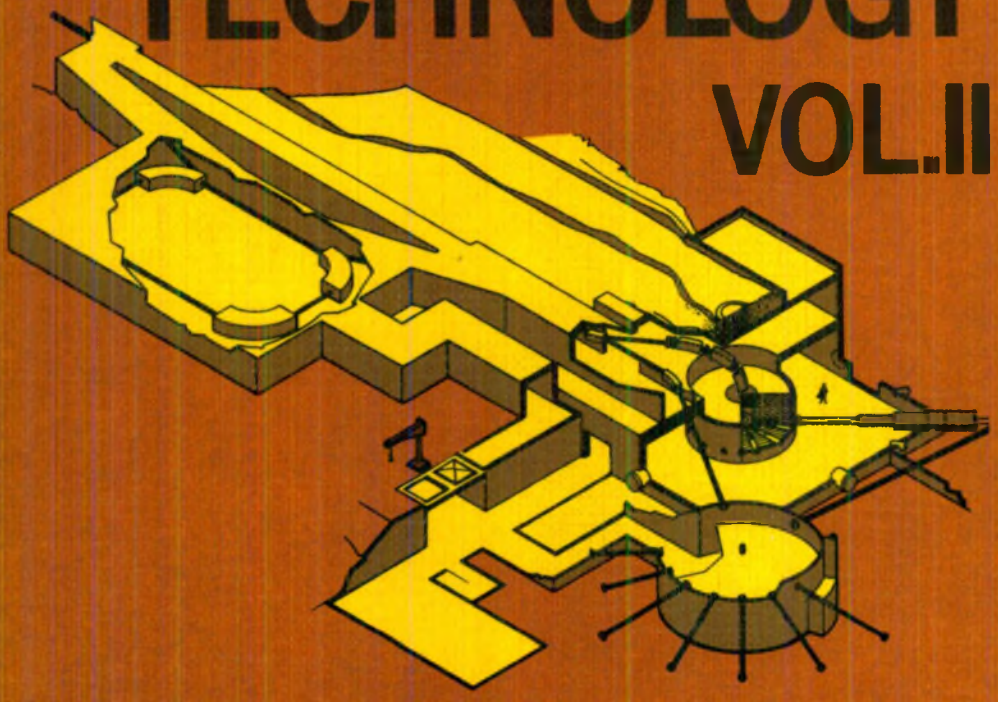
NUCLEAR DATA IN SCIENCE AND TECHNOLOGY VOL. II



PROCEEDINGS OF A SYMPOSIUM, PARIS, 12-16 MARCH 1973

INTERNATIONAL ATOMIC ENERGY AGENCY, VIENNA, 1973

NUCLEAR DATA
IN SCIENCE AND TECHNOLOGY

VOL. II

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VOL. II

INTERNATIONAL ATOMIC ENERGY AGENCY
VIENNA, 1973

NUCLEAR DATA IN SCIENCE AND TECHNOLOGY
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FOREWORD

The IAEA Symposium on "Applications of Nuclear Data in Science and Technology" was convened by the International Atomic Energy Agency on 12 - 16 March 1973 in Paris at the invitation of the French Government. The meeting was held on the recommendation of the International Nuclear Data Committee (INDC) and the International Working Group on Nuclear Structure and Reaction Data (IWGNSRD). The main purpose of the Symposium was to illuminate the needs for nuclear data in the technological and scientific community. Over 200 delegates attended, representing 30 countries and five international organizations. A total of 74 papers was presented, including the Keynote Address and the Symposium Summary.

For many years the mechanisms for satisfying the nuclear data needs related to neutron-induced reactions have been fairly well organized by those concerned with neutron-reactor technology, which is a major field of application for this kind of data. However, for several years it has become increasingly evident that there is a strong need for better, up-to-date compilations of nuclear data for a large number of other applications. The IAEA was therefore requested to convene a symposium in order to review the status of and needs for new nuclear data evaluation activities. During the preparation, it became evident that the symposium should emphasize data needs in the various applications rather than existing data compilation activities.

The program committee attempted to achieve a balance between reactor and non-reactor applications as well as a balance between the needs for various applications and the needs for compilation work. As a result, four of the sixteen regular sessions were devoted to applications related to nuclear energy, seven to other applications and five to topics related to data compilations. In contrast to the International Conferences on Nuclear Data for Reactors (Paris, 17 - 21 October 1966, and Helsinki, 15 - 19 June 1970), this Symposium was not meant to be a forum for the presentation of experimental data.

The Symposium demonstrated that the compilation of structure and decay data would benefit from considerably increased support. These data are basic to most other data-application-oriented compilations. The long delay in bringing compilations of this type up to date causes unacceptable hold-ups in the process of bringing such data from producers to users. Under the heading 'Symposium Summary', Dr. W.B. Lewis, at the end of the meeting, discussed the most important conclusions to be drawn from the symposium with regard to data needs and the status of compilations. A further summary of the meeting, by L. Hjärne, appears in Atomic Energy Review, 1973, Vol.11, No.2, 395.

The Proceedings are divided into two volumes, the first of which contains, besides the Keynote Address, thirty-two papers in the fields of Future Technology Requirements, Reactor Technology, Safeguards, Life Sciences, Radioisotopes in Chemistry, Fission-Product Nuclear Data and

Accelerator and Space Shielding. The second volume – with forty contributions, in addition to the Symposium Summary – covers the fields of Fusion Research, Evaluated Neutron Data Files, Activation Analysis (General and Neutrons), Compilation and Evaluation – Data Centres, Large-Volume Compilations, Various Applications, Activation Analysis: Charged Particles and Photons, and Application-Oriented Computations and Evaluations.

The Agency wishes to thank the French authorities for their hospitality and active support of the Symposium, and the authors and participants for their valuable contributions. Special thanks are due to the Chairmen of the individual sessions for their successful efforts in guiding the discussions.

EDITORIAL NOTE

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For the sake of speed of publication the present Proceedings have been printed by composition typing and photo-offset lithography. Within the limitations imposed by this method, every effort has been made to maintain a high editorial standard; in particular, the units and symbols employed are to the fullest practicable extent those standardized or recommended by the competent international scientific bodies.

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FUSION RESEARCH

Chairman

Yu.F. CHERNILIN (IAEA)

НЕКОТОРЫЕ ПРОБЛЕМЫ ИЗМЕРЕНИЯ ЯДЕРНЫХ КОНСТАНТ ДЛЯ ТЕРМОЯДЕРНЫХ РЕАКТОРОВ

Е.А.КУЗЬМИН, А.А.ОГЛОБЛИН,
Н.И.СИДОРОВ, А.Р.ФАЙЗИЕВ,
Л.В.ЧУЛКОВ, Г.Б.ЯНЬКОВ
Институт атомной энергии им.И.В.Курчатова,
Москва,
Союз Советских Социалистических Республик

Abstract–Аннотация

PROBLEMS OF MEASURING NUCLEAR CONSTANTS FOR THERMONUCLEAR REACTORS.

From the available nuclear data it appears that a tritium breeding coefficient greater than unity can be obtained in a thermonuclear reactor using the D-T cycle and with a breeding zone consisting of a mixture of lithium isotopes. However, the present inaccuracies in the nuclear data may considerably influence tritium breeding and the optimum relationship between breeder and construction materials. A fairly extensive programme of neutron research in the neutron energy range 7-14 MeV is therefore necessary. The main feature of neutron measurements within this range is that there are no monoenergetic neutron sources available. The authors consider the possibility of using normal sources – the reactions D + D, D + T, T + p. Measurement data are presented on the continuous neutron spectrum in the reaction T + p, which limits the use of this reaction as a source of monoenergetic neutrons. Neutron sources based on the use of alpha particles and ^{3}He ions are considered and currently available sources of continuous-spectrum neutrons are discussed. It is shown that the optimum source of a continuous spectrum of neutrons in the required energy range is deuteron disintegration at $E_d \simeq 25$-40 MeV. The authors consider which nuclear constants can be used in continuous neutron spectrum measurements.

НЕКОТОРЫЕ ПРОБЛЕМЫ ИЗМЕРЕНИЯ ЯДЕРНЫХ КОНСТАНТ ДЛЯ ТЕРМОЯДЕРНЫХ РЕАКТОРОВ.

Из существующих в настоящее время ядерных данных следует, что в термоядерном реакторе, использующем D-T цикл и зону воспроизводства из смеси изотопов лития, может быть получен коэффициент воспроизводства трития больше единицы. Однако, имеющиеся неточности в ядерных данных могут существенно повлиять на воспроизводство трития и на оптимальное соотношение между воспроизводящими и конструкционными материалами. Это делает необходимым проведение довольно большой программы нейтронных исследований в диапазоне энергий нейтронов 7-14 МэВ. Основная особенность нейтронных измерений в этой области состоит в том, что для нее отсутствуют моноэнергетические источники нейтронов. Рассматривается возможность использования обычных источников – реакций D+D, D + T, T + p, ограничивающего использование этой реакции как источника моноэнергетических нейтронов. Рассматриваются источники нейтронов с использованием α-частиц и ионов ^{3}He. Анализируются современные источники нейтронов сплошного спектра. Показано, что оптимальным источником сплошного спектра нейтронов в требуемом диапазоне энергий является процесс развала дейтрона при $E_d \simeq 25$-40 МэВ. Рассматривается вопрос о том, какие ядерные константы могут быть получены в измерениях на нейтронном сплошном спектре.

1. ВВЕДЕНИЕ

Термоядерные реакторы будущего требуют знания ядерных констант для решения двух основных задач: определения характеристик горючего в плазме и рационального использования образующегося в реакциях синтеза проникающего излучения. Хотя параметры плазмы, необходимые для протекания термоядерных реакций еще не получены и путь получения

энергии благодаря реакции синтеза не выбран, уже ведется широкое обсуждение различных моделей термоядерного реактора [1].

Анализ современного состояния данных о протекающих в плазме реакциях между легкими ядрами показывает [2], что достаточно полно известны характеристики только для процессов d + d, d + t и d + ^{3}He.

За редким исключением, как например реакция ^{3}He + d, все реакции синтеза, по крайней мере в одной из ветвей, приводят к образованию быстрых нейтронов. В настоящее время считается, что наиболее достижимым является плазменный цикл, основанный на горении равнокомпонентной смеси дейтерия и трития. В этой первичной реакции образуются нейтроны с энергией около 14 МэВ. Однако из-за вторичных процессов в плазме могут присутствовать нейтроны и больших энергий. В плазме и в материалах, окружающих плазму, нейтроны теряют свою энергию в результате упругих и неупругих соударений, создают значительные потоки γ-излучения, воспроизводят использованную в реакциях синтеза компоненту горючего, отсутствующего в природе, создают другие изотопы и т.п. Характеристики указанных процессов, такие как угловые распределения упруго-рассеянных нейтронов, угловые и энергетические распределения вторичных нейтронов для процессов неупругого рассеяния и реакции (n, 2n), сечения образования γ-лучей и их энергетические спектры, сечения реакций воспроизводства и конкурирующих процессов, сечения образования различных изотопов и другие – для широкого круга элементов и изотопов необходимо знать для рационального использования нейтронов в термоядерных реакторах.

Потребности в ядерных данных для начальной стадии изучения термоядерных реакторов, подробно обсуждались в работах [2-4]. Хотя определенная часть этих потребностей является общей с потребностями реакторов деления, значительно более широкая область энергий нейтронов, используемых в термоядерных реакторах, выдвигает существенно новые задачи измерения ядерных констант. Данных о взаимодействии нейтронов с различными ядрами особенно мало для области энергий нейтронов 7-14 МэВ. В то же время измерения на нейтронных пучках в этой области являются наиболее сложными. Особенность измерений состоит в том, что почти для всех значений энергий нейтронов здесь отсутствуют, в обычном понимании, пучки моноэнергетических нейтронов, которые широко используются для измерений ядерных данных, в первую очередь для изучения угловых и энергетических распределений вторичного нейтронного излучения.

Обсуждению особенностей измерения ядерных констант, главным образом в области энергий нейтронов 7-14 МэВ, посвящена данная работа.

2. ИСТОЧНИКИ МОНОЭНЕРГЕТИЧЕСКИХ НЕЙТРОНОВ*

В качестве моноэнергетических источников нейтронов в различных частях энергетической области 7-14 МэВ обычно используются реакции, протекающие между изотопами водорода: T(p,n)^{3}He (Q= - 0,764 МэВ), D(d,n)^{3}He (Q= 3,27 МэВ) и T(d,n)^{4}He (Q= 17,59 МэВ). На рис.1-3 представлены дифференциальные сечения образования нейтронов на

* Рассматриваемые ниже реакции называются моноэнергетическими источниками условно.

изотопах водорода под углом 0° в лабораторной системе координат в зависимости от энергии протонов и дейтронов [5]. Подробные сведения об этих реакциях, включая табличные данные о зависимости энергии вылетающих нейтронов от энергии падающих частиц и угла вылета нейтрона, приводятся в работах [6,7]. Ядра ^{3}He и ^{4}He не имеют низколежащих уровней возбуждения, поэтому вылетающие нейтроны обладают строгой моноэнергетичностью. Однако, начиная с определенных энергий заряженных частиц, в пучке моноэнергетических нейтронов появляются нейтроны меньших энергий, вследствие процессов развала сначала на три (а затем и на большее число) частицы согласно реакциям T(p,pn)D,

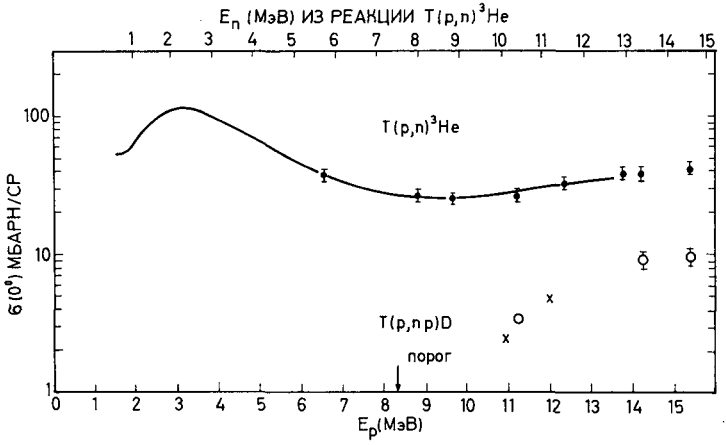

Рис.1. Сечения образования нейтронов под 0° при взаимодействии протонов с тритием.
Сплошная кривая - сечение реакции T(p, n) ^{3}He [5].
• - сечение реакции T(p,n) ^{3}He - данные ИАЭ.
x - верхний предел сечения развала трития [10].
o - сечение развала трития - данные ИАЭ.

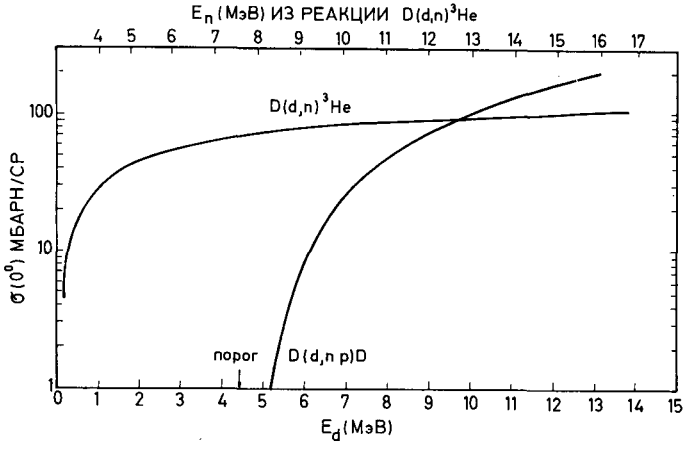

Рис.2. Сечения образования нейтронов под 0° при взаимодействии дейтонов с дейтерием [5].

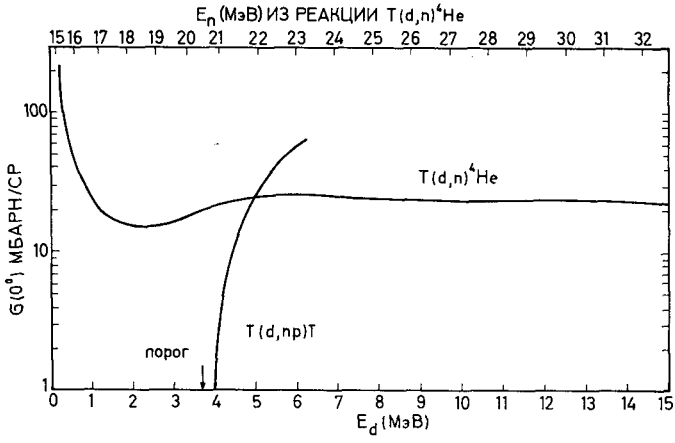

Рис.3. Сечения образования нейтронов под 0° при взаимодействии дейтонов с тритием [5].

D(d,pn)D, T(d,pn)T. Нейтроны развала преимущественно летят в направлении движения ускоряемых частиц. При планировании экспериментов, а также при обработке полученных данных необходимы сведения о сечениях процессов развала, об угловых распределениях и энергетических спектрах вылетающих нейтронов.

2.1. Нейтроны из реакций протонов с тритием

Дифференциальные сечения реакции T(p,n)^{3}He в настоящее время известны в области энергий протонов от порогового значения (E_{thr} = 1,019 МэВ) примерно до 13,5 МэВ с точностью около 15%. Эффективное сечение под углом 0° измерено с погрешностью 5-7% [8,9]. При энергии протонов 8,36 МэВ достигается порог реакции развала трития T(p,pn)D(Q= - 6,26 МэВ) и появляется группа нейтронов с непрерывным энергетическим распределением. При энергии протонов выше 11,34 МэВ в пучке возможно присутствие нейтронов из реакции T(p,p 2n)H. До последнего времени литературные данные о величине сечения процесса развала трития на водороде отсутствовали. В работах [10,11] приводилась лишь оценка верхнего предела сечения при энергиях протонов 11-12 МэВ.

С целью изучения характеристик пучка нейтронов, возникающего при взаимодействии протонов с тритием, во всей области, представляющей интерес для термоядерных реакторов, на циклотроне в ИАЭ им.И.В.Курчатова была проведена в 1972 году соответствующая работа. Определялись дифференциальные сечения под углом 0° реакции T(p,n)^{3}He в интервале энергий протонов 6-15,3 МэВ, сечения процесса образования нейтронов развала и энергетические спектры этих нейтронов также под углом 0° для энергий протонов 11,2 МэВ, 14,2 МэВ и 15,3 МэВ. Относительный ход сечения измерялся методом времени пролета, порог регистрации нейтронов составлял 1,5 МэВ. Абсолютные значения сечения реакции T(p,n)^{3}He определялись нормировкой по данным работы [9]. На рис.1 представлены результаты измерений для T(p,n)^{3}He и для про-

цесса развала трития. Кружками с экспериментальными ошибками (6% для $T(p,n)^3He$) указаны данные ИАЭ, крестами – данные [10], кривая – данные [5]. Верхний предел сечения развала трития при энергии протонов 11,2 МэВ оценивается равным 3,5 мбарн/ср, при энергиях протонов 14,2 МэВ и 15,3 МэВ сечения соответственно равны 10,1 ± 1,6 мбарн/ср и 10,0 ± 1,4 мбарн/ср. Энергетические спектры нейтронов развала трития на водороде под углом 0° (для нейтронов с энергией выше 2 МэВ) показаны на рис.4. Разброс экспериментальных значений сечений во всех областях усреднения шириной 0,5 МэВ, за исключением высокоэнергетичной граничной области, не превышал 20%. На основании полученных данных можно сказать, что при энергиях протонов выше примерно 11 МэВ в использовании моноэнергетического пучка нейтронов из реакции $T(p,n)^3He$ возникают дополнительные трудности, вследствие присутствия в пучке также и нейтронов развала трития, доля которых составляет около 20%.

Определенным выходом из этого положения является использование той же реакции $H(t,n)^3He$, но при ускорении не протонов, а ядер трития [11]. Хотя ускорение ядер трития сопряжено с известными техническими трудностями, использование указанной реакции в качестве источника нейтронов имеет свои преимущества: значительно возрастает выход нейтронов под углом 0°; порог реакции $H(t,np)D$ достигается здесь при энергии трития E_{thr} = 25,03 МэВ, поэтому нейтроны из реакции

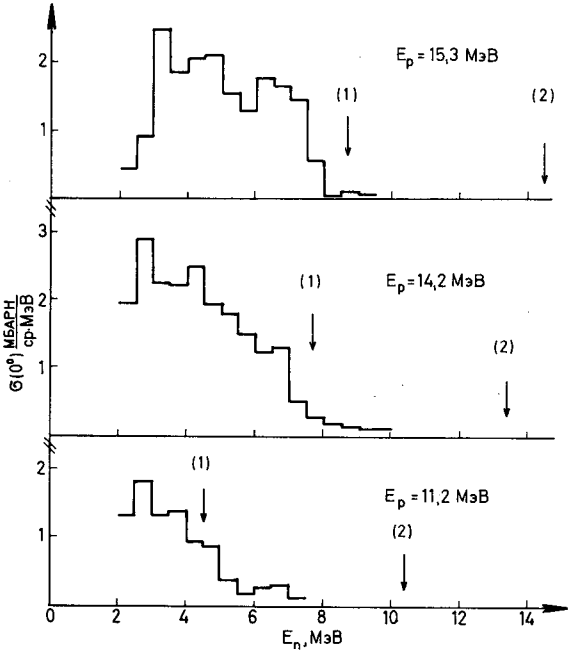

Рис.4. Энергетические спектры нейтронов из реакции развала трития при взаимодействии трития с протонами с энергией 11,2, 14,2 и 15,3 МэВ. Стрелками (1) и (2) обозначены, соответственно, расчетная E_{max} нейтронов из реакции развала трития и E_n нейтронов из реакции $T(p,n)^3He$. Присутствие нейтронов правее стрелки (1) вызвано ошибками в учете фона.

$H(t,n)^3He$ вплоть до энергии 17,5 МэВ свободны от присутствия развальной группы. Пороговое значение энергии ядер трития в реакции $H(t,n)^3He$ равно E_{thr} = 3,06 МэВ, при этом нейтроны под углом 0° имеют энергию 0,575 МэВ. Выше порога нейтроны заключены в конусе со значением половинного угла при вершине, определяемым из соотношения $\sin\theta = (E_t - E_{thr})^{1/2} E^{-1/2}$. При возрастании E_t значение θ стремится к пределу, равному 90°. За счет движения центра масс под каждым углом в лабораторной системе координат испускаются нейтроны двух различных энергий. На рис.5 представлены сечение образования нейтронов в направлении 0° и энергии соответствующих нейтронных групп. Нейтроны низкоэнергетической группы имеют весьма малую энергию и интенсивность, поэтому их присутствие в пучке для ряда экспериментов может быть несущественным. Выход под углом 0° нейтронов высокой энергии оказывается гораздо больше выхода в реакциях $T(p,n)^3He$ и $D(d,n)^3He$. Так, например, при E_t = 15 МэВ энергия нейтронов высокоэнергетичной группы составляет 10 МэВ, а выход нейтронов 450 мбарн/ср; энергия нейтронов низкоэнергетической группы составляет при этом около 0,03 МэВ. Реакции $T(p,n)^3He$ и $D(d,n)^3He$ для нейтронов с энергией 10 МэВ имеют выход 28 мбарн/ср и 83 мбарн/ср, соответственно.

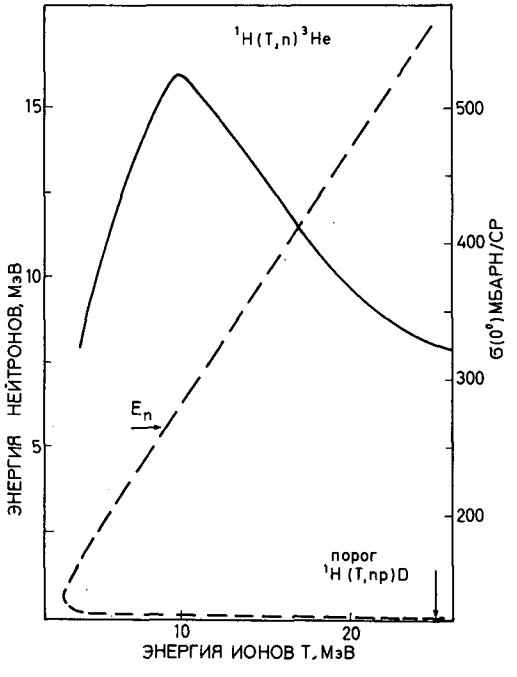

Рис.5. Энергия групп нейтронов из реакции $H(t,n)^3He$ под 0° (пунктирная кривая). Сечение образования высокоэнергитичной группы нейтронов из реакции $H(t,n)^3He$ под 0° (сплошная кривая).

2.2. Нейтроны из реакций взаимодействия дейтронов

Реакция $D(d,n)^3He$ широко используется как источник моноэнергетических нейтронов с энергией примерно до 7,5 МэВ. Характеристики реакции подробно изучены вплоть до энергий дейтронов 19 МэВ. Дифференциальные сечения измерены с точностью около 2,5% в области энергий дейтонов 2-6 МэВ [12] и с погрешностью 4-5% при больших энергиях [13-14]. При энергии дейтонов 4,45 МэВ достигается порог реакции развала дейтона $D(d,np)D$ ($Q = -2,23$ МэВ) и в пучке появляется группа нейтронов с непрерывным энергетическим распределением. Энергетическая зависимость сечения реакции $D(d,np)D$ под углом 0° представлена на рис.2. Выход нейтронов развала быстро растет с увеличением энергии дейтонов и при значении энергии около 9,5 МэВ становится больше выхода моноэнергетической группы. Энергетические спектры и угловые распределения нейтронов из реакции $D(d,np)D$ изучались методом времени пролета при энергиях дейтонов 7,5-11 МэВ [15]. На рис.6 показаны спектры нейтронов развала дейтонов под углом 0° в лабораторной системе координат. Погрешность определения величины сечения составляла около 20%. С увеличением угла вылета спектры нейтронов становятся мягче. Наиболее энергичные нейтроны сосредоточены в малых углах. На рис.7 показано изменение спектрального состава нейтронов в зависимости от угла вылета в лабораторной системе координат при энергии дейтонов около 10 МэВ. Спектры имеют гладкую колоколообразную форму, аналогичную представленной на рис.6, и при более высоких энергиях дейтонов 12-19 МэВ [14].

Известны попытки отделить пучок нейтронов из реакции $D(d,n)^3He$ от нейтронов развала. В работе [16] исследовались возможности регистрации совпадений импульсов от нейтронов с импульсами от сопутствующих нейтронам ядер ^3He. При этом возникает ряд серьезных трудностей. При энергиях дейтонов больше 3,27 МэВ ядра ^3He вылетают в лабораторной системе координат только в переднее полупространство,

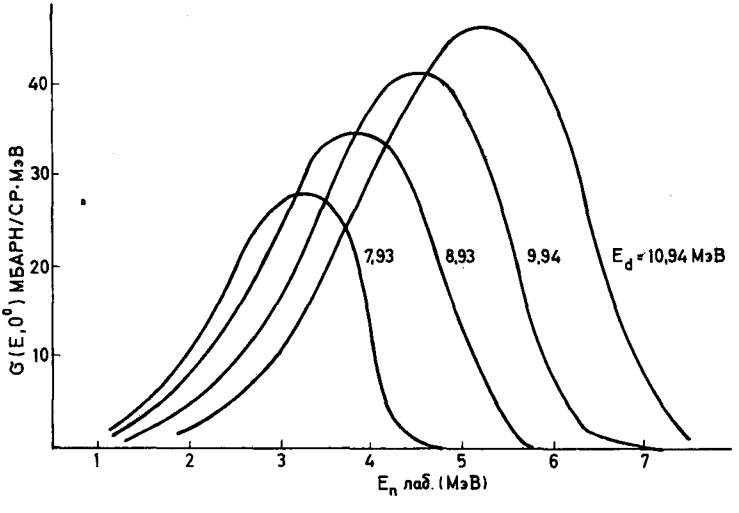

Рис.6. Спектры нейтронов под 0° из реакции D(d,np)D [15].

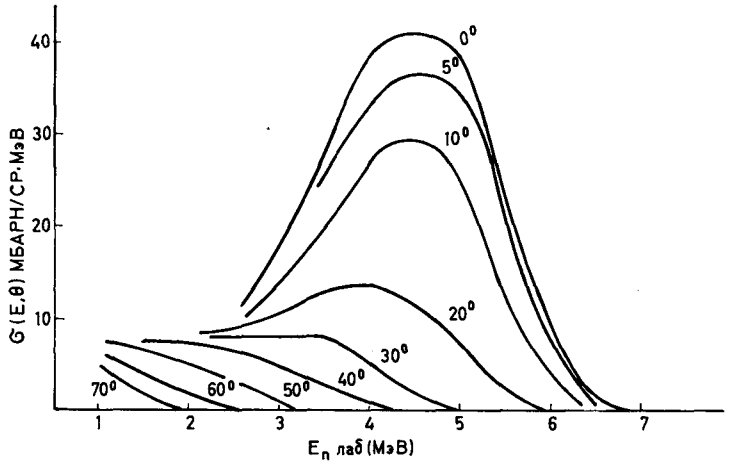

Рис.7. Спектры нейтронов под разными углами из реакции D(d,np)D. Для $\theta \leqslant 30°$ E_d = = 9,94 МэВ. Для $\theta > 30°$ E_d = 9,75 МэВ [15].

причем направления вылета ограничены конусом с величиной половинного угла при вершине, определяемого соотношением $\sin^2\theta = \frac{1}{3}(1 + 6{,}54/E_d)$, где E_d есть энергия дейтонов в МэВ. Под каждым углом будут наблюдаться две группы частиц ^{3}He, обладающих различными энергиями. Ядра ^{3}He,имеющие меньшие значения энергии, соответствуют нейтронам с большим значением энергий, вылетающим также в переднее полупространство. По абсолютным значениям энергия ядер ^{3}He равна примерно 1-2 МэВ. Регистрацию ядер ^{3}He приходится вести в условиях огромного фона от рассеянных дейтонов. Используя схему совпадений с разрешающим временем 3,5 нсек и мишень из дейтерированного полиэтилена толщиной около 25 мк, автору работы [16] удалось получить узкие пучки моноэнергетических нейтронов с энергией 2-12 МэВ и интенсивностью, равной 2,2·10 нейтр/мкКл·мср. Таким образом, в настоящее время получены обнадеживающие результаты по исследованию возможностей выделения моноэнергетической части пучка, соответствующей реакции D(d,n)^{3}He, однако на этом пути предстоит еще многое сделать, чтобы метод ассоциированных частиц вошел в практику.

2.3. Реакция T(d,n)^{4}He

Основная термоядерная реакция T(d,n)^{4}He широко используется и как источник моноэнергетических нейтронов с энергией около 14 МэВ и выше. Резонанс в кривой сечения (σ_{max} = 5 барн) при энергии дейтонов около 110 кэВ обуславливает важность этой реакции как источника нейтронов, главным образом, при работе с низковольтными ускорителями. При этом относительно просто осуществляется регистрация сопутствующих нейтронам α-частиц с энергией около 3,5 МэВ. Здесь этим методом широко пользуются как для мониторирования, так и для абсолютного определения потока нейтронов. На рис.3 представлены зависимости сече-

ния под углом 0° реакций $T(d,n)^4He$ и $T(d,np)T$. Однако процесс развала дейтона на ядре трития становится заметным при тех значениях энергий нейтронов (E_n = 20 МэВ), которые выходят за пределы основных интересов данной работы. Отметим, что для получения моноэнергетических нейтронов с энергией в интервале значений 12-14 МэВ можно использовать нейтроны, вылетающие в заднее полупространство из реакции $T(d,n)^4He$ при энергиях дейтонов в несколько МэВ. Однако использование нейтронных пучков, идущих в заднее полупространство, для применения весьма неудобно и, кроме того, сечение реакции в этой области энергий дейтонов невелико. Для основной массы измерений ядерных констант для термоядерных реакторов в точках с энергией около 14 МэВ использование реакции $T(d,n)^4He$ особых трудностей не встречает. Однако, например, для осуществления полного опыта при изучении реакций (n,2n), когда экспериментально определяются энергетические и угловые характеристики обоих нейтронов, выход нейтронов из обычных устройств оказывается недостаточным и даже в случае больших значений сечений реакции (n,2n) длительность экспозиции при одном угловом положении занимает несколько суток [17]. В этом отношении весьма интересны исследования [18], проводимые с целью создания мощных источников нейтронов для применения при изучении радиационных повреждений и терапии рака.

2.4. Реакция взаимодействия лития с водородом

Реакция $^7Li(p,n)^7Be$ (Q = - 1,64 МэВ) является одним из наиболее изученных и широко применяемых источников нейтронов при энергиях протонов 1,9-2,4 МэВ. Особенностью реакции является наличие низколежащего уровня возбуждения ядра ^7Be (E^* = 0,43 МэВ); поэтому при энергиях протонов выше 2,4 МэВ в пучке нейтронов появляется дополнительная линия, соответствующая этому возбужденному состоянию ядра ^7Be. Измерения показывают [7,19], что при энергиях протонов ниже 6 МэВ интенсивность нейтронов второй группы составляет не более 15% от интенсивности основной группы. С ростом энергии протонов сечение реакции с образованием основного состояния ядра ^7Be быстро уменьшается и поэтому интенсивности нейтронных групп становятся сравнимыми [20,21]. Кроме того, при энергиях протонов выше 7 МэВ определенный вклад дает и второе возбужденное состояние ^7Be (Q = - 6,29 МэВ). Например, при энергии протонов 14 МэВ выход нейтронов этой группы под углом 0° в системе центра масс только примерно в 2,5 раза меньше суммарного выхода нейтронов с основного и первого возбужденного состояний бериллия [21]. Все это ограничивает применимость этой реакции областью энергий нейтронов, лежащей вне рассматриваемого интервала.

Расчет, аналогичный расчету характеристик реакции $^1H(t,n)^3He$, был проведен авторами данной работы для реакции $^1H(^7Li,n)^7Be$. Область энергий нейтронов от 7 до 14 МэВ требует ускорения ионов лития в интервале энергий примерно 23-40 МэВ. Образующиеся в реакции нейтроны заключены в конусе с половинным углом при вершине, определенным соотношением $\sin^2 Q = 1 - \frac{13,2}{E_{Li}}$, где E_{Li} есть энергия ионов ^7Li в МэВ. На рис.8 представлены эффективные сечения реак-

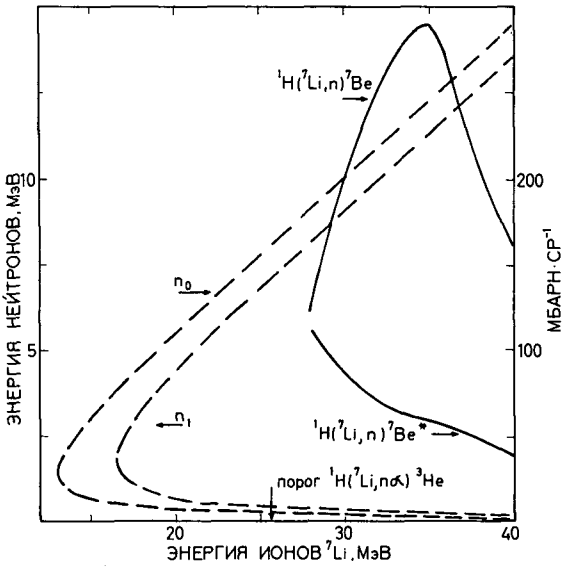

Рис.8. Энергия групп нейтронов из реакции $^1H(^7Li,n)^7Be$ и $^1H(^7Li,n)^7Be^*$ под 0° (пунктирные кривые). Сечение реакций $^1H(^7Li,n)^7Be$ и $^1H(^7Li,n)^7Be^*$ под 0° (сплошные кривые).

ций $^1H(^7Li,n)^7Be$ и $^1H(^7Li,n)^7Be^*$ (0,43 МэВ) для угла 0°, рассчитанные по данным работы [19]. На том же рисунке изображены значения энергий нейтронов, соответствующих ядру ^7Be в основном и первом возбужденном состояниях при угле вылета нейтронов 0°. Значения энергии нейтронов для указанных групп отличаются примерно на 1 МэВ; в районе энергий нейтронов 12 МэВ интенсивности указанных групп относятся примерно как 5 : 1. Такая немоноэнергетичность пучка нейтронов может оказаться вполне приемлемой для некоторых задач определения ядерных констант в области энергий нейтронов больше 10 МэВ. Концентрация потока нейтронов в пределах сравнительно узкого конуса углов и слабая зависимость энергии нейтронов от энергии ионов лития, допускающая работу с относительно толстыми мишенями, может обеспечить сравнительно мощные потоки быстрых нейтронов с интенсивностью около $4 \cdot 10^9$ нейтр/ср при токе однозарядных ионов лития 1 мкА. Второе возбужденное состояние ^7Be достигается здесь при энергии ионов лития около 50 МэВ, что лежит за пределами рассматриваемого интервала. В пучке нейтронов будут присутствовать также нейтроны, вылетающие в системе центра масс в заднее полупространство. Энергия этих нейтронов, как видно из рис.8, не превышает нескольких сотен кэВ, а сечения их образования малы. При энергии ионов лития больше 25,8 МэВ энергетически возможна реакция $^1H(^7Li,n,\alpha)^3He$, нейтроны из которой будут иметь сплошной энергетический спектр.

2.5. Реакции с ионами гелия

Реакции $^{12}C(^3He,n)^{14}O$, $^{16}O(^3He,n)^{18}Ne$, $^{12}C(^4He,n)^{15}O$, $^{13}C(^4He,n)^{16}O$ дают моноэнергетические нейтроны в ограниченных

интервалах энергий, которые находятся вне области, представляющей интерес. Ниже указываются максимальные значения энергии моноэнергетических нейтронов под 0° при энергиях ионов гелия, равных порогу реакции на первое возбужденное состояние конечного ядра:
$^{12}C(^{3}He,n)^{14}O$ – 6,4 МэВ, $^{16}O(^{3}He,n)^{18}Ne$ – 2,4 МэВ,
$^{12}C(^{4}He,n)^{15}O$ – 7,5 МэВ, $^{13}C(^{4}He,n)^{16}O$ – 7,0 МэВ.

3. ИСТОЧНИКИ НЕЙТРОНОВ СПЛОШНОГО СПЕКТРА

Пучки быстрых нейтронов сплошного спектра в большинстве случаев создаются с помощью ускорения дейтонов, бомбардирующих толстые мишени из различных элементов. В последнее время для этих целей стали использоваться линейные ускорители электронов [22] с мишенями специальных конструкций из набора определенных элементов. Однако выход нейтронов с энергиями в области 7 ÷ 14 МэВ из фото-нейтронных реакций значительно меньше выхода из реакции (d,n). Возникающие в реакциях (d,n) нейтроны обусловлены, главным образом, двумя процессами: испарением из сильно возбужденного ядра и развалом дейтона в поле ядерных и кулоновских сил ядра мишени. Энергетический спектр нейтронов в первом случае является испарительным, а угловое распределение симметрично относительно 90° и близко к изотропному. Нейтроны развала характеризуются широким колоколообразным энергетическим спектром с максимальной интенсивностью при энергиях нейтронов равных примерно половине энергии дейтонов. Нейтроны развала направлены преимущественно по движению дейтона. Энергетическое и угловое распределение этих нейтронов удовлетворительно описывается выражениями [23]:

$$n(E)\,dE = (1 + y^2)^{-3/2}\,dy, \quad y = (E - 1/2\,E_d)/\sqrt{E_d B},$$
$$n(\theta)\,d\theta = (1 + x^2)^{-3/2}\,dx, \quad x = \theta\sqrt{E_d/B}.$$

Здесь E и E_d – энергии нейтрона и дейтона, соответственно, B – энергия связи дейтона (2,23 МэВ) и θ – угол вылета нейтрона.

Спектры из мишеней различных элементов близки по форме и плавно меняются с изменением энергии дейтонов. Общей особенностью спектров является также уменьшение энергии нейтронов с ростом заряда ядра-мишени [24]. Полный выход нейтронов из толстых мишеней быстро растет с увеличением энергии дейтона и падает с ростом заряда ядра-мишени. Наибольший выход дают толстые мишени из дейтерия, трития и бериллия. На рис.9 представлены эффективные сечения процессов d-^{2}H, d-^{3}H и d-Be, приводящих к рождению нейтронов в направлении 0° в области энергий дейтонов до 20 МэВ [18]. Хотя сечения процессов d - Be превышают соответствующие сечения на тяжелых изотопах водорода, расчетные значения выхода нейтронов из толстых газообразных дейтериевых и тритиевых мишеней при энергиях дейтонов 10-16 МэВ оказываются большими, чем значения выхода из бериллия [25]. Спектры быстрых нейтронов в процессах d-^{2}H обладают весьма привлекательным для нейтронных измерений качеством: резким обрывом распределения со стороны высоких энергий. На рис.10 представлены расчетные спектры нейтронов из толстой газообразной дейтериевой мишени для дейтонов с энергиями 8, 10, 11,75 и 16 МэВ [25]. Такая форма спектра ведет при измерениях в ограниченном энергетическом интервале к уменьшению фона из-за отсутствия неиспользующихся нейтронов с большими энер-

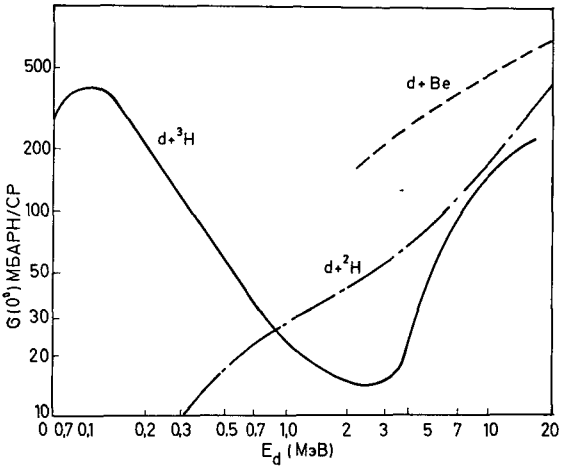

Рис.9. Сечения процессов образования нейтронов под 0° из реакций d + ^{3}H, d + ^{2}H, d + Be [18].

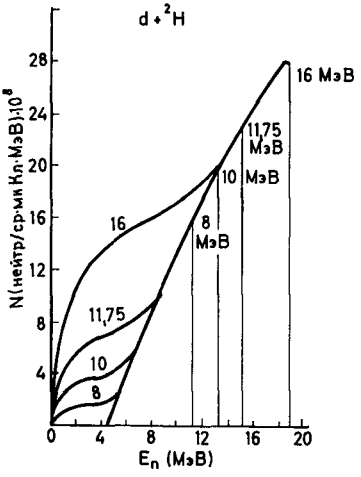

Рис.10. Расчетные нейтронные спектры под 0° при падении моноэнергетических дейтонов с энергиями 8, 10, 11,75 и 16 МэВ на толстую дейтериевую мишень [25].

гиями, облегчает сооружение защиты и коллимацию нейтронного пучка. Однако из-за большой величины пробега дейтона в дейтерии (200 мг/см^2 при E_d = 12 МэВ, что соответствует пробегу около 11 м при нормальных условиях) практическое использование толстых газообразных или жидкостных мишеней сопряжено с большими техническими затруднениями.

Большие возможности представляет использование толстой бериллиевой мишени. На рис.11 показано изменение потока нейтронов в направлении 0° и в интервале энергий дейтонов от 8 до 30 МэВ по дан-

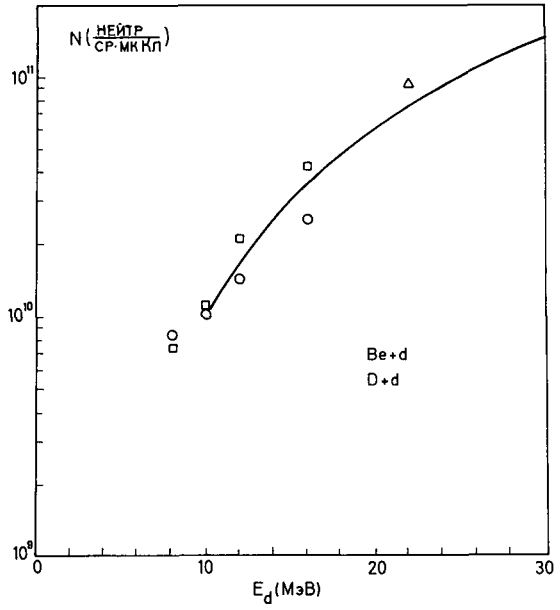

Рис.11. Зависимость выхода нейтронов из толстых Be и D мишеней под 0° от энергии дейтонов.

Сплошная кривая - [18].
○ - [25].
△ - [26] - Be-мишень.
□ - [25] - D-мишень.

ным ряда авторов [18,25,26]. На том же рисунке для сравнения приведены значения выхода нейтронов из дейтериевой мишени [25]. Как видно из этого рисунка выход нейтронов из бериллиевой мишени при энергии дейтонов около 28 МэВ составляет величину большую 10^{11} нейтр/мкКл·ср. Детальное сравнение нейтронов высокой энергии из толстых бериллиевых мишеней при энергиях дейтонов 15, 20, 24, 40 и 53,8 МэВ показывает, что энергетические спектры обладают геометрическим подобием и могут характеризоваться положением максимума интенсивности и полушириной распределения $\Delta E_{1/2}$ [27]. На основании такого подобия авторы работы предлагают практический метод конструирования энергетических распределений нейтронов для энергий дейтонов 15-50 МэВ. На рис.12 показан рассчитанный таким образом спектр при энергии дейтонов 28,4 МэВ. Энергетические распределения нейтронов, в первом приближении, остаются неизменными для всех углов вылета. Однако при больших углах спектры оказываются несколько обедненными нейтронами высоких энергий.

Значительные возможности для измерения ядерных констант на сплошных спектрах открываются при использовании изохронных циклотронов [28], в которых наряду с малой длительностью импульса тока на мишень, удается сохранить сравнительно большой средний ток. Источником нейтронов здесь является мишень, находящаяся внутри камеры циклотрона, на которую за время 1-2 нсек путем отклонения попадают

дейтоны, находящиеся в процессе ускорения на нескольких десятках орбит. На рис.13 приведен спектр нейтронов, полученный на толстой урановой мишени, при попадании на нее ионов дейтерия с энергиями в интервале 40 ÷ 50 МэВ. Поток нейтронов в направлении 0° составлял $5 \cdot 10^{11}$ нейтр/мкКл·ср.

Необходимо отметить, что для получения интенсивных потоков нейтронов в диапазоне энергий 7-15 МэВ энергия дейтонов может не превосходить значения 30-35 МэВ. В ИАЭ им.И.В.Курчатова разработана подобная система [29], которая может быть использована для двухдуантного изохронного циклотрона с максимальной энергией дейтонов 30 МэВ.

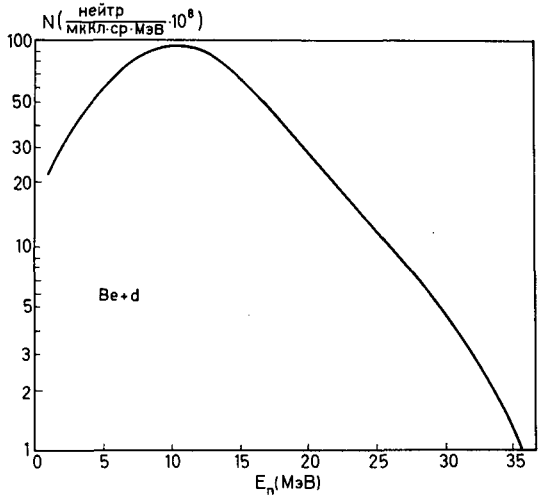

Рис.12. Расчетный спектр нейтронов из толстой Be мишени при энергии дейтонов 28,4 МэВ [27].

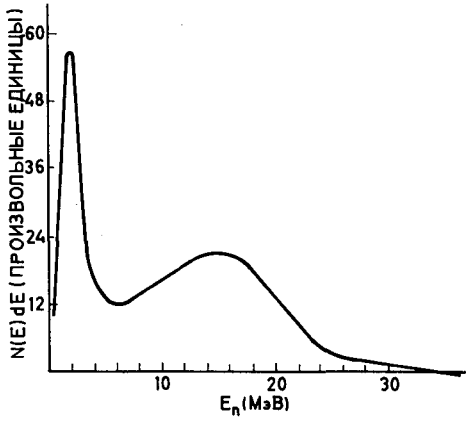

Рис.13. Спектр нейтронов из толстой урановой мишени, бомбардируемой дейтонами с энергией 40 - 50 МэВ [28].

Была получена частота повторения импульсов 110 кГц и оценки показывают, что при длительности импульсов 1,5 нсек поток нейтронов от бериллиевой мишени в направлении 0° будет составлять величину около $3 \cdot 10^{12}$ нейтр/ср·сек.

Преимущества использования сплошных спектров – "белого" пучка – наиболее четко проявляются при систематическом изучении энергетических зависимостей сечений в широкой области энергий нейтронов. Использование в таких измерениях моноэнергетических источников нейтронов требует очень больших затрат времени. Достигнутое на "белом" пучке от изохронного циклотрона энергетическое разрешение в широкой области энергии нейтронов (0,5-30 МэВ) является рекордным и составляет $\leqslant$ 1,7 кэВ при 0,8 МэВ, $\leqslant$ 110 кэВ при 13 МэВ [30]. Это позволяет проводить в одном опыте за сравнительно короткое время измерения ряда сечений таких как, полное сечение [31], сечение образования γ-лучей [30], сечения деления [32] и др. Сечения образования γ-лучей при неупругом рассеянии нейтронов с меньшим разрешением, но достаточным для практических применений, измеряются также на "белых" пучках от линейных ускорителей электронов [33, 34]. То же относится и к исследованиям полных сечений в области энергий нейтронов примерно до 40 МэВ [35]. В более ограниченной области энергий нейтронов для измерений полных сечений применяются и ускорители Ван-де-Граафа, например [36, 37]. Величина энергетического разрешения здесь уступает достигнутому на циклотроне, а интенсивность нейтронов в сравнимых областях энергий составляет, по оценке авторов работы [37], примерно 0,1 интенсивности "белого" пучка от изохронного циклотрона.

"Белый" нейтронный пучок в сочетании с техникой времени пролета используется во многих лабораториях мира для изучения полных сечений на быстрых нейтронах. Однако, как указывается в работе [38], при характерных статистических погрешностях результатов примерно в 1%, расхождения данных различных авторов составляют величину около 10%. Систематические ошибки, по-видимому, связаны с трудностями правильного учета фона и с неточностями, обусловленными высокими скоростями счета. При сравнении результатов, полученных на моноэнергетических источниках и на "белых" пучках, наблюдаются также расхождения (0,1-1%) в энергетической шкале.

Исследования вторичного нейтронного спектра, возникающего из-за процессов упругого и неупругого рассеяния нейтронов и реакций типа (n, 2n) и т.п., ведутся в настоящее время на моноэнергетических источниках нейтронов. Определенную информацию о процессе неупругого рассеяния нейтронов можно получить и на "белом" пучке, исследуя γ-излучения, сопровождающие этот процесс. Однако при этом информация является только частичной, так как γ-излучение есть продукт вторичной реакции и ее характеристики могут полностью не отражать характер основной реакции. Кроме того, не всегда неупругое рассеяние нейтронов сопровождается γ-излучением, примером тому являются изотопы лития.

Использование "белого" пучка для измерений дифференциальных сечений упругого и неупругого рассеяния нейтронов в области энергий 0,2-16 МэВ обсуждается в работе [39]. Сочетание техники времени пролета с амплитудным анализом рассеянного нейтронного излучения позволило авторам измерить поперечные сечения и угловые распределения нейтронов, рассеянных на основном и первом возбужденном состоя-

нии ^{56}Fe в интервале энергий 2,70-6,62 МэВ с энергетическим разрешением около 5%. Неотъемлемым требованием метода является высокое амплитудное разрешение и необходимость проведения сложного анализа спектров.

4. ЗАКЛЮЧЕНИЕ

Развитие термоядерных исследований вызывает необходимость проведения достаточно большой программы нейтронных исследований значительно более широкого диапазона энергий, чем это требуется для реакторов деления. Характерной особенностью измерений здесь является почти полное отсутствие источников моноэнергетических нейтронов. Реакция взаимодействия протонов с тритием представляет наименее "засоренный" источник моноэнергетических нейтронов, особенно при ускорении трития. Однако ускорение ядер трития делает этот источник труднодоступным для широкого применения. В некоторых случаях могут использоваться и другие источники нейтронов, выбор которых зависит от поставленной задачи.

Значительная часть измерений ядерных данных, представляющих интерес для реакторов синтеза, проводится в настоящее время с помощью источников нейтронов сплошного спектра. Оптимальным источником в рассматриваемой области энергий является процесс развала дейтона при энергиях до 30-35 МэВ.

Сочетание систематических измерений на "белом" пучке с измерениями в отдельных точках на моноэнергетических нейтронах дает возможность получить ядерные данные в нужном энергетическом диапазоне. Существующие трудности измерений вызывают потребность в тщательном отборе ядерных констант, подлежащих определению, в физическом обосновании удовлетворяющих погрешностей изучаемых параметров, в допустимых энергетических разрешениях и т.п., аналогично тому, как это делается при формировании потребностей в ядерных данных для быстрых реакторов.

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ВЛИЯНИЕ ЯДЕРНЫХ ДАННЫХ НА ВОСПРОИЗВОДСТВО ТРИТИЯ В ТЕРМОЯДЕРНОМ РЕАКТОРЕ

А.А.ОГЛОБЛИН, Л.В.ЧУЛКОВ, Г.Б.ЯНЬКОВ

Институт атомной энергии им.И.В.Курчатова,
Москва,
Союз Советских Социалистических Республик

Abstract–Аннотация

INFLUENCE OF NUCLEAR DATA ON TRITIUM BREEDING IN A THERMONUCLEAR REACTOR.

The authors consider the influence of nuclear data on the tritium breeding coefficient in a thermonuclear reactor using the D-T cycle. A spherical isotropic source of 14-MeV neutrons surrounded by a monoisotropic wall and a tritium breeding zone is taken as a model. A homogeneous mixture of ^{6}Li, ^{7}Li and some isotope which simulates the construction materials within the breeding zone without forming tritium under the action of neutrons is chosen as the tritium breeding zone. The tritium breeding coefficient is calculated by the Monte Carlo method for various wall and zone thicknesses and various isotopic compositions within the zone. Total and differential cross-sections characterizing the main processes of interaction between neutrons and lithium isotopes, the shape of the energy spectrum of the secondary neutrons for lithium and cross-sections characterizing the wall material and the impurities are used as input data. The interaction of secondary charged particles with the blanket substance is ignored. Within the limits of the experimental errors, the authors vary some of the input data and study the sensitivity of the tritium breeding coefficient to such variations; the also investigate the influence of such variations on the relationship between breeder and construction materials. On the basis of their calculations, the authors make recommendations regarding the priority and required accuracy of neutron constant measurements for a thermonuclear reactor.

ВЛИЯНИЕ ЯДЕРНЫХ ДАННЫХ НА ВОСПРОИЗВОДСТВО ТРИТИЯ В ТЕРМОЯДЕРНОМ РЕАКТОРЕ.

Рассмотрено влияние ядерных данных на коэффициент воспроизводства трития в термоядерном реакторе, использующем Д-Т-цикл. В качестве модели реактора был взят изотропный цилиндрический источник нейтронов с энергией 14 МэВ, окруженный моноизотопной стенкой и зоной воспроизводства трития. В качестве зоны воспроизводства была выбрана гомогенная смесь лития-6, лития-7 и ванадия, иммитирующего конструкционные материалы внутри зоны и не образующего трития под действием нейтронов. Коэффициент воспроизводства трития вычислялся методом Монте-Карло для различных толщин стенки и различного изотопного состава зоны. В качестве входных ядерных данных использовались полные и дифференциальные сечения, характеризующие основные процессы взаимодействия нейтронов с изотопами лития, форма энергетического спектра вторичных нейтронов для лития, а также сечения, характеризующие материал стенки и примесь. Взаимодействием вторичных заряженных частиц с веществом бланкета пренебрегалось. В пределах экспериментальных погрешностей проводились вариации некоторых входных данных и исследовалась чувствительность коэффициента воспроизводства трития относительно этих вариаций. На основании проведенных расчетов делаются некоторые рекомендации о приоритете и точности измерения нейтронных констант для термоядерного реактора.

ВВЕДЕНИЕ

Плазменный цикл, использующий реакцию $T(d,n)^{4}He$ (Q= 17,6 МэВ), считается в настоящее время наиболее легко осуществимым источником термоядерной энергии. В этом случае бланкет – оболочка, окружающая плазму в реакторе, должен выполнять две основные функции: превращать энергию нейтронов в тепловую и воспроизводить тритий, сгоревший в плаз-

ме. Естественная смесь изотопов лития дает возможность эффективно регенерировать тритий во всей области энергий нейтронов, вплоть до 14 МэВ. Для этой цели на промежуточных и медленных нейтронах может использоваться реакция $^{6}Li(n,\alpha)T$ ($Q = 4{,}78$ МэВ), так как ее сечение растет по мере уменьшения энергии нейтронов. Реакция $^{7}Li(n,n'\alpha)T$ является эндотермической ($Q = 2{,}47$ МэВ) и поэтому может использоваться только на быстрых нейтронах; важно, что при этом не теряется нейтрон, который затем может вновь вызвать реакцию с образованием трития. Коэффициент воспроизводства трития T обычно представляют в виде суммы $T = T_6 + T_7$, где T_6 есть коэффициент воспроизводства трития вследствие реакции $^{6}Li(n,\alpha)T$, а T_7 – вследствие реакции $^{7}Li(n,n'\alpha)T$.

Одной из проблем, возникших при нейтронных расчетах бланкета и затрудняющих уже на данном этапе инженерные разработки моделей термоядерных реакторов, является большая неопределенность в ядерных данных. Неопределенность в ядерных данных для одних элементов, их отсутствие для других – могут существенно повлиять на выбор и оптимальное соотношение между воспроизводящими и конструкционными материалами в бланкете, на его размеры, на коэффициент воспроизводства трития, картину тепловыделения в бланкете, а также на оценку величины радиационных повреждений, активации материалов и т.п. В настоящее время формулируются первоочередные потребности в ядерных данных. По мере дальнейшего развития идей по осуществлению управляемых термоядерных реакторов, а особенно – работ по технологии реакторов синтеза, потребности в ядерных данных значительно возрастут.

Это делает необходимым проведение достаточно широкой программы экспериментальных исследований по ядерным данным, главным образом в интервале энергий 5-15 МэВ. Однако проводить нейтронные исследования в этой области гораздо труднее, чем при более низких энергиях. Трудности измерений усугубляются отсутствием моноэнергетических источников нейтронов почти для всех значений энергии в указанном интервале. Более подробно этот вопрос обсуждается в работе [1].

В связи с этим представляет интерес рассмотреть вопрос о чувствительности основных параметров, характеризующих бланкет, к изменению ядерных данных. Решение этой задачи во всей ее полноте является весьма трудной проблемой. Данное исследование представляет попытку решения более частной задачи: изучение чувствительности коэффициента воспроизводства трития к изменению параметров основных реакций, протекающих в бланкете. Частично этот вопрос обсуждался в литературе. В работе [2] было показано, что учет резонансной самоэкранировки для процесса (n,γ) для ниобия привел к увеличению T_6 примерно на 5%, а увеличение сечения реакции $^{93}Nb(n,2n)^{92}Nb$ в 2,5 раза при одновременном уменьшении сечения $^{93}Nb(n,n'\gamma)^{93}Nb$ привело к увеличению T_6 примерно на 10%. При этом величина T_7 осталась неизменной. В трудах Международной рабочей сессии по технологии реакторов синтеза [3] имеется указание на то, что существующая неопределенность в сечении реакции $^{7}Li(n,n'\alpha)T$, равная 15-25% для различных интервалов энергий нейтронов, приводит к неопределенности в значении T около 0,1.

В данной работе использовались две модели зоны воспроизводства в бланкете. Одна из них содержала только литий, вторая – литий и ванадий, который добавлялся в качестве конструкционного материала; в обоих случаях зона была окружена отражателем из углерода. Коэффициент воспроизводства трития вычислялся методом Монте-Карло.

Для модели зоны с ванадием вычисления проводились при различных толщинах стенки из ванадия, отделяющей источник термоядерных нейтронов от зоны воспроизводства, а также для различного количества ванадия в литии. В обеих зонах поглощение промежуточных и медленных нейтронов практически определялось реакцией $^{6}Li(n,\alpha)T$. Задача определения оптимальных инженерных характеристик бланкета не ставилась.

При расчетах литиевой зоны изменялись сечение реакции $^{7}Li(n,n'\alpha)T$ и форма спектра вторичных нейтронов из этой реакции. При расчетах зоны, состоящей из лития и ванадия, изменялись сечения реакций $(n,n'\gamma)$ и (n, 2n) для ванадия. Взаимодействием вторичных заряженных частиц с веществом бланкета пренебрегалось. Основное внимание уделялось изменениям T_7, так как эта величина характеризует воспроизводство трития именно в этой области, где ядерные данные хуже известны, а измерения их представляют небольшие трудности.

МЕТОДИКА РАСЧЕТА И МОДЕЛЬ БЛАНКЕТА

Составленная программа предназначалась для расчетов энергетических спектров нейтронов внутри бланкета, скоростей различных ядерных реакций в нем и для определения коэффициента воспроизводства трития в различных интервалах энергий нейтронов.

Основной недостаток метода Монте-Карло заключается в больших затратах времени ЦВМ при моделировании редких событий. Известно несколько способов уменьшения дисперсии результатов расчета. Алгоритм настоящей программы основан на методах "расщепления" и "русской рулетки". Идея метода [4] состоит в том, что поток нейтронов по мере углубления в вещество не уменьшается, а искусственно поддерживается на постоянном уровне. Для этого толстый слой вещества разбивается на ряд более тонких слоев. При моделировании частица, пересекающая i-тую из введенных границ в направлении уменьшения потока, расщепляется на n_i одинаковых частиц, где n_i – произвольное целое число. Каждой частице приписывается новый вес, равный $1/n_i$ веса первичной частицы. Если же частица пересекает границу в обратном направлении, то к ней применяется метод рулетки: с вероятностью $1-1/n_i$ моделирование истории частицы прекращается, а с вероятностью $1/n_i$ – продолжается; при этом вес частицы увеличивается в n_i раз.

Применение описанного метода позволило сохранить примерно одинаковую статистическую точность полученных результатов во всем рассматриваемом диапазоне энергий нейтронов. В данной программе расчета толщина слоя выбиралась примерно равной толщине слоя половинного ослабления и n = 2. Программа была написана на языке ФОРТРАН применительно к вычислительной машине БЭСМ-6.

Величины сечений, использованные в расчетах, так же как и угловые распределения нейтронов задавались в отдельных энергетических точках. Значения между выбранными точками определялись линейной интерполяцией значений в соседних точках. Дифференциальные сечения при данной энергии представлялись в виде таблиц для 11 и 21 значения углов рассеяния в зависимости от сложности углового распределения.

Взаимодействие нейтронов с изотопами лития описывалось сечениями реакций, приведенных в табл.I. В расчетах использовались величины сечений указанных реакций при 60 значениях энергий нейтронов в интервале 10 кэВ – 14,5 МэВ. Ниже 10 кэВ сечения упругого рассеяния ней-

ТАБЛИЦА I. РЕАКЦИИ НА ИЗОТОПАХ ЛИТИЯ, ИСПОЛЬЗОВАННЫЕ В РАСЧЕТАХ

Реакции	Q (МэВ)	Реакции	Q (МэВ)
$^{6}Li(n,n)^{6}Li$	-	$^{7}Li(n,n)^{7}Li$	-
$^{6}Li(n,n'\gamma)^{6}Li$	-3,56	$^{7}Li(n,n'\gamma)^{7}Li$	-0,48
$^{6}Li(n,n'd)^{4}He$	-1,47	$^{7}Li(n,n'\alpha)T$	-2,47
$^{6}Li(n,2n'p)^{4}He$	-3,70	$^{7}Li(n,2n)^{6}Li$	-7,25
$^{6}Li(n,\alpha)T$	-4,78	$^{7}Li(n,2n'd)^{4}He$	-8,72
$^{6}Li(n,p)^{6}He$	-2,75	$^{7}Li(n,d)^{6}He$	-7,78

тронов полагались независимыми от энергии нейтронов, а сечение реакции $^{6}Li(n,\alpha)T$ определялось аналитически [5]. Угловые распределения упруго рассеянных нейтронов и вторичных нейтронов из реакций $^{6}Li(n,n'd)^{4}He$ и $^{7}Li(n,n'\alpha)T$ задавались при 14 значениях энергии нейтронов; спектры вторичных нейтронов из этих реакций формировались в виде гистограмм, однако в программе была предусмотрена возможность формирования спектра, используя аналитические выражения. Спектры вторичных нейтронов из реакций (n,2n) принимались испарительными. Ядерные данные для изотопов лития были взяты из работы [6].

Взаимодействия нейтронов с ядрами ванадия учитывались четырьмя процессами: упругое и неупругое рассеяние, реакция (n,2n) и поглощение нейтронов; поглощение нейтронов учитывало любое взаимодействие, приводящее к гибели нейтрона. Использованные величины сечений были представлены в виде таблиц для 45 значений энергий нейтронов в интервале 100 эВ – 14,5 МэВ. Подробно была описана только группа резонансов в сечении упругого рассеяния при энергиях нейтронов 3-10 кэВ. Ниже 100 эВ сечение упругого рассеяния сохраняло свое значение при 100 эВ, а сечение захвата нейтронов увеличивалось обратно пропорционально скорости нейтрона. Анизотропным в системе центра масс принималось только упругое рассеяние при энергиях выше 1 МэВ, угловые распределения которого были определены при 15 значениях энергий нейтронов. Энергетические спектры неупруго рассеянных нейтронов и нейтронов из реакции (n,2n) принимались испарительными. Ядерные данные для ванадия были взяты из работы [7].

Взаимодействие нейтронов с ядрами углерода описывалось реакциями упругого и неупругого рассеяния и $^{12}C(n,\alpha)^{9}Be$. Радиационный захват нейтронов и реакция $^{12}C(n,n'2\alpha)^{4}He$ не учитывались. Величины сечений были определены при 35 значениях энергий нейтронов в интервале 2-14,5 МэВ. Дифференциальные сечения упругого рассеяния нейтронов с энергией выше 2 МэВ задавались при 10 значениях энергии. Ниже 2 МэВ величина сечения упругого рассеяния представлялась полиномом четвертой степени от энергии [8]. При определении значений сечений для углерода использовались данные из работ [9-11].

Избранная для расчетов модель бланкета показана на рис.1. Изотропные источники нейтронов с энергией 14,2 МэВ распределялись равномерно внутри бесконечной цилиндрической зоны 1; зона 4 (зона воспроизводства трития) состояла из смеси лития и ванадия; графит ис-

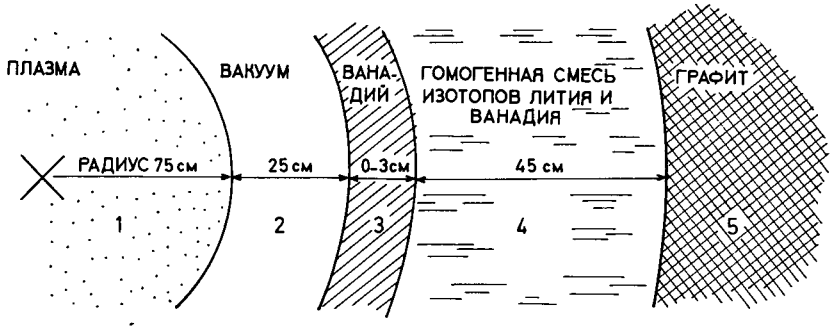

Рис.1. Модель бланкета, использованная в расчетах.

пользовался в качестве замедлителя и отражателя нейтронов. Судьба нейтрона прослеживалась в интервале энергий от 14,2 МэВ до 10 эВ; принималось, что практически все нейтроны, замедлившиеся ниже 10 эВ, поглощались ядрами лития-6 с образованием трития.

ЛИТИЕВАЯ ЗОНА ВОСПРОИЗВОДСТВА

На рис.2 приведены энергетические спектры нейтронов внутри зоны воспроизводства, которая состояла только из лития; стенка из ванадия также отсутствовала. Смесь изотопов лития содержала 10% ядер лития-6. Спектры были нормированы на один термоядерный нейтрон. Гистограммы 1 и 2 представляют соответственно нейтронные спектры в зоне 4 на глубине около 9 и 45 см от внутреннего края литиевой зоны. Поток нейтронов на глубине 45 см составлял примерно 0,3 от потока на глубине 9 см. Энергетическое распределение изменялось на этом расстоянии значительно сильнее: доля нейтронов с энергией выше 1 МэВ уменьшилась примерно от 55% до 25%.

На рис.3 представлена энергетическая зависимость коэффициентов воспроизводства трития T_6 и T_7 (левая шкала), а также энергетическая зависимость сечений соответствующих реакций (правая шкала). Как и ожидалось, реакция $^6Li(n,\alpha)T$ играла роль только при энергиях нейтронов, меньших значения порога реакции $^7Li(n,n'\alpha)T$; доля ядер трития в T_6, образованная нейтронами с энергией выше 3 МэВ составляла около 1%, а с энергией выше 0,1 МэВ – 25%. В то же время около 80% всех ядер трития в T_7 создавались нейтронами с энергией выше 10 МэВ. Известно, что при малых энергиях нейтронов сечение реакции $^6Li(n,\alpha)T$ определено с хорошей точностью [5]. Выше 0,1 МэВ погрешность возрастает и составляет для различных интервалов энергий нейтронов от 5% до 15%. Таким образом, для расчетов воспроизводства трития, дополнительные измерения сечения реакции $^6Li(n,\alpha)T$ требуются главным образом в интервале энергий нейтронов 0,1-3,0 МэВ.

В табл.II представлены скорости основных реакций, протекающих в бланкете с литиевой зоной воспроизводства. Скорость реакции определялась как отношение числа реакций данного типа к числу первичных нейтронов из источника. В первом столбце таблицы не указаны реакции, скорости которых были меньше 0,01, однако в строках, относящихся к

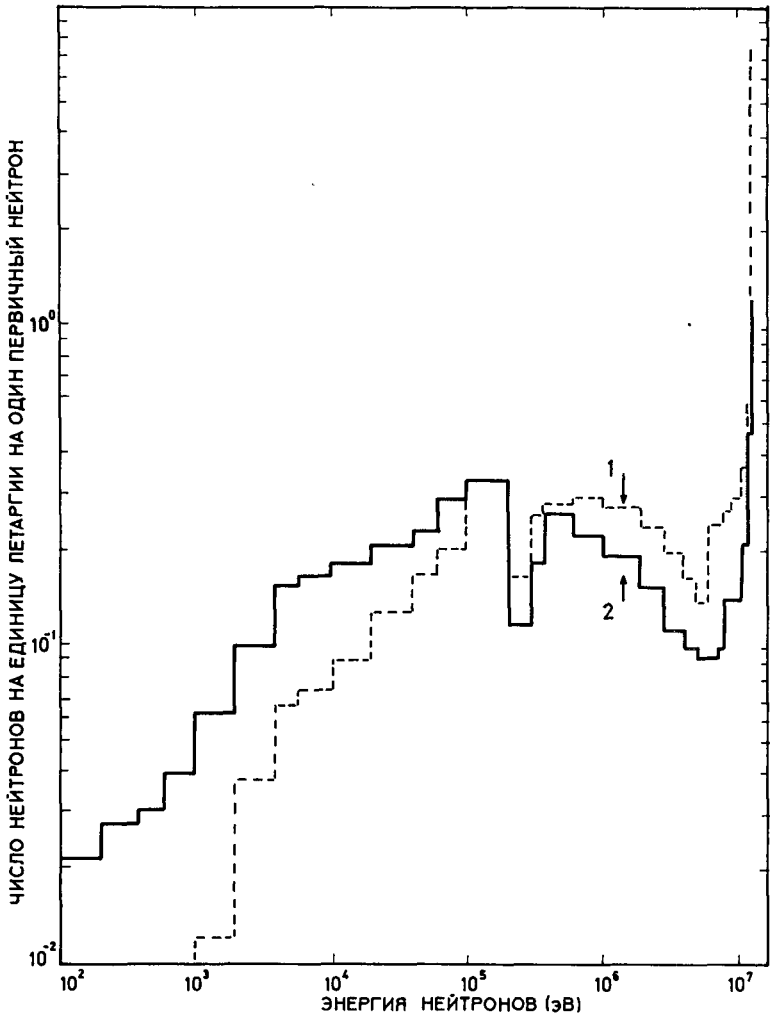

Рис.2. Спектры нейтронов в литиевой зоне воспроизводства.
1 – на глубине 9 см от внутреннего края зоны;
2 – на глубине 45 см от внутреннего края зоны.

суммарному поглощению и размножению нейтронов в бланкете, эти реакции учитывались. В силу того, что процесс упругого рассеяния нейтронов не оказывает прямого существенного влияния на производство трития, скорости соответствующих реакций для изотопов лития и углерода также не приведены в таблице, хотя величины скоростей реакций для этих процессов значительно превышают любую из указанных.

Во втором столбце таблицы представлены скорости реакций, когда величины сечений, использованные в расчетах, соответствовали рекомендованным значениям. Размножение нейтронов вследствие реакции

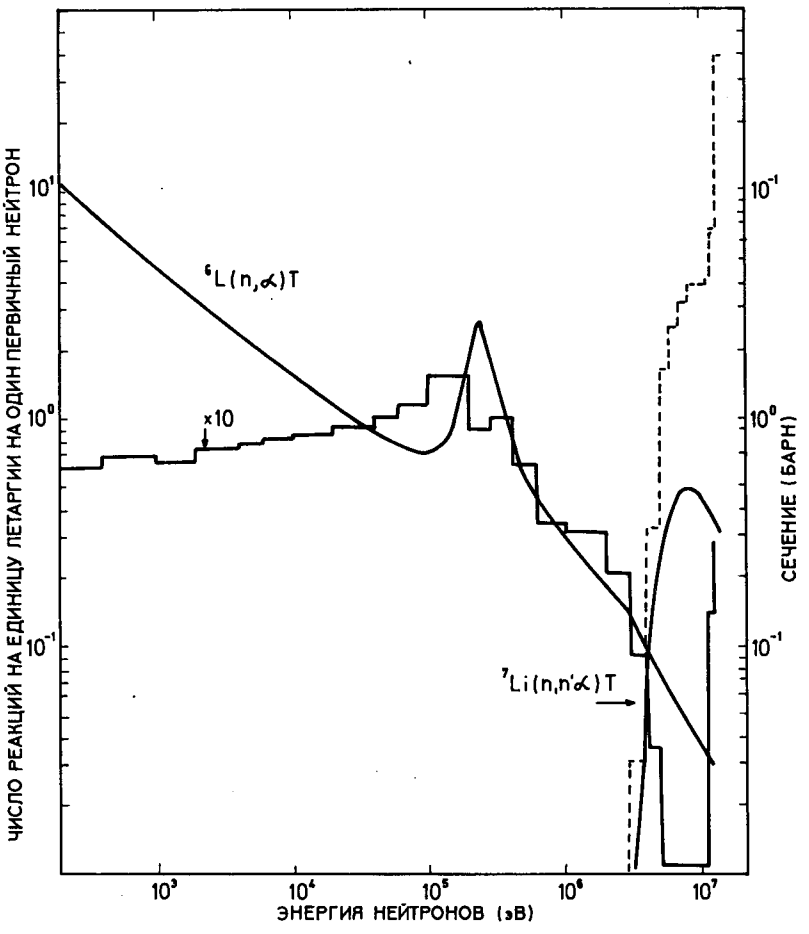

Рис.3. Зависимость воспроизводства трития от энергии нейтронов: —⌐⌐— T_6; --⌐⌐-- T_7. (Гладкими сплошными линиями показаны сечения соответствующих реакций).

(n,2n) в таком бланкете невелико, поэтому величина T_6 несколько меньше единицы. Реакция ^{7}Li(n, n'α)T, конкурируя с реакциями упругого рассеяния, ^{7}Li(n,n'γ)^{7}Li и ^{6}Li(n,n'd)^{4}He давала сравнительно большое значение $T_7 = 0{,}655 \pm 0{,}012$.

Числа в столбцах 3 и 4 соответствуют тем вариантам расчета, когда были изменены значения сечения реакции ^{7}Li(n,n'α)T на ± 10% по отношению к рекомендованным значениям для всех энергий нейтронов от порогового значения до 14,2 МэВ. Такое изменение соответствует примерно разбросу экспериментальных данных для указанного сечения. При этом соответствующим образом варьировалось и сечение упругого рассеяния нейтронов. Как видно из таблицы, изменение величины сечения на 20% привело к примерно такому же изменению величины T_7 (около 15%) и не изменило в пределах ошибок расчета других скоростей реакций, в

ТАБЛИЦА II. СКОРОСТИ РЕАКЦИЙ В БЛАНКЕТЕ С ЛИТИЕВОЙ ЗОНОЙ ВОСПРОИЗВОДСТВА

Параметры / Реакции	Исходные значения	Сечение реакции $^{7}Li(n, n'\alpha)T$		Спектр нейтронов $^{7}Li(n, n'\alpha)T$	
		Увеличено на 10%	Уменьшено на 10%	На уровень 4,63 МэВ	Испарительный
$^{6}Li(n, \alpha)T$ T_6	0,987 ±0,007	0,989 ±0,008	0,996 ±0,008	0,992 ±0,007	0,997 ±0,007
$^{6}Li(n, n'd)^{4}He$	0,108 ±0,011	0,107 ±0,014	0,101 ±0,013	0,118 ±0,012	0,100 ±0,010
$^{7}Li(n, n'\gamma)^{7}Li$	0,532 ±0,036	0,524 ±0,045	0,532 ±0,046	0,583 ±0,039	0,536 ±0,036
$^{7}Li(n, n'\alpha)T$ T_7	0,655 ±0,012	0,686 ±0,015	0,595 ±0,013	0,728 ±0,013	0,632 ±0,012
$^{7}Li(n, 2n)$	0,069 ±0,006	0,061 ±0,007	0,067 ±0,008	0,072 ±0,006	0,067 ±0,006
$^{12}C(n, \alpha)^{9}Be$	0,075 ±0,005	0,070 ±0,008	0,069 ±0,008	0,078 ±0,005	0,068 ±0,005
Суммарное поглощение	1,075 ±0,007	1,066 ±0,008	1,074 ±0,009	1,083 ±0,007	1,075 ±0,007
Суммарное размножение	0,075 ±0,007	0,066 ±0,008	0,074 ±0,009	0,083 ±0,007	0,075 ±0,007
$T = T_6 + T_7$	1,642 ±0,014	1,675 ±0,017	1,591 ±0,015	1,720 ±0,014	1,629 ±0,014

том числе и T_6. Отсутствие изменений в T_6 объясняется тем, что в литиевой зоне нет процесса поглощения нейтронов, который конкурировал бы с реакцией $^{6}Li(n, \alpha)T$. Однако даже в этом случае изменение сечения реакций $^{7}Li(n, n'\alpha)T$ в пределах экспериментальных погрешностей привело к изменению T примерно на 6%. Уменьшение неопределенности в значениях сечения этой реакции позволит более точно определить коэффициент воспроизводства трития.

Числа в столбцах 5 и 6 представляют величины скоростей реакций при измененном спектре вторичных нейтронов в реакции $^{7}Li(n, n'\alpha)T$. Рекомендованный спектр вторичных нейтронов [6] имел сглаженную форму, соответствующую экспериментальным данным работы [12], в которой утверждалось, что в основном указанная реакция идет через возбужденное состояние лития-7 с энергией возбуждения 4,63 МэВ. Однако при энергиях первичных нейтронов выше 10 МэВ экспериментальные данные [12] показывали, что имеется определенное число событий, отвечающих возбуждениям более высоких уровней ядра литий-7. В данном варианте расчета (столбец 5) принималась рекомендованная форма спектра для нейтронов с энергией, меньшей значения порога возбуждения уровня 4,63 МэВ (E_{trh} = 5,29 МэВ), а при энергиях первичных нейтронов выше порога предполагалось, что реакция протекает только через это возбужденное состояние. Такой спектр нейтронов является более жестким, чем рекомендованный. Это подтверждается ростом величины T_7, который составлял примерно 10%. Другие величины в пределах ошибок

не изменили своих значений. В столбце 6 приведены скорости реакций, когда использовалась испарительная форма спектра с постоянной температурой, равной 1,5 МэВ. Существенных изменений по сравнению с исходным вариантом расчета не наблюдалось. Аналогичный результат был получен и для случая, когда рекомендованная сглаженная форма спектра 6 была заменена гистограммами, непосредственно отвечающими экспериментальным данным работы [12]. Полученные результаты показали, что только значительная деформация спектра вторичных нейтронов в жесткой части может оказать существенное влияние на оценку величины T_7.

КОНСТРУКЦИОННЫЙ МАТЕРИАЛ В ЗОНЕ ВОСПРОИЗВОДСТВА

При наличии внутренней стенки (зона 3, рис.1) и добавлении в литий конструкционного материала, энергетический спектр нейтронов на небольших глубинах зоны воспроизводства становится значительно мягче. Доля быстрых нейтронов уменьшается за счет неупругих соударений с ядрами конструкционного материала главным образом за счет процесса неупругого рассеяния, в результате которого образуются нейтроны в основном испарительного спектра со средней энергией вблизи 1 МэВ. Для случая, когда в литии содержалось 10% ванадия по объему и стенка из ванадия имела толщину 3 см, доля нейтронов с энергией выше 1 МэВ на глубине 9 см составляла величину около 35%. Форма энергетического спектра для нейтронов с энергией ниже 1 МэВ зависит от соотношения сечений поглощения и рассеяния для выбранного конструкционного материала. Ванадий обладает значительно меньшим поглощением нейтронов за счет реакций (n, γ), чем, например, ниобий. Поэтому в зоне даже со значительным количеством ванадия поглощение промежуточных и медленных нейтронов практически определялось реакцией $^{6}Li(n, \alpha)T$.

По мере углубления в зону формы спектров сближаются. На рис.4 приведены для сравнения энергетические спектры нейтронов на глубине 45 см для литиевой зоны (пунктирная кривая) и зоны с ванадием (сплошная кривая). В последнем случае стенка имела толщину 3 см, а смесь содержала 10% ванадия. Спектры были нормированы на один термоядерный нейтрон. Величина потока нейтронов для зоны с ванадием составляла около 0,7 от величины потока для зоны без ванадия и без стенки. С учетом этого обстоятельства спектры являются весьма сходными.

В табл.III представлены скорости основных реакций в зоне с ванадием. Для облегчения сравнения во втором столбце приведены скорости реакций для литиевой зоны. Как и ранее, не указаны реакции, скорости которых были меньше 0,01, а также реакции упругого рассеяния. В столбцах 3-6 приведены величины для двух толщин стенки (3 см, 1 см) и для различного объемного содержания ванадия в литии (10%, 5%, 0%).

Сравнение данных в столбцах 2 и 3 показывает, что T_6 возросло примерно на 25%, а T_7 упало почти вдвое. Изменение T_6 и T_7 скомпенсировали друг друга так, что величина T уменьшилась всего на 5%. Рост T_6 обусловлен значительным увеличением размножения нейтронов вследствие реакции $^{51}V(n, 2n)\ ^{50}V$ при небольшом увеличении поглощения нейтронов. Сравнение данных в столбцах 3 и 4 для реакции $^{51}V(n, 2n)\ ^{50}V$ показывает, что около 70% этих событий происходило в толстой стенке, что естественно было ожидать, учитывая высокий энергетический порог

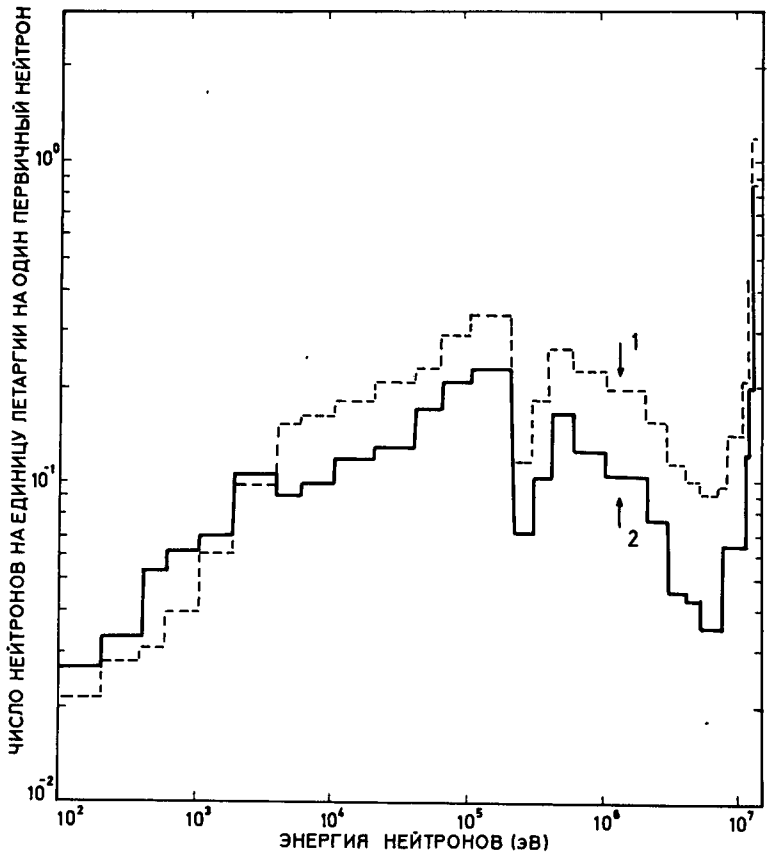

Рис.4. Спектры нейтронов на глубине 45 см от внутреннего края зоны воспроизводства. 1 – литиевая зона; 2 – зона с ванадием, толщина стенки 3 см, в смеси 10% ванадия.

этой реакции (E_{trh} = 11,3 МэВ). Однако небольшую величину скорости (из приведенных в таблице) имел процесс неупругого рассеяния нейтронов на ванадии.

Включение стенки толщиной 3 см и добавление в литий 10% ванадия оказывало примерно одинаковое влияние на уменьшение величины T_7. Зависимость T_7 от количества ванадия в литии проявлялась сильнее при тонкой стенке (1 см), чем при толстой стенке (3 см). Аналогично этому зависимость T_7 от толщины стенки проявлялась сильнее при меньшем количестве ванадия в литии.

В табл.IV приведены данные, показывающие как менялись скорости основных реакций в зоне с ванадием при изменении сечений реакции $^{51}V(n, n'\gamma)\,^{51}V$ (столбцы 3-5) и $^{51}V(n, 2n)\,^{50}V$ (столбцы 6,7). Зона воспроизводства содержала 10% ванадия в литии и была отделена от источника нейтронов стенкой толщиной 3 см. Условия составления табл.IV были аналогичны условиям составления табл.II. Второй столбец таблицы соответствует исходным рекомендованным значениям сечений [7].

ТАБЛИЦА III. СКОРОСТИ РЕАКЦИЙ ПРИ РАЗЛИЧНЫХ КОНСТРУКЦИЯХ БЛАНКЕТА

Параметры / Реакции	Толщина стенки из ванадия (T см) и его содержание в литии (P %)				
	T=0 P=0	T=3 P=10	T=3 P=0	T=1 P=10	T=1 P=5
$^{6}Li(n, \alpha)T$ T_6	0,987 ±0,007	1,223 ±0,006	1,153 ±0,008	1,151 ±0,010	1,098 ±0,007
$^{6}Li(n, n'd)^{4}He$	0,108 ±0,011	0,056 ±0,004	0,075 ±0,008	0,078 ±0,010	0,071 ±0,008
$^{7}Li(n, n'\gamma)^{7}Li$	0,532 ±0,036	0,303 ±0,014	0,438 ±0,030	0,342 ±0,030	0,381 ±0,026
$^{7}Li(n, n'\alpha)T$ T_7	0,655 ±0,012	0,335 ±0,004	0,456 ±0,008	0,412 ±0,009	0,502 ±0,009
$^{7}Li(n, 2n)$	0,069 ±0,006	0,033 ±0,002	0,051 ±0,005	0,046 ±0,006	0,051 ±0,005
$^{51}V(n, n'\gamma)^{51}V$	-	0,630 ±0,026	0,366 ±0,022	0,426 ±0,033	0,300 ±0,018
$^{51}V(n, 2n)^{50}V$		0,288 ±0,005	0,191 ±0,005	0,207 ±0,007	0,139 ±0,004
поглощение на V		0,055 ±0,007	0,029 ±0,006	0,053 ±0,012	0,032 ±0,006
$^{12}C(n, \alpha)^{9}Be$	0,075 ±0,005	0,040 ±0,002	0,057 ±0,003	0,042 ±0,003	0,058 ±0,003
суммарное поглощение	1,075 ±0,007	1,326 ±0,006	1,258 ±0,007	1,256 ±0,009	1,197 ±0,007
суммарное размножение	0,075 ±0,007	0,326 ±0,006	0,258 ±0,007	0,256 ±0,009	0,197 ±0,007
$T = T_6 + T_7$	1,642 ±0,014	1,558 ±0,007	1,609 ±0,011	1,563 ±0,014	1,600 ±0,012

Сопоставление данных в столбцах 3 и 4 показывает, что варьирование сечения неупругого рассеяния нейтронов в пределах 60% во всем интервале энергий при одновременном изменении сечения упругого рассеяния привело к сравнительно небольшому (около 4%) изменению величины T; при этом T_7 возросло примерно на 16%, а T_6 не изменилось. В столбце 5 приведены величины, соответствующие варианту расчета, когда сечение неупругого рассеяния варьировалось на 30% в более узком интервале энергий, а именно от 10 до 14,2 МэВ. Полученные данные показывают, что рост T_7 целиком объясняется изменением сечения только в этом интервале энергий. Отсутствие изменения величины T_6 было связано с тем, что в зоне с ванадием, также как и в литиевой зоне, для промежуточных и медленных нейтронов нет сильного процесса поглощения нейтронов, конкурирующего с реакцией $^{6}Li(n, \alpha)T$.

Таким образом, для более точной оценки величины T_7 необходимо уменьшение неопределенности в значениях сечения неупругого рассеяния нейтронов для конструкционных материалов, главным образом в интерва-

ТАБЛИЦА IV. СКОРОСТИ РЕАКЦИЙ В ЗОНЕ ВОСПРОИЗВОДСТВА С ВАНАДИЕМ (ТОЛЩИНА СТЕНКИ 3 см, В ЗОНЕ 10% ВАНАДИЯ)

Параметры / Реакции	Исходные значения	Сечение реакции $^{51}V(n, n'\gamma)^{51}V$			Сечение реакции $^{51}V(n, 2n)^{50}V$	
		Увеличено на 30%	Уменьшено на 30% а)		Увеличено на 30%	Уменьшено на 30%
$^{6}Li(n, \alpha)T$ T_6	1,223 ±0,006	1,221 ±0,006	1,233 ±0,006	1,231 ±0,008	1,299 ±0,008	1,172 ±0,008
$^{6}Li(n, n'd)^{4}He$	0,056 ±0,004	0,052 ±0,004	0,061 ±0,004	0,065 ±0,007	0,054 ±0,006	0,052 ±0,006
$^{7}Li(n, n'\gamma)^{7}Li$	0,303 ±0,014	0,280 ±0,014	0,324 ±0,016	0,296 ±0,020	0,304 ±0,020	0,308 ±0,021
$^{7}Li(n, n'\alpha)T$ T_7	0,335 ±0,004	0,312 ±0,004	0,362 ±0,004	0,363 ±0,007	0,302 ±0,005	0,360 ±0,006
$^{7}Li(n, 2n)$	0,033 ±0,002	0,040 ±0,003	0,038 ±0,003	0,038 ±0,005	0,031 ±0,004	0,036 ±0,005
$^{51}V(n, n'\gamma)^{51}V$	0,630 ±0,026	0,770 ±0,032	0,501 ±0,021	0,568 ±0,034	0,629 ±0,038	0,636 ±0,038
$^{51}V(n, 2n)^{50}V$	0,288 ±0,005	0,280 ±0,005	0,297 ±0,005	0,290 ±0,007	0,360 ±0,009	0,234 ±0,006
поглощение на V	0,055 ±0,007	0,070 ±0,009	0,063 ±0,008	0,064 ±0,012	0,063 ±0,011	0,064 ±0,012
$^{12}C(n, \alpha)^{9}Be$	0,040 ±0,002	0,035 ±0,002	0,033 ±0,002	0,035 ±0,003	0,026 ±0,002	0,033 ±0,003
суммарное поглощение	1,326 ±0,006	1,324 ±0,006	1,342 ±0,006	1,337 ±0,009	1,394 ±0,010	1,275 ±0,008
суммарное размножение	0,326 ±0,006	0,324 ±0,006	0,342 ±0,006	0,337 ±0,009	0,394 ±0,010	0,275 ±0,008
$T = T_6 + T_7$	1,558 ±0,007	1,533 ±0,007	1,595 ±0,007	1,594 ±0,010	1,601 ±0,009	1,532 ±0,010

а) В столбце 5 сечение изменено в интервале энергий 10-14,2 МэВ.

ле энергий нейтронов 10-15 МэВ. Кроме того, отклонение спектра вторичных нейтронов от испарительной формы, особенно в его жесткой части, может оказать существенное влияние на величину T_7.

Сравнение данных в столбцах 6-7, когда сечение реакции $^{51}V(n, 2n)\ ^{50}V$ варьировалось в пределах 60%, показывает, что величины T_6 и T_7 изменились примерно на 10% и 20%, соответственно; при этом величина T изменилась всего на 5%. Такая компенсация изменений произошла потому, что одновременно с изменением сечения реакции (n, 2n) было изменено сечение упругого рассеяния. Если бы избыток сечения из реакции (n, 2n) был включен в сечение неупругого рассеяния, то, как показали расчеты, величина T_7 не изменилась бы. Этот результат согласуется с данными работы [2], когда изменение сечения реакции Nb(n, 2n) в 2,5 раза при одновременном изменении сечения неупругого рассеяния не повлияло на величину T_7.

Таким образом, для правильной оценки величины T_6 и T_7 необходимо хорошее знание парциальных сечений для процессов, в результате которых нейтроны не теряются.

ЗАКЛЮЧЕНИЕ

Целью настоящей работы являлось исследование чувствительности коэффициента воспроизводства трития к изменению характеристик основных реакций, протекающих в зоне воспроизводства. Главное внимание уделялось той части коэффициента воспроизводства, которая создается за счет реакции на литии-7. Это определялось тем обстоятельством, что в интервале энергий нейтронов 5-15 МэВ ядерные данные известны гораздо хуже, чем при меньших энергиях, где определяющей в коэффициенте воспроизводства является реакция на литии-6. В то же время изучение ядерных данных с высокой точностью в указанном интервале быстрых нейтронов представляет большие трудности.

Модель бланкета, использованная в данной работе, обладает пониженной чувствительностью T_6 к изменению сечений, так как не содержала материалов, сильно поглощающих промежуточные и медленные нейтроны. Отсутствовала и утечка нейтронов из бланкета. Однако эти ограничения не оказывали существенного влияния на воспроизводство трития вследствие реакции на литии-7 на быстрых нейтронах.

Полученные данные свидетельствуют о том, что коэффициент воспроизводства трития не является величиной, сильно зависящей от изменения ядерных данных для воспроизводящих и конструкционных материалов. В определенной степени это связано с тем, что все идеализированные модели бланкета богаты нейтронами. Естественно ожидать, что в реальном бланкете коэффициент воспроизводства окажется меньше и требования к его определению будут жестче.

Несмотря на простоту рассмотренных моделей бланкета, можно сделать некоторые рекомендации для определения ядерных данных.

Реакция $^{6}Li(n, \alpha)T$ не играет существенной роли при энергиях нейтронов выше порога реакции $^{7}Li(n, n'\alpha)T$. Ниже 0,1 МэВ сечение этого процесса известно с достаточной точностью. Поэтому наибольший интерес представляют дополнительные измерения в области энергий нейтронов 0,1-3,0 МэВ.

Сечение реакции $^{7}Li(n, n'\alpha)T$ известно в среднем с точностью 20%. Изменение величины сечения в этих пределах приводит к примерно такому же изменению в T_7. Дальнейшее уточнение сечения указанной реакции важно в интервале энергий, примыкающим к значению 14 МэВ, так как 80% всего количества трития в T_7 дают нейтроны с энергией выше 10 МэВ.

Тонкая структура спектра вторичных нейтронов из реакции $^{7}Li(n, n'\alpha)T$ не оказывает влияния на воспроизводство трития. Только значительные отклонения формы спектра от имеющихся данных, причем в его наиболее жесткой части, смогут оказать влияние на оценку T_7.

Наиболее существенным процессом, конкурирующим с реакцией $^{7}Li(n, n'\alpha)T$, является процесс неупругого рассеяния нейтронов на конструкционных материалах. Неопределенность в ядерных данных для этого процесса (сечение, форма спектра вторичных нейтронов в жесткой части), особенно при энергиях нейтронов выше 10 МэВ, должна быть значительно уменьшена.

Процесс (n, 2n) на конструкционных материалах влияет как на T_6, так и на T_7, но в противоположных направлениях. Как правило, сечение этого процесса для большинства конструкционных материалов известно с недостаточной точностью. Экспериментальные данные по спектрам вторичных нейтронов практически отсутствуют. Дальнейшие измерения здесь крайне желательны.

В заключение необходимо подчеркнуть, что данные рекомендации сделаны на основании изучения чувствительности к вариациям ядерных данных только для одного параметра модели реактора-коэффициента воспроизводства трития. Более полная картина может быть получена только после всестороннего обсуждения этих вопросов для всех основных параметров термоядерного реактора.

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DISCUSSION

TO PAPERS IAEA-SM-170/22 AND 21

G. CASINI: Have you tested your calculation method and nuclear data against the benchmark calculation known as the Blow and Steiner reference case?

G.B. YANKOV: We have not set ourselves the task of making an engineering calculation of the blanket. The purpose of the calculation was to obtain information on the sensitivity of the breeding ratio of tritium, especially T_1, to variations in nuclear data. We have not compared our calculations with those of Blow and Steiner since we do not have detailed data or detailed values.

G. CASINI: Concerning the use of vanadium, could you give the source of the nuclear data you used and indicate whether you have data calculations related to transmutation effects and decay heat of this material?

G.B. YANKOV: We took our nuclear data for vanadium from a paper of PENNINGTON, E.M., GAJNIAK, J.C., ANL-7387 (1968). We have not made calculations for transmutation effects and decay heat.

Yu.F.CHERNILIN (Chairman): It has been decided that this would be an appropriate time for the presentation of two special comments. The first is concerned with the use of a semi-classical optical model for the calculation of fast neutron cross-sections.

J. CSIKAI: Data requirements for shielding calculations [1] include a knowledge of fast-neutron total and elastic cross-sections. A systematic

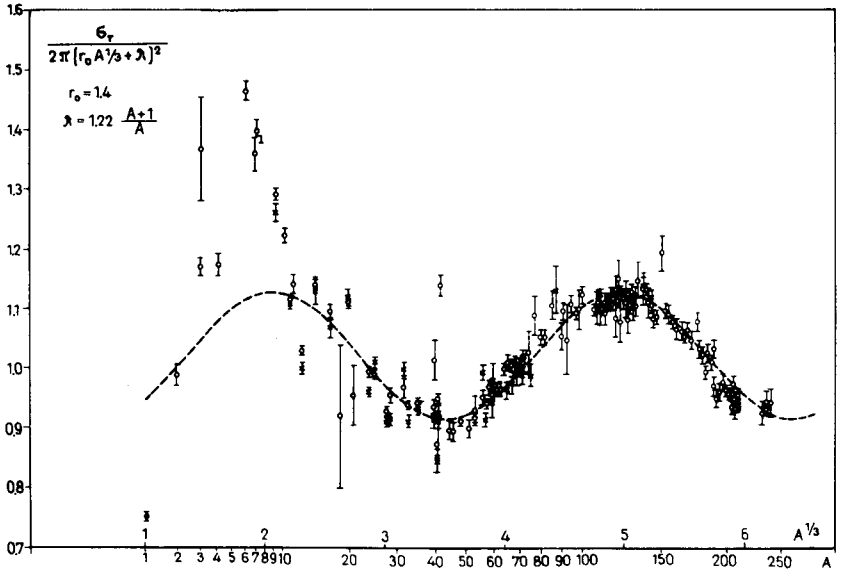

FIG. A. Cross-sections described by simple empirical expression.

[1] ABAGYAN, A.A., DUBININ, A.A., SUVOROV, A.P., Nuclear Data Requirements for Reactor Shielding Calculations INDC(CCP)-28/U (1972). Report presented at the Panel on Nuclear Data Requirements for Shielding Calculations held during the Fourth International Conference on Reactor Shielding, Paris (9-13 October 1972).

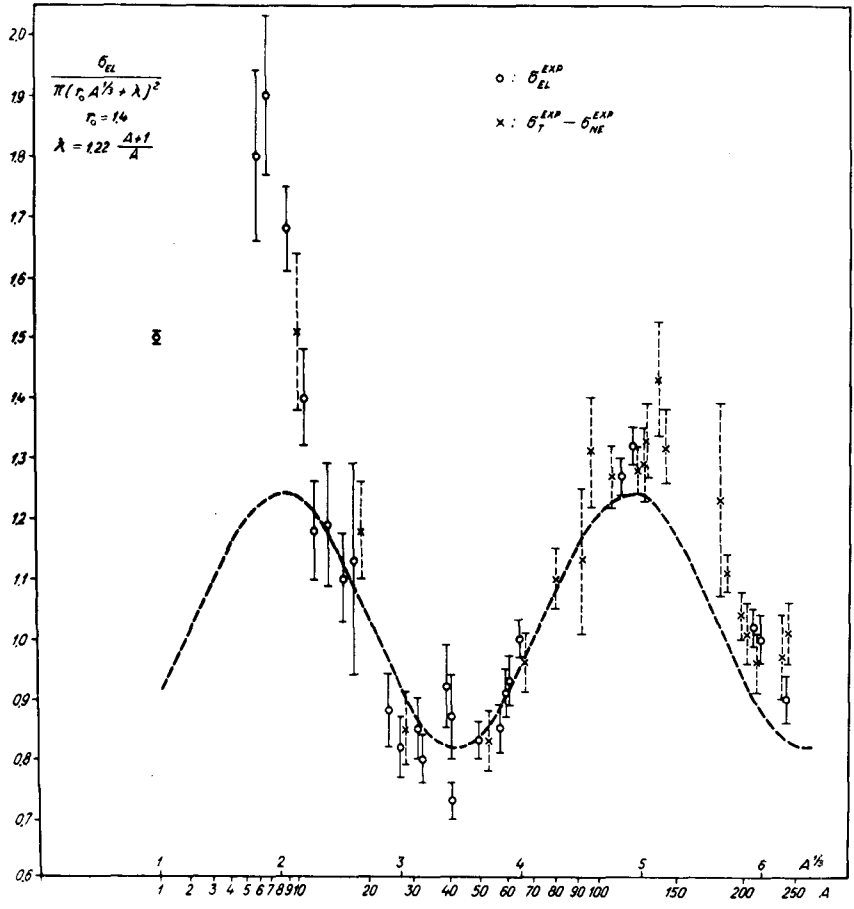

FIG. B. Integrated elastic cross-sections.

analysis of total and of integrated elastic and non-elastic cross-sections performed at the Institute of Experimental Physics, Kossuth University, Debrecen, has shown that total cross-sections can be described by a simple empirical expression in a wide range of energy and mass number (Fig. A) which can be interpreted by a semi-classical model [2, 3].

Further investigations [4], at 14 MeV, resulted in a description of integrated elastic (Fig. B) and non-elastic cross-sections (Fig. C); the latter work led to the conclusion that, in calculations of neutron cross-sections, a mass-dependent nuclear-radius parameter γ_0/A should be used. This mass dependence is similar to that of the electromagnetic γ_0/A values.

At present, work is in progress on describing the differential scattering cross-sections for fast neutrons by the same simple model, i.e. with much less computing time being required than in the case of quantum-mechanical

[2] ANGELI, I., CSIKAI, J., Nucl. Phys. A158 (1970) 389;
[3] ANGELI, I., CSIKAI, J., Nucl. Phys. A170 (1971) 577.
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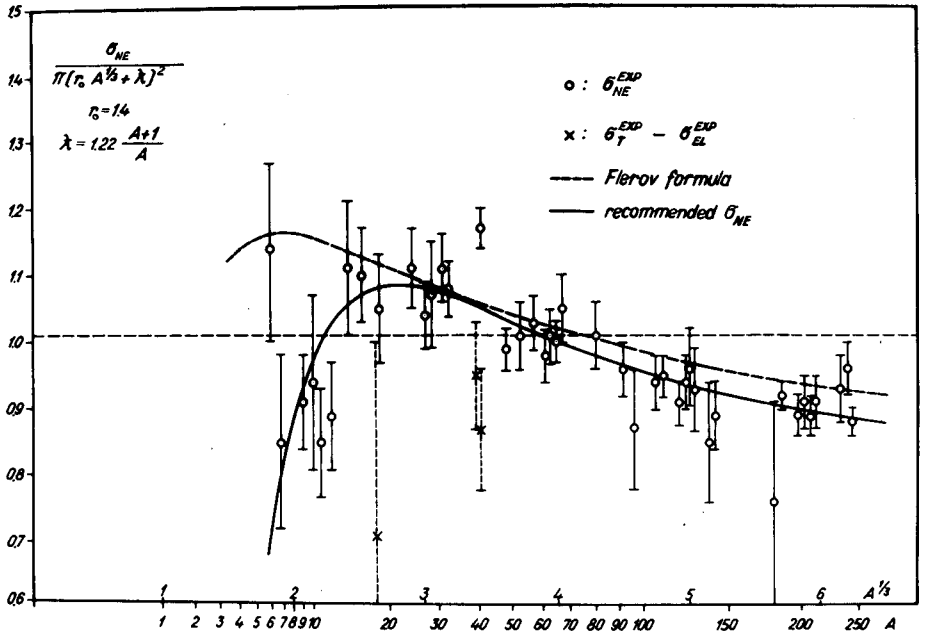

FIG. C. Integrated non-elastic cross-sections.

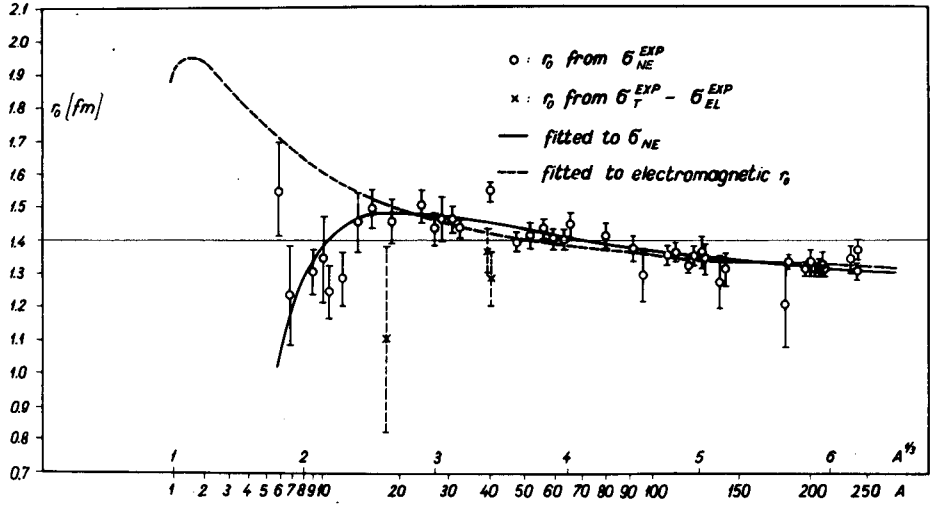

FIG. D. r_0 (fm) versus $A^{1/3}$.

optical models. This is important, for example, in Monte-Carlo shielding calculations, where angular distributions are frequently computed. According to the preliminary results, angular distributions – especially in the forward peak – are well described both in form and absolute value. This is perhaps of interest for the energy transport processes. Detailed results will be published later.

Yu. F. CHERNILIN (Chairman): The second comment is on the subject of (n, 2n) and (n, t) cross-sections for D+T neutrons.

J. CSIKAI: It is well known that for thermonuclear devices, cross-sections of D+T and D+D neutrons are needed with an accuracy of 5% (e.g. for (n, t), (n, 2n) and (n, 3n) reactions, from the point of view of tritium regeneration and neutron multiplication). According to our survey, (n, 2n) cross-sections have been measured for about 30% of the nuclei for D+T neutrons. On the basis of experimental data and N-Z systematics, an evaluation procedure was followed to obtain the most probable cross-section values. Because of the large inaccuracies in the experimental (n, 2n) data, no standard averaging methods could be applied. Therefore a generalized averaging procedure was used. In the case of those nuclei for which measurements are difficult to perform, the cross-sections were estimated by means of N-Z systematics. The cross-sections for elements have also been calculated at 14.7 MeV by averaging over isotopic abundances (Fig. a). The comparisons with empirical formulae (Fig. b) show that Pearlstein's formula gives the best agreement with experimental data. Results can be found in INDC (HUN)-10/L (1973).

Data on (n, t) reactions induced by fast neutrons are rather scarce since the very low cross-section prevents direct detection of the emitted tritons. Measurement of the activity of residual nuclei is also difficult in most cases because of interfering nuclear reactions and radiation.

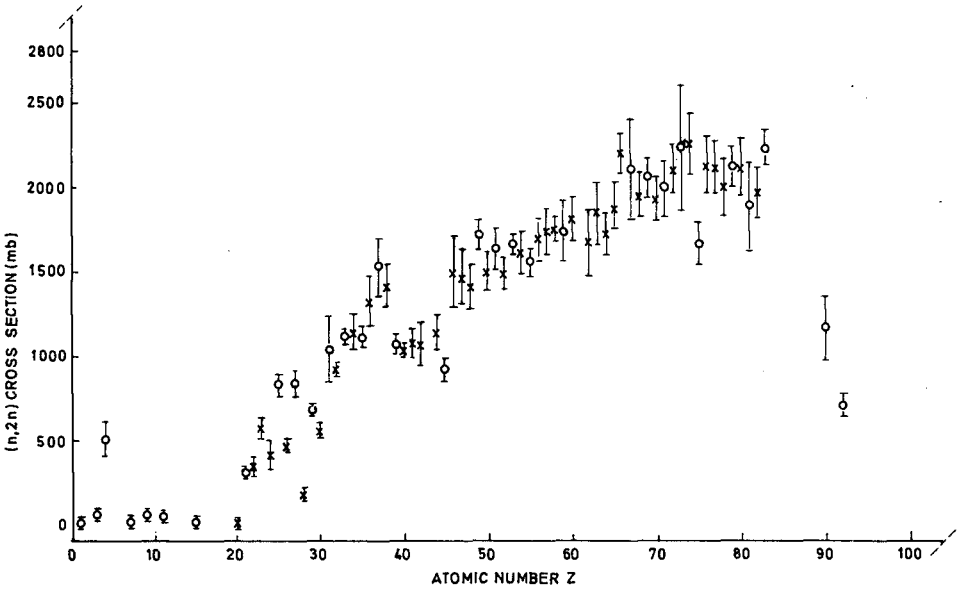

FIG. a. (n, 2n)-cross-sections for elements at 14.7 MeV.

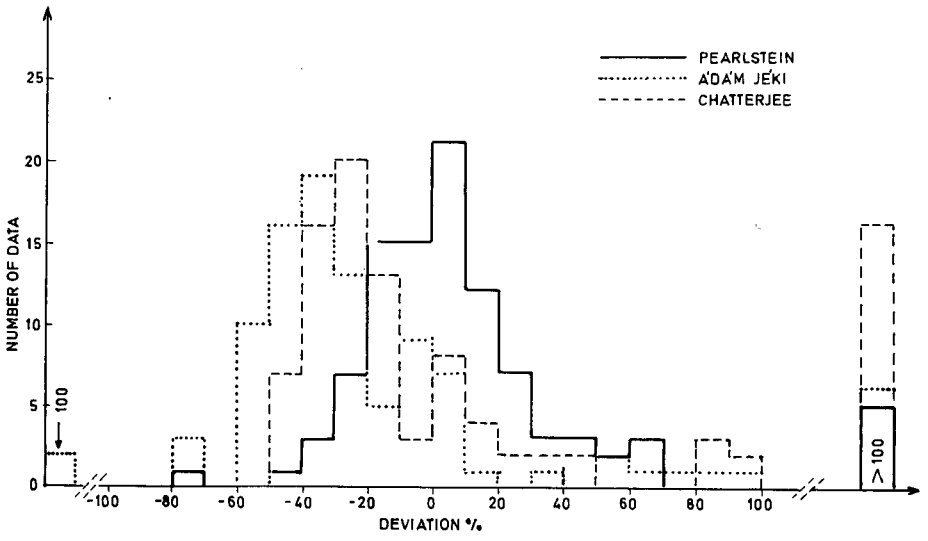

FIG. b. Distribution of relative differences between experimental and theoretical cross-sections.

Measurements of tritium beta activity afford a means of interference-free cross-section determination on any kind of nuclei. This method can be used for determining cross-sections for natural elements. The data for (n, t) cross-sections are few and inconsistent. As can be seen in Fig. c, the cross-sections for very light nuclei are relatively large (~100 mb), while for medium and heavy nuclei they are in the microbarn region. There are no theoretical methods or systematics by which the unknown data can be estimated. Further measurements are needed to determine the cross-sections for the structural materials of thermonuclear devices.

R. NICKS: My question concerns inelastic scattering cross-sections. Did you calculate total inelastic scattering cross-sections or partial cross-sections corresponding to the different excitation levels? Did you also deduce the associated gamma spectra? If you did not, it seems to me that your calculations are less useful for shielding calculations.

J. CSIKAI: We have a compilation for the total inelastic-scattering cross-sections and for some levels, mainly for isomeric states. The evaluation of these cross-sections is in progress. So far, we have no results for the associated gamma-ray spectra.

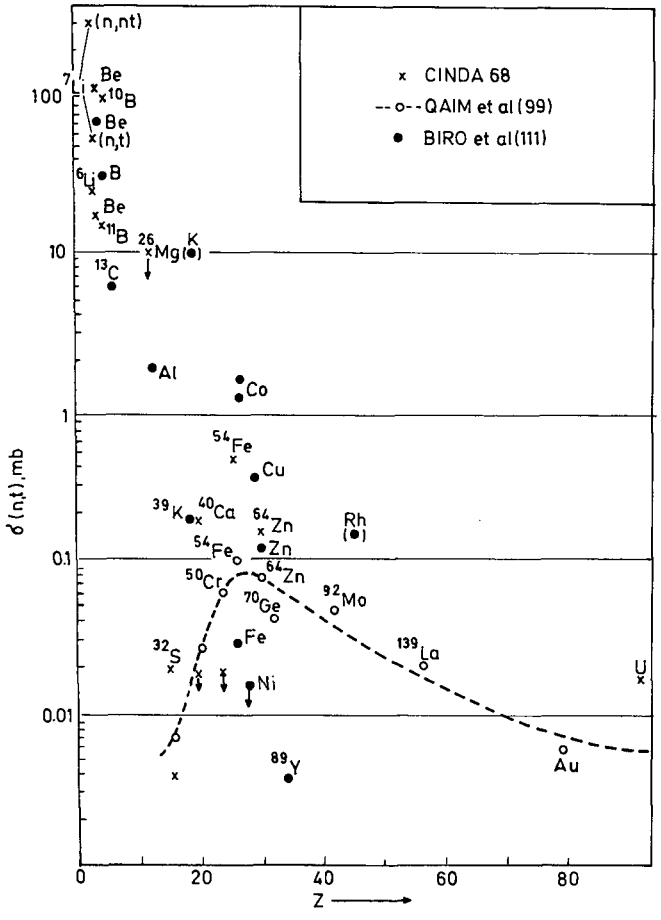

FIG. c. σ(n, t) versus Z.

NUCLEAR-FUSION CHAIN-REACTION APPLICATIONS IN PHYSICS AND ASTROPHYSICS

J. RAND McNALLY, Jr.
Oak Ridge National Laboratory,* Oak Ridge, Tenn.,
United States of America

Abstract

NUCLEAR-FUSION CHAIN-REACTION APPLICATIONS IN PHYSICS AND ASTROPHYSICS.

Nuclear-fusion chain reactions have been proposed for supernovae and controlled fusion reactors. The concept of fusion chain reactions will be traced with emphasis on possible applications in physics and astrophysics. Reaction-kinetics calculations of D-T and D-^{3}He fusioning plasmas reveal the presence of suprathermal particles as the end products of the reactions. These suprathermal end products can be active chain centres for the propagation of energy-releasing fusion chain reactions with fuels having $Z \geq 3$. A ^{6}LiD-fuelled fusion reactor has a calculated Lawson number, $n\tau$, lower by a factor of three compared to a pure-deuterium-fuelled fusion reactor. The first ^{6}LiD-fuelled fusion reactor will be sub-critical with the electrons sustained at MeV temperatures by electron cyclotron heating. If net power producers using ^{6}LiD-fuel are eventually feasible they would be large in size, well reflected, and have a high beta ($\beta = 8\pi nkT/B^2$) in order to minimize synchrotron radiation losses. Closed magnetic configurations would also be required to ensure the propagation of fusion chain reactions. Higher-powered d.c. accelerators would be essential to exploit this field of controlled fusion reactors, although a scientific feasibility experiment may be executed with existing accelerators ($\sim$ 2 mA, 4 MeV H_2^+). Present-day astrophysical calculations appear to be inadequate to account for the rapid nuclear processes in astrophysical explosions inasmuch as such calculations neglect the presence of suprathermal chain centres and multiplying chain reactions. The 55-day light decay of Type-I supernovae may possibly be explained by the production of ^{7}Be(53.61 d) in chain-reaction burning of neon and oxygen followed by ^{7}Li-burning in the residual star. Evaluation of these new prospects will require more accurate data on and broader coverage of the pertinent nuclear reaction cross-sections for fuels Li - S and n, p, d, t, ^{3}He, α-particles as suprathermal (E up to 20 MeV) chain centres.

PREFACE

No present controlled fusion experiment is within a factor of 1000 of the temperature, density, and confinement time product, $Tn\tau > 10^{22}$ K cm^{-3} sec, condition required for a scientific demonstration for fusion analogous to that obtained in 1942 by Fermi for the fission process. I shall present some ideas on a relatively new, bold and expensive approach to a scientific feasibility experiment for controlled fusion. The major ingredient of this approach is the concept of charged particle nuclear fusion chain reactions at MeV energies for fuels like ^{6}LiD and catalysts or suprathermal chain centers like p, d, t, ^{3}He, and α which permit the more rapid burning of ^{6}LiD to alpha particles than the direct reaction does. One of the chief requirements for evaluating the concept of nuclear fusion chain reactions in controlled fusion research is the accurate, wide-ranging determination of absolute nuclear reaction cross sections as well as those for nuclear elastic and inelastic processes.

A fitting corollary to these ideas is the concept of nuclear fusion chain reactions in the field of astrophysics, such as the gigantic explosions of supernovae. Here again there is a real need for accurate

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* Operated by Union Carbide Corporation for the US Atomic Energy Commission.

nuclear data which could lead to the evaluation of new and pregnant ideas on nuclear fusion chain reactions involving fuels like C, N, O, Ne, etc. Such processes might explain the production of the light elements, Li, Be and B, which is apparently an unresolved puzzle in astrophysical systems.

1. INTRODUCTION

Nuclear fusion chain reactions are very analogous to chemical chain reactions. In contradistinction to fission chain reactions which give a multiplication of neutrons in one reaction step, the nuclear fusion and chemical chain reactions generally produce a multiplication of catalysts or suprathermal chain centers in two or more reaction steps with an overall net energy release in the complete chain sequence. These suprathermal particles speed up the reaction rate by by-passing the direct thermal (or thermonuclear) reaction. Thermal or thermonuclear reactions between two fuel nuclei will generate additional chain centers and thus can lead to an increased possibility of exponentially growing chain reactions. The complete process should probably be referred to as a chain-thermal or chain-thermonuclear reaction.

INITIATION STEP	$h\nu + Cl_2 \longrightarrow 2Cl + (E_{h\nu} - 2.48)$ ev
PROPAGATION CYCLE	$Cl + H_2 \longrightarrow HCl + H - 0.04$ ev
	$H + Cl_2 \longrightarrow HCl + Cl + 2.00$ ev
LINEAR TERMINATION STEPS	$H + WALL \longrightarrow H{\cdot}WALL + \Delta E_1$
	$Cl + WALL \longrightarrow Cl{\cdot}WALL + \Delta E_2$
MUTUAL TERMINATION STEPS	$H + Cl + X \longrightarrow HCl + X + 4.44$ ev
	$H + H + X \longrightarrow H_2 + X + 4.48$ ev
BRANCHING STEP	$KE + Cl_2 \longrightarrow 2Cl + (KE - 2.48)$ ev

FIG. 1. Chemical chain reactions in hydrogen-chlorine gas with free atoms of H and Cl as chain centres.

The first chemical chain reactions were recorded by Max Bodenstein, a German chemist, who observed that a single initiation reaction in molecular hydrogen and chlorine gas mixtures stimulated a sequence of chain reactions which propagated through the chemical fuel several tens of thousands of times[1]. His proposed chemical chain reaction processes were later correctly identified as involving free atoms of H and Cl as the active chain centers by Nernst (Fig. 1). The propagation chain involves a free chlorine atom reacting with H_2 to produce HCl and a free hydrogen atom which then can react with a chlorine molecule to produce both a net energy release and the regeneration of a new free chlorine atom, which could keep the chain reaction going. Chain breaking reactions include wall or plasma recombination processes and will tend to stop the chain. On the other hand, chain branching reactions, such as the dissociation of HCl, H_2 or Cl_2 molcules by photon, electron, or atom collision processes, will continue or even proliferate the chains. In general, the active chain centers are slightly suprathermal in energy since they carry energy provided by the center of mass of the energetic colliding species as well as the energy released in the reaction. The presence of suprathermal chain centers and multiplying chain reactions is extremely important in the explanation of rapid combustion or explosive processes.

Practically all chemical combustion or explosion processes depend on chain or chain-thermal reaction steps. In the case of nuclear fusion chain reactions, we find analogous examples of the initiation steps, propagation chains, and both chain breaking and chain branching steps of chemical chain reactions, as well as breeding reactions which can produce exotic nuclear fuels like tritium or ^{3}He as highly active chain centers.

At high plasma temperatures, where one expects endothermic and inverse reactions to occur as well as increased radiation losses, the important property of a negative temperature coefficient for the control of the burning or operating temperature of the chemical or nuclear system becomes of importance. The nuclear operating temperature is quite distinct from the kindling or ignition temperature for the nuclear fire. It was apparently first discussed for the thermonuclear case by Huss[2] and later by Ohta, et al[3] and others. By proper control of fuel feed, ash loss, and confinement configuration one can maintain a stable burning temperature. The reacting system is thermally unstable at, or just above, the ignition temperature and flashes over to the "burning temperature" of the system. As ash build-up occurs this burning temperature decreases in general to a somewhat lower operating temperature though still above the ignition temperature if burning continues. If, on the other hand, the ashes include suprathermal particles which can operate as active chain centers to speed up the reaction, the system will be again thermally unstable and a somewhat higher, secondary burning temperature will be established.

The concept of "temperature" is ill defined in both chemical and nuclear combustion processes: the chain centers are suprathermal, i.e., somewhat hotter than the electrons or fuel atoms or nuclei. This has been shown in calculations at the Culham and Lawrence Livermore Laboratories for d·t and d·^{3}He nuclear fusioning systems in which the protons and alpha particles are calculated to be at least ten times hotter than the electrons.

Nuclear fusion chain reactions were apparently first proposed by Ulrich Jetter, a German engineer, back in 1950[4](Fig.2). He chose ^{6}LiD as the basic nuclear fusion fuel and selected neutrons and tritons as the active chain centers. The propagation cycle which he proposed utilized the two most reactive light element fusion reactions (d·t and n·^{6}Li) and would burn ^{6}LiD to alpha particles much faster than the direct reaction $^6\text{Li} + \text{D} \rightarrow 2\alpha + 22.4$ MeV. In addition, he suggested thermal or thermonuclear chain branching reactions involving the $d \cdot d_t$ and $d \cdot d_n$ processes. He also proposed neutron branching reactions involving the fast neutron dissociation of the deuteron or ^{9}Be, or the fission of uranium. These branched reactions give a multiplication of the number of active neutron and triton chain centers, and thus can speed up the reaction many-fold.

Fuel: ^{6}LiD; Chain Centers: n and t

Propagation Chain	$^6\text{Li} + n \longrightarrow t + \alpha +$	4.8 Mev
	$d + t \longrightarrow n + \alpha +$	17.6 Mev
Branching Steps	$d + d \longrightarrow n + {}^3\text{He} +$	3.3 Mev
	$d + d \longrightarrow t + p +$	4.0 Mev
	$n + d \longrightarrow p + 2n -$	2.2 Mev
	$(n + {}^9\text{Be} \rightarrow 2n + 2\alpha -$	1.7 Mev)

Ulrich Jetter, Physik. Blätter 6, 199 (1950).

FIG.2. The first proposed nuclear-fusion chain reaction.

PROPAGATION CHAIN

$$\begin{cases} {}^3He + d \longrightarrow p + \alpha + 18.4\ MeV \\ p + {}^6Li \longrightarrow {}^3He + \alpha + 4.0\ MeV \end{cases}$$

BRANCHING CHAINS

$$d + d \longrightarrow p + t + 4.0\ MeV$$
$$d + d \longrightarrow n + {}^3He + 3.3\ MeV$$

RE-INJECTION OF ^3He AND t-DECAY PRODUCT (^3He) INTO LEAKY MAGNETIC MIRROR

*R. F. POST, *NUCLEAR FUSION SUPPLEMENT*, PART 1 (1962) 119.

FIG. 3. The first proposed charged-particle nuclear-fusion chain reaction for controlled fusion applications [5].

It is of historical interest to note that, shortly after the discovery of the neutron by James Chadwick, Leo Szilard proposed a chain reaction probably involving the $^9Be(n,2n)2\alpha$ - 1.7 MeV neutron multiplying reaction, but apparently Szilard could not find any way to keep the neutrons hot to maintain the chain reaction.

In 1961 Richard Post of Lawrence Livermore Laboratory proposed a nuclear fusion chain reaction involving only charged particles[5]—such a process might permit the magnetic containment of the energetic reaction products in controlled fusion reactors. Figure 3 illustrates the essential elements of his scheme. Since Post was at that time interested in open-ended, leaky magnetic mirror confinement systems using D·^{3}He fuel with added ^{6}Li, he visualized re-injecting the ^{3}He end product from the p·^{6}Li reaction back into the fusion reactor to fuse with deuteron fuel ions and thus regenerate the 14 MeV protons to continue the cycle. Breeding (or chain branching) reactions of the type $d(d,n)^3He$ + 3.3 MeV and $d(d,p)t$ + 4.0 MeV would generate both of the energy-rich t and ^{3}He in such a ^{6}Li·D fueled fusion reactor. However, Post limited his considerations to leaky or open-ended magnetic mirror confinement configurations and presumed very efficient reinjection of the t and ^{3}He.

In 1963, completely unaware of these various nuclear fusion chain reaction prospects, I discovered a whole plethora of charged particle fusion chain reactions[6]. However, I could find no way at that time to overcome the stopping power losses of the energetic chain centers on the colder electrons in order to maintain a meaningful burning temperature for ^{6}Li or ^{6}LiD fueled plasmas[7]. More recently, the prospects for overcoming the stopping power losses have brightened as the result of the development of electron cyclotron heating techniques which could lead to a sub-critical fusion chain reactor. In addition, the principle of closing the ends of a leaky magnetic mirror has been experimentally demonstrated. Thus, it may be possible in the near future to develop controlled, but sub-critical, fusion chain reactors using ^{6}Li or ^{6}LiD fuels and in which some of the "ashes" behave as active chain centers at MeV energies. Net fusion power producers, if they ever become feasible, would involve major engineering developments and efficient microwave reflecting walls as well as high β plasmas. Ignition of such devices would require powerful d.c. accelerators while the understanding and design of such reactors will require more accurate charged-particle nuclear cross sections covering a wide range of energies up to about 20 MeV. More than 50 reactions are involved among the various light isotopes up to at least beryllium.

In late 1963, I discovered the first of many nuclear fusion chain reactions which involve medium heavy element fuels like C, N, O, and Ne reacting with active chain centers such as energetic n, p, d, t, ^{3}He, and α particles[8]. It appears that these latter nuclear chain reactions can occur only in super-dense media like pre-supernovae stars where the degenerate electron gas has a grossly reduced stopping power for the fast ions.

My presentation today deals with prospective applications of nuclear fusion chain reactions in physics and astrophysics. In the physics case of controlled fusion reactors, the basic reactions are primarily of the charged particle type inasmuch as neutrons do not appear to be conservable in a magnetically confined plasma. The escaping neutrons can be used for energy or tritium multiplication in an external Na or Li blanket. In the astrophysics case of exploding dense stars, neutrons will play a very important role as one of the active chain centers in the fast nuclear processes taking place in these very high temperature systems. These latter nuclear fusion chain reactions will supplement the thermal or thermonuclear reactions originally proposed by Hoyle and Fowler for supernovae explosions[9].

2. NUCLEAR FUSION CHAIN REACTION PROSPECTS IN PHYSICS

It is too early to claim that charged particle nuclear fusion chain reactions may play a decisive role in controlled fusion reactors, but the prospects are much brighter now than in 1961 when Post first suggested charged particle fusion chain reactions. Closed magnetic confinement devices would be required such as stoppered magnetic mirrors (closed by large internal plasma currents), bumpy tori (which are multiple magnetic mirrors connected toroidally), or tori of the Tokamak or stellarator varieties. Magnetic bremsstrahlung at the high electron temperatures would have to be grossly reduced by utilizing large reactor sizes, by operating with plasmas having a high plasma beta ($\beta = 8\pi nkT/B^2$), and by providing efficiently reflected microwave and/or infrared radiation. The magnetic bremsstrahlung or synchrotron radiation power is reduced proportional to (1-R) and (1-β), respectively. $R > 0.99$ may be expected for clean metallic walls in the infrared and microwave region and $\beta > 0.5$ has been actually obtained in a number of plasma experiments.

Figure 4 illustrates some of the major developments which have occurred in CTR magnetic mirror research. A mini breakthrough in energetic ion trapping in magnetic mirrors has been marginally attained at Culham on the PHOENIX II device and more definitively established at Lawrence Livermore Laboratory on the Baseball II device. At Livermore large scale trapping (30 fold increase over the Lorentz or $v \times B$ trapping by dissociation of excited hydrogen atoms) has been observed for the injected neutrals by the already trapped plasma. A very important process called exponential build-up of the proton density over the charge exchange losses has been conclusively demonstrated at Livermore although it eventually dies out at a density approaching 10^{10} cm^{-3}[10]. The Fermi fission feasibility experiment of 1942 demonstrated an exponential growth process for the build-up of neutrons in a fission experiment. The plasma exponential process for trapping of protons under a more optimum choice of injection energy would permit the large scale exponential build-up of the plasma into the density range of real fusion interest[11]. In various magnetic mirror devices, such as the original Soviet OGRA and the American DCX facilities, the ion "temperature" and confinement times were of thermonuclear interest, but the density was

I.	Plasma Trapping and Incipient Exponential Build-Up of Density (Culham, Livermore)	E
II.	Scaling Laws for Scatter-Dominated Mirrors ($n\tau \propto E^{\frac{3}{2}}$) (Livermore, Culham, Oak Ridge)	T
III.	Hot Electron Plasmas by ECH ($T_e \geq 1$ MeV) (Oak Ridge)	E
IV.	Exponential Build-Up of Density at MeV Energies with ECH (Oak Ridge)	T
V.	Closed Magnetic Mirror (Livermore, Cornell)	E
VI.	MeV Energies for Fusion Reaction Products (Culham, Livermore)	T
VII.	Fusion Chain Reactions (^{6}Li or ^{6}LiD Fuel) (Oak Ridge)	T

FIG.4. Major developments in controlled fusion research on magnetic mirror systems. E = Experimental; T = Theoretical.

orders of magnitude too low. Now, a realistic exponential scaling law has been demonstrated at Culham and especially at Livermore somewhat analogous to the critical exponential test of Fermi for fission. If instabilities do not limit the build-up process a large scale exponential test for fusion may be demonstrated with multi-MeV injection[11].

The scaling law for the density × confinement time product, $n\tau$, for scatter-dominated losses of energetic ions in magnetic mirror machines indicates that the crucial $n\tau$ number increases as the 3/2 power of the energy of the injected ions[12]. Thus, at 1 MeV injection energy for the ions, $n\tau$ is a factor of 1000 times more favorable than at 10 keV injection energy. The electrons must also be raised to very high energy in order to reduce their stopping power effects—however, this has been satisfactorily accomplished at Oak Ridge by resonance and upper off-resonance electron cyclotron heating of the electrons to MeV energies in devices such as the ELMO magnetic mirror facility[13].

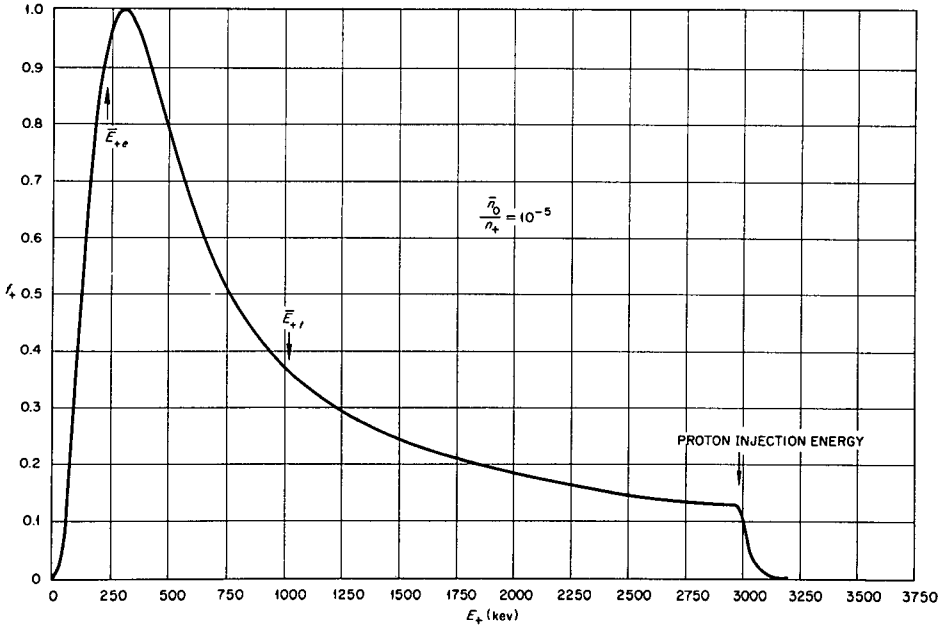
The possibility of the large scale exponential build-up of plasmas to high densities at MeV energies and for long confinement times was outlined by me in 1970 and 1971[11] and involved injection of 2 mA of 4 MeV H_2^+ ions into a simple magnetic mirror. Closure of the magnetic mirror by the self-current of the trapped protons would require a 10-20 mA, 4 MeV H_2^+ beam to produce a sufficiently large circulating ion current to reverse the magnetic field on the axis—a small scale version of the proton E-layer of the late N. C. Christofilos of Livermore.

The experimental prospect for closing a magnetic mirror by a large internal current of relativistic electrons—the so-called E-layer—has been studied at Livermore and eventually was successfully demonstrated in 1971 and 1972 at Cornell University[14]. Such closed magnetic mirror devices might be termed "orthogonal Tokamaks" since the plasma

current is at right angles to the applied magnetic field whereas in the Tokamak device the plasma current is parallel to the magnetic guide field.

Reaction kinetics calculations of fusioning plasmas fueled with D·T and D·^{3}He have been carried out at Culham and Livermore by means of Fokker-Planck equations which include the slowing down and energy diffusion of the charged reaction products, p and α, in the reacting plasma[15,16]. These calculations revealed mean ion energies greater than 1 MeV for both the protons and the alpha particles even without supplemental microwave heating of the electrons. However, the calculations omitted radiation losses so that electron cyclotron heating would probably be required in any small scale laboratory device to offset the synchrotron losses and maintain the protons and alphas at MeV energies.

Nuclear fusion chain reactions require such suprathermal or MeV particle energies to propagate the fusion chain reactions in fuels like ^{6}Li or ^{6}LiD. Since even in leaky magnetic mirrors the fusion "ashes" can have multi-MeV energies, the overall prospect for viable nuclear fusion chain reactions in closed magnetic configurations looks extremely favorable. Initially such systems would be sub-critical because of the synchrotron losses, but large systems may prove to have a net power.



Distribution Function for Protons in DCX Plasma for 6- Mev H_2^+ Injection.

FIG.5. Fokker-Planck calculations of energy distribution function for 3 MeV protons injected into a 2/1 magnetic mirror with $T_e \sim 30$ keV. $\bar{E}_{+e}$ = average energy of escaping protons, $\bar{E}_{+t}$ = average energy of trapped protons. Obtained by Mozelle Rankin in 1964 at ORNL.

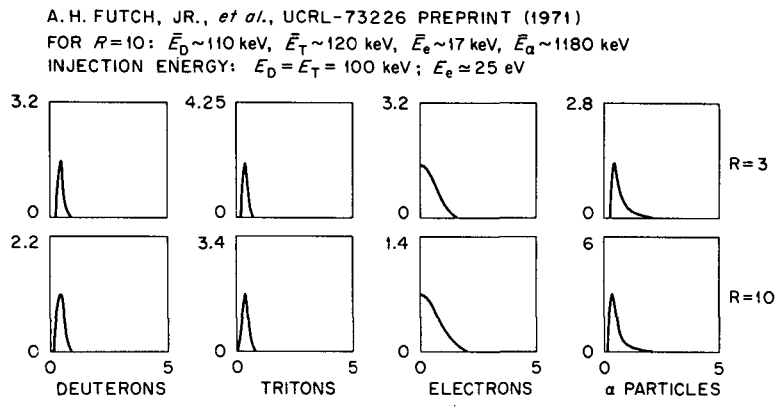


FIG.6. Fokker-Planck calculations of particle velocity distribution functions in magnetic mirrors for d-t reactions [18]. Abscissa scale for electrons to be multiplied by 6 x 10^9 and for ions by 6 x 10^8 cm/s.

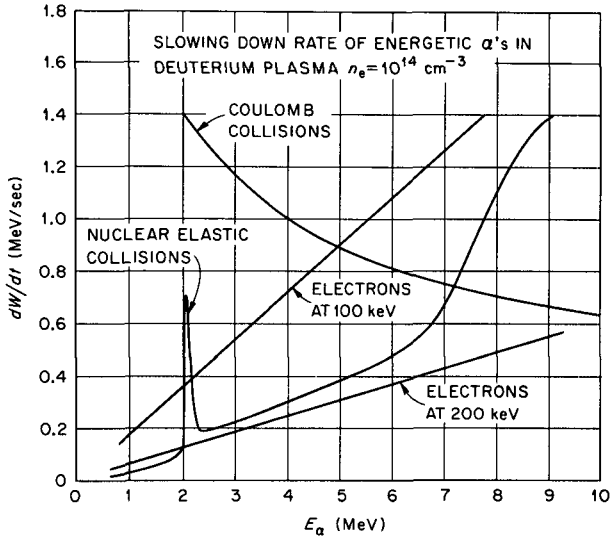


FIG.7. Energy gain and loss rates for alpha particles via Coulomb collisions and nuclear elastic collisions with deuterons and electrons.

Figure 5 shows the calculated energy distribution function for a 3 MeV proton slowing down in a mirror confined plasma having $T_e \sim 30$ keV. Figure 6 illustrates Livermore calculations on the particle velocity distribution functions for different ion and electron species in a fusioning D·T plasma confined in a magnetic mirror having a mirror ratio R (= B_{max}/B_0) of 3 and 10, respectively[16]. For the R = 10 case the alpha products slow down and diffuse to an average energy of 1.2 MeV whereas the D and T fuel ions move from their injected energy of 100 keV

to a mean energy slightly higher than 100 keV. In contrast, the electrons are essentially cold, being injected initially into the plasma at the same velocity as the fuel ions. The electrons eventually attain an electron temperature of only about 17 keV. Despite the expected strong stopping power of the electrons for the 100 keV deuterons and tritons, the average energy of the latter increases slightly, primarily as a result of collisional heating by the initially 3.5 MeV alphas.

Figure 7 illustrates that alpha heating of the deuterons would be even more pronounced if one included nuclear elastic scattering effects. Here, the nuclear elastic collisional heating rate of deuterons by alpha particles is based on the excellent paper by Devaney and Stein of Los Alamos Scientific Laboratory for some light element nuclear elastic scattering collisions[17]. More elastic and inelastic collision cross sections are needed for the elements Li through at least neon.

Most astrophysical and physics calculations of nuclear reaction processes generally assume all particles (fuel, electrons, and reaction products) have the same temperature, i.e., that local thermodynamic equilibrium (LTE) prevails. Spectroscopic and other diagnostic studies of dynamic plasma systems in the laboratory reveal markedly different "temperatures" for each specie. A good example of a physical system having a gross departure from temperature equilibrium is the fast breeder reactor in which the neutrons will have a so-called "temperature" of order 3×10^9°K whereas the fuel, sodium heat exchanger, and associated structures will be at about 1000°K. The energetic, magnetically confined, long carbon arc plasma at Oak Ridge revealed ions at 5×10^6°K whereas the electrons were at only about 4×10^4°K[18]. The Burnout series of experiments at Oak Ridge revealed kilo-electron volt ions immersed in electrons at only a few eV temperature[19]. The DCX experiments at ORNL had a kinetic "temperature" for the ions in excess of 3×10^9°K whereas its electrons were at 10^7°K or less[20]. Similar non-LTE results have been observed in numerous plasma experiments in the CTR world community.

A nuclear fusioning plasma will probably have a fuel mix at moderate fuel ion and electron temperatures ($\sim 10^9$°K), but the fusion reaction products which enter the system at multi-MeV energies, may well have an ion "temperature" of order 10^{10}°K or more. This is beautifully illustrated by the reaction kinetics calculations at the Culham and Lawrence Livermore Laboratories where the α product from the d·t reaction has a mean energy of about 1.2 MeV, whereas protons from the d·^{3}He reaction have a mean energy of $\sim$ 5 MeV[15,16]. Thus, these very energetic charged particles from the fusion reactions may react very favorably with added nuclear fuels such as ^{6}Li or ^{6}LiD to initiate nuclear fusion chain reactions. Subsequent generations of active chain centers can, in principle, maintain the criticality condition or reproduction factor, k, under appropriate conditions of fuel feed, ash exhaust, and controlled electron temperature. Whether such reactions can proceed in moderate density, magnetically confined plasmas having large radiation losses remains for future experiments to determine. It is quite possible that the first nuclear fusion reactors will be sub-critical fusion chain reactors in which the electrons are sustained at MeV temperatures by electron cyclotron heating (ECH). Such sub-critical fusion reactors may also be of significant value as pilot plant fusion reactors having greater application for nuclear reaction kinetic analyses than the most complicated electronic computer—although the diagnostic readouts will be difficult to evaluate.

In Plasma

$$^{7}Li + t \rightarrow \begin{cases} n + {}^{9}Be + 10.4\ MeV \\ 2n + 2\alpha + 8.8\ MeV \end{cases}$$

In Blanket

$$\begin{cases} n_{fast} + {}^{7}Li \rightarrow n + t + \alpha - 2.5\ MeV \\ n_{fast} + {}^{238}U \rightarrow \text{Fission Products} + 2.5\ n + 200\ MeV \\ n + {}^{238}U \rightarrow {}^{239}Pu + 2e + 5.8\ MeV \\ n + {}^{6}Li \rightarrow t + \alpha + 4.8\ MeV \end{cases}$$

*C.J.H. Watson, Appendix III, EUR-CEA-FC-628-AG (1971).

FIG. 8. Neutron-multiplying fusion reactions with ^{7}Li fuel and tritons in a hot plasma and triton- or neutron-multiplying reactions in the blanket [21].

BREEDING REACTIONS (THERMAL REACTIONS GIVING CHAIN BRANCHING)

$$d + d \rightarrow n + {}^{3}He + 3.3\ MeV$$
$$d + d \rightarrow p + t + 4.0\ MeV$$
$$d + {}^{6}Li \rightarrow 2\alpha + 22.4\ MeV$$
$$d + {}^{6}Li \rightarrow p + {}^{7}Li + 5.0\ MeV$$
$$d + {}^{6}Li \rightarrow p + t + \alpha + 2.6\ MeV$$
$$d + {}^{6}Li \rightarrow n + {}^{7}Be + 3.4\ MeV$$
$$d + {}^{6}Li \rightarrow n + {}^{3}He + \alpha + 1.8\ MeV$$

FIG. 9. Thermonuclear reactions in ^{6}LiD-fuelled plasmas leading to branched chains.

Figure 8 illustrates a possible multiplying fusion chain reaction proposed by Watson of the Culham Laboratory[21]. Here, a single triton reacting with ^{7}Li can generate as many as 2 neutrons which can then react further in a lithium blanket to breed tritons in excess. The new tritons must eventually be reintroduced into the reacting plasma to replace the burn-up of the original tritons in the plasma. Blow and Watson have calculated that a T - ^{7}Li - ^{238}U system can produce a very large energy multiplication—about 100 MeV per fusion event or as much as 200 MeV per fusion event if Pu breeding is taken into account[21]. Such a complex system would, like the fission reactor, be energy rich rather than neutron rich; however, there are major complications in such a system (fission products, reinjection problems, tritium inventory, sustainment, etc.).

As mentioned earlier, the process of nuclear fusion chain reactions involves both chain and thermal (or thermonuclear) reactions and thus should more precisely be called a chain-thermal or chain-thermonuclear reaction. Figure 9 shows several thermonuclear reactions which give chain branching in a ^{6}LiD fueled plasma, i.e., the reactions are breeding reactions for t and ^{3}He as well as for energetic protons and alphas.

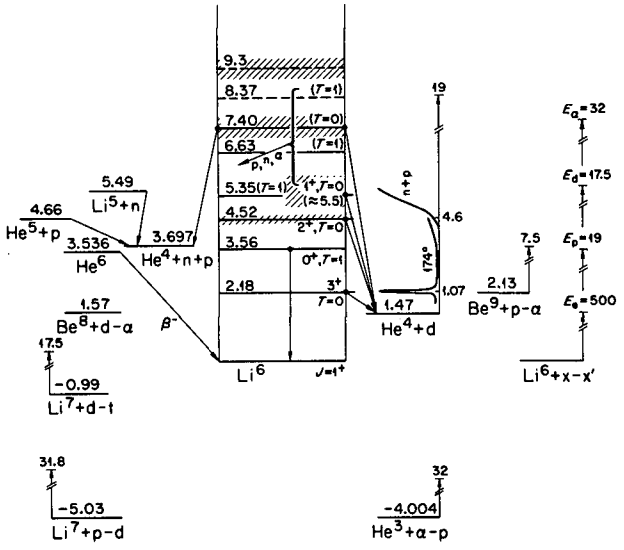


FIG. 10. Nuclear mass-energy level diagram of ^{6}Li showing the important 2.18-MeV state which dissociates into energetic d and alpha particles [22].

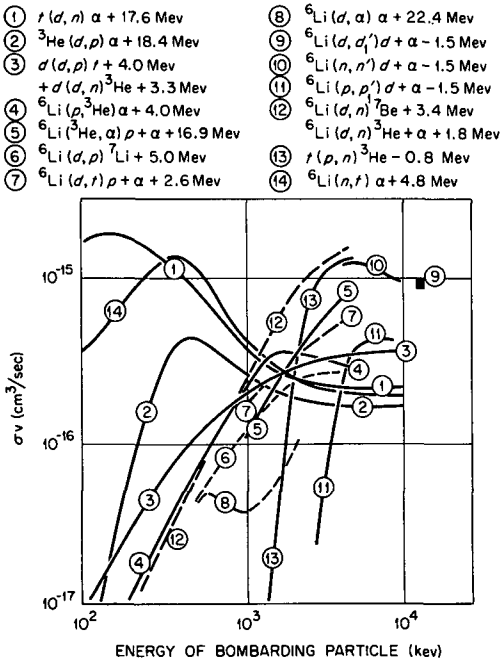


FIG. 11. Various reaction rate parameters, σv, for several important light-element reactions as a function of projectile energy. Data only approximate in most cases.

The energetic protons, ^3He, t, and alpha particles, will all end up in the reaction mix as suprathermal ions capable of further reactions with deuterons or ^6Li fuel nuclei. In the case of pure deuterium fueled plasmas the reaction can be catalyzed by the t and ^3He ashes for only one more generation. In the case of ^6Li or ^6LiD fuel, many further reaction generations may be expected—the reactions can be auto-catalytic, i.e., lead automatically to the next reaction. The use of ^6LiD fuel will permit true auto-catalytic reactions to occur so long as new fuel is added and cold ashes removed.

Even some of the very energetic alphas and protons can regenerate new fast deuterons from ^6Li dissociation. Figure 10 shows the nuclear mass-energy level diagram for ^6Li[22]. The low-lying 2.2 MeV state is extremely important for converting very fast alphas or protons to colder ashes plus a fast deuteron when the excited 2.2 MeV state decays to d + α. The 9 MeV alphas originating from the $^6Li(^3He,\alpha)^5Li \rightarrow p + \alpha$ + 16.9 MeV and 11 MeV alphas from $^6Li(d,\alpha)\alpha$ + 22.4 MeV probably have a large cross section for excitation of this state. The $^6Li(p,p')^6Li^*$ - 2.2 MeV reaction has a fairly flat σv from 10 MeV down to almost 3 MeV proton energy as shown in Fig. 11[6]. Figure 11 illustrates the importance of the reactions (1) and (14) to Jetter's proposed chain reaction cycle.

Note the five important d·^6Li reactions, (6), (7), (8), and also (12) for which there are two reaction branches possible. The reactivity product, $\sigma v Q_+$, where Q_+ is the energy release in charged particles, is about four times larger for d·^6Li than for the two d·d reaction channels. This fact leads to a value of $n\tau$ of about 1.8×10^{15} sec/cm^3 for a proposed d·^6Li fueled fusion reactor as compared to a value of 6×10^{15} needed for the d·d system[23]. The five d·^6Li reactions have the additional advantage that they can initiate nuclear fusion chain reactions whereas the d·d breeding cycle, in which the tritium and ^3He

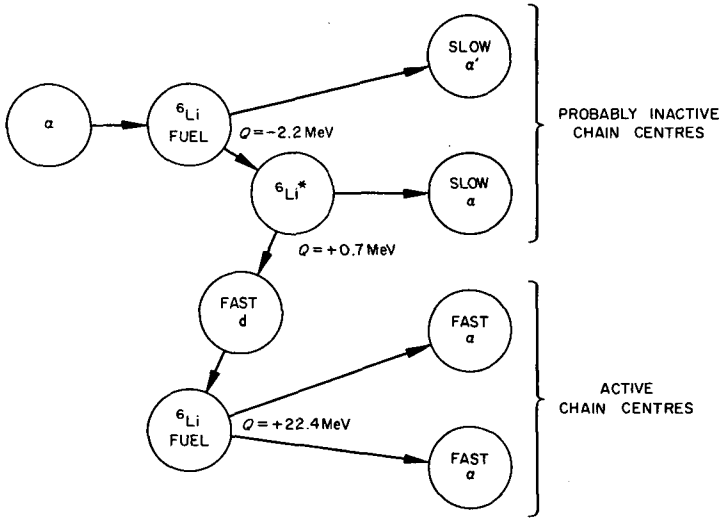


FIG. 12. Fusion chain reaction initiated by fast alphas on ^6Li-fuel nuclei. The $^6Li^*$-excited state emits a 0.5-MeV deuteron.

$$\begin{cases} p + {}^{6}\mathrm{Li} \rightarrow {}^{3}\mathrm{He} + \alpha + 4.0\ \mathrm{MeV} \\ {}^{3}\mathrm{He} + d \rightarrow p + \alpha + 18.4\ \mathrm{MeV} \end{cases}$$

$$\begin{cases} p + {}^{6}\mathrm{Li} \rightarrow {}^{3}\mathrm{He} + \alpha + 4.0\ \mathrm{MeV} \\ {}^{3}\mathrm{He} + {}^{6}\mathrm{Li} \rightarrow p + 2\alpha + 16.9\ \mathrm{MeV} \end{cases}$$

$$\begin{cases} p + {}^{6}\mathrm{Li} \rightarrow {}^{3}\mathrm{He} + \alpha + 4.0\ \mathrm{MeV} \\ \alpha + {}^{6}\mathrm{Li} \rightarrow p + {}^{9}\mathrm{Be} - 2.1\ \mathrm{MeV} \end{cases}$$

$$\begin{cases} d + {}^{6}\mathrm{Li} \rightarrow t + p + \alpha + 2.6\ \mathrm{MeV} \\ t + {}^{6}\mathrm{Li} \rightarrow d + {}^{7}\mathrm{Li} + 1.0\ \mathrm{MeV} \end{cases}$$

$$\begin{cases} d + {}^{6}\mathrm{Li} \rightarrow {}^{3}\mathrm{He} + n + \alpha + 1.8\ \mathrm{MeV} \\ {}^{3}\mathrm{He} + {}^{6}\mathrm{Li} \rightarrow d + {}^{7}\mathrm{Be} + 0.1\ \mathrm{MeV} \end{cases}$$

FIG. 13. Several charged-particle fusion chain reactions involving ^{6}Li- or ^{6}LiD-fuel.

can react only in one second generation reaction, will revert back to a pure thermal or thermonuclear cycle after the second generation.

Reactions (9) and (10) illustrate the importance of converting very energetic particles to reactive deuterons. The analogous $^{6}\mathrm{Li}(\alpha,\alpha')^{6}\mathrm{Li}^{*} \rightarrow d + \alpha - 1.5$ MeV has a σv of almost 2×10^{-15} cm^3/sec at 31 MeV but needs to be measured accurately down to about 3 MeV. In fact all of these reaction rate parameters, σv, are poorly known despite the seeming authority of this prepared figure. There are many lacunae in the data, many cross sections have been measured only once; also, differential cross sections are frequently measured for only one angle and gross uncertainties have been introduced in converting the differential cross sections to absolute cross sections.

Figure 12 illustrates a multiplying chain for suprathermal alpha particles with ^{6}Li nuclei. Here, one fast alpha reacts with the first ^{6}Li nucleus to produce a fast deuteron ($\sim$ 0.5 MeV not including the center of mass energy contribution) which can then react with a second ^{6}Li fuel nucleus to give two fast alpha particles each with an energy above 11 MeV; thus, this two step reaction has a theoretical multiplication factor of 2.

Figure 13 illustrates five charged particle propagation chain reactions with ^{6}Li or ^{6}LiD fuel. In the first case, ^{3}He is produced by the reaction of fast protons with ^{6}Li and then reacts with d to regenerate a 14 MeV proton which can continue to propagate the chain cycle. In the second reaction cycle the fast ^{3}He can react with a ^{6}Li fuel nucleus to regenerate a fast proton as well as two energetic alphas. The energy spectrum of the alphas in this second reaction cycle extends up to about 9 MeV. It should be noted that the maximum multiplication factor in the first reaction cycle of Fig. 13 is unity so that any chain center losses would have to be replenished via d·d or d·^{6}Li chain branching thermonuclear reactions (Fig. 9).

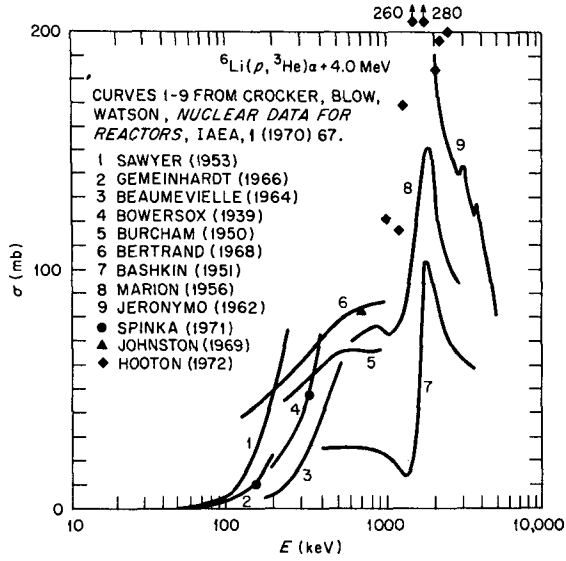


FIG. 14. Various measurements of the cross-section of the ^{6}Li (p, ^{3}He) α + 4.0 MeV reaction as a function of energy [24, 25].

To illustrate how a very simple reaction such as $^6Li(p,^3He)\alpha$ + 4.0 MeV has widely varying cross section measurements, consider Fig. 14 which is taken from Crocker, Blow and Watson of the Culham and Harwell Laboratories[24]. More recent data of Spinka, Johnston, Hooton, et al[25] is shown. This p·^{6}Li reaction could be an excellent standard for the much more difficult 5 channel d·^{6}Li reactions as well as others involving ^{6}Li, but it needs to be measured accurately from 100 keV to at least 15 or 20 MeV.

It appears that considerably more nuclear cross section measurements—both theoretical and experimental—are required to evaluate more thoroughly the ultimate prospects of nuclear fusion chain reactions in controlled fusion applications.

3. NUCLEAR FUSION CHAIN REACTION PROSPECTS IN ASTROPHYSICS

Nuclear fusion chain reactions in astrophysical systems do not appear to have received the degree of study they probably merit. Present day astrophysical calculations, especially for rapid phenomena such as occur in supernovae, presume local thermodynamic equilibrium (LTE) among all species and determine $\langle\sigma v\rangle$ vs. T for a large class of possible reactions. This type of LTE calculation omits or severely reduces the prospect for certain high energy reactions for which σv may be unusually large. This is especially true in the case of endothermic reactions having a high energy threshold. I first proposed the possibility of nuclear fusion chain reactions involving such fuels and suprathermal chain centers in 1964 in an ORNL-Status and Progress Report[8]. The ORNL report stated briefly, "Other light isotopes also possess chain-reacting capabilities, at least in principle. It is quite possible that

THERMONUCLEAR REACTIONS LEADING TO CHAIN BRANCHING

$$^{12}C + {}^{12}C \rightarrow \begin{cases} p + {}^{23}Na + 2.2\ MeV & E_p > 2.1\ MeV \\ \alpha + {}^{20}Ne + 4.6\ MeV & E_\alpha > 3.6\ MeV \end{cases}$$

$$^{14}N + {}^{14}N \rightarrow \begin{cases} n + {}^{27}Si + 9.1\ MeV & E_n > 8.8\ MeV \\ p + {}^{27}Al + 15.6\ MeV & E_p > 15.0\ MeV \\ \alpha + {}^{24}Mg + 17.3\ MeV & E_\alpha > 14.8\ MeV \end{cases}$$

$$^{16}O + {}^{16}O \rightarrow \begin{cases} n + {}^{31}S + 1.6\ MeV & E_n > 1.5\ MeV \\ p + {}^{31}P + 7.7\ MeV & E_p > 7.5\ MeV \\ \alpha + {}^{28}Si + 9.7\ MeV & E_\alpha > 8.5\ MeV \end{cases}$$

$$^{20}Ne + {}^{20}Ne \rightarrow \begin{cases} n + {}^{39}Ca + 6.0\ MeV & E_n > 5.8\ MeV \\ p + {}^{39}K + 12.4\ MeV & E_p > 12.1\ MeV \\ \alpha + {}^{36}Ar + 13.7\ MeV & E_\alpha > 12.3\ MeV \end{cases}$$

plus numerous isotope mixtures such as $^{12}C + {}^{14}N$, etc.

FIG.15. Thermonuclear reactions among C, N, O, or Ne nuclear fuels indicating energy of the light reaction product (centre-of-mass energy of the reacting nuclei would increase E).

the various light-element thermonuclear chain reactions can occur only in optically thick media, such as are found in astrophysical systems, and possibly may contribute to the generation of novas and supernovas. In such cases, neutrons would play a more important role than in laboratory plasmas." Since that time a fuller development of the prospect for nuclear fusion chain reactions in astrophysical systems has evolved and is outlined here.

Figure 15 depicts the production of suprathermal chain centers by thermonuclear reactions among the fuels C, N, O, and Ne. Each reaction gives rise to a light product, n, p, or α, which will carry away the bulk of the reaction Q from center of mass considerations. Thus for a nitrogen burning system the neutron enters the reaction mix at 9 MeV or more, the proton at 15 MeV or more, and the α particle at 15 MeV or more depending on the Q and the incremental amount of energy available in the center of mass of the colliding nuclei. In fast reacting systems such as supernovae, these energetic particles will be suprathermal chain centers as they slow down on the colder electrons and on the nuclear fuel materials via Coulomb and nuclear elastic and inelastic collision processes.

In very dense systems such as supernovae, the plasma is usually treated as a degenerate electron gas (see Fermi and Teller[26] and Gryzinski[27]) and the stopping power of the cold electrons is very greatly reduced. This effect can be illustrated by Fig. 16 which gives the Coulomb logarithm, $\ell n\, \Lambda_{ie}$, for a 1 MeV proton slowing down in a non-degenerate plasma of varying electron density and $T_e \sim 10^9$ °K. Note that at supernovae densities the stopping power, which is proportional to the Coulomb logarithm, $\ell n\, \Lambda_{ie}$, approaches zero asymptotically whereas the stopping power for the faster electrons on the protons as measured

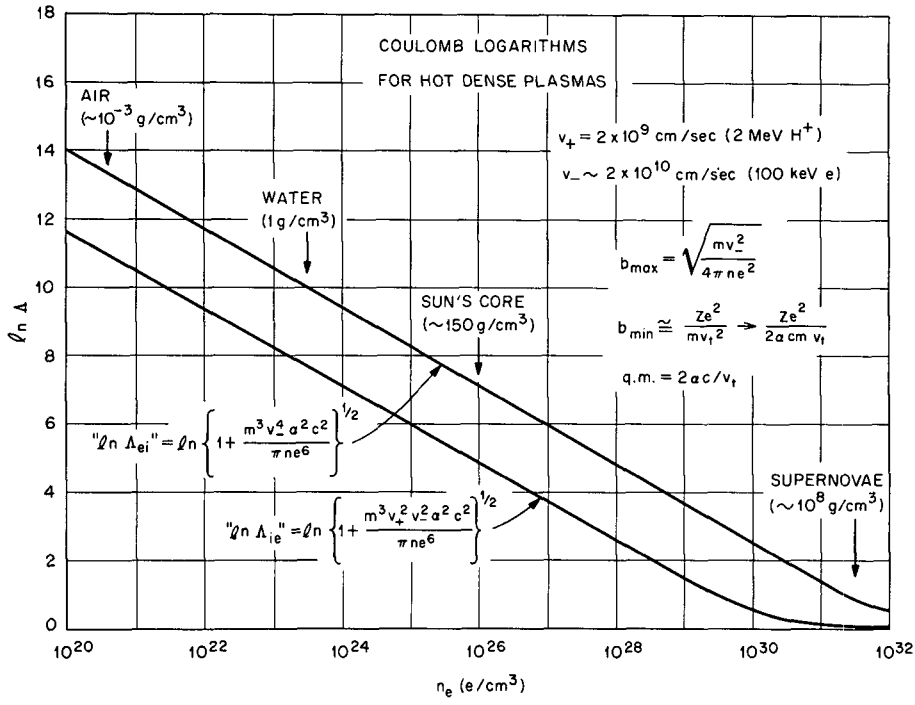


FIG.16. Coulomb logarithm for 1-MeV proton slowing down on 100-keV electron gas at various densities ($\ln \Lambda_{ie}$) and for 100-keV electron slowing down on 1 MeV protons ($\ln \Lambda_{ei}$).

by $\ell n\, \Lambda_{ei}$ is significantly larger. The net effect of this is to permit the energetic proton to remain suprathermal much longer than in more ordinary lower density plasmas such as those having densities near that of air, water, or the sun's core. In fact, the slowing down time becomes independent of density whereas the nuclear reaction time continues to decrease as the density increases. Thus, in very dense systems such as pre-supernovae stars, the suprathermal fusion reaction products will slow down primarily on the fuel nuclei keeping the nuclear or ion "temperature" somewhat higher than that of the electrons. The neutrons are of course uncharged and do not suffer Coulomb slowing down collisions. A local severe fluctuation in the nuclear reaction rate can produce a strong compressional shock wave in a dense star which may shift the electrons from non-degenerate to degenerate and the explosion may then propagate through a large fraction of the whole system.

Figure 17 illustrates multiplying chain reactions in nitrogen fuel. The chains are now more complex than those involving ^{6}Li or ^{7}Li but show that in principle these multiplying chain reactions may supplement the ordinary thermonuclear burning of nitrogen. The overall process in cycle A utilizes two initially fast alpha particles and converts one nitrogen nucleus by (α,p) and (α,n) reactions up to neon whereas a second nitrogen nucleus is reduced to alpha particles by (p,α) and

A.
$$^{14}N(\alpha,p)^{17}O - 1.20\ MeV \qquad ^{14}N(n,\alpha)^{11}B - 0.15\ MeV$$
$$^{17}O(\alpha,n)^{20}Ne + 0.61\ MeV \qquad ^{11}B(p,\alpha)2\alpha + 8.67\ MeV$$
$$2^{14}N + 2\alpha \rightarrow {}^{20}Ne + 4\alpha + 7.93\ MeV$$

B.
$$^{14}N(\alpha,\alpha')^{16}O - 3.12\ MeV$$
$$^{14}N(\alpha',\alpha)^{12}C^{*} \rightarrow 3\alpha + 6.29\ MeV$$
$$2^{14}N + \alpha \rightarrow {}^{16}O + 4\alpha + 3.17\ MeV$$

C.
$$^{14}N(\alpha,\alpha')^{12}C + d - 10.27\ MeV$$
$$^{14}N(\alpha',\alpha)^{12}C + 13.58\ MeV$$
$$2^{14}N + \alpha \rightarrow 2^{12}C + 2\alpha + 3.31\ MeV$$

Possible Multiplying Nuclear Fusion Chain Reaction Sequences in Burning Nitrogen in Supernovae.

FIG. 17. Multiplying nuclear-fusion chain reactions in nitrogen.

(n,α) reactions. There is a net energy release of 7.93 MeV and a multiplication factor of two since four energetic alphas are produced in the complete chain cycle.

Figure 18 shows several propagation chains which are possible in nitrogen fuel. Here, the theoretical reproduction factor is unity. The loss of a suprathermal chain center either due to too much slowing down or by a nonproductive neutron capture reaction will terminate or break such propagation chains. In this case new neutron, proton, or alpha chain centers must be produced by thermonuclear reactions between two nitrogen fuel nuclei as depicted earlier in Fig. 15. It should be noted that nitrogen is probably not a major constituent in pre-supernovae stars.

Fusion chain reaction possibilities have been discovered for fuel elements at least up to silicon. The most abundant elements in pre-supernovae stars were thought to be C, O, and Ne[9]. One of the most interesting cases is that of chain reaction burning of an oxygen and a neon nucleus as shown in Fig. 19. Here, in the closed chain, four alpha particles can burn neon up to ^{32}S and produce protons and neutrons which subsequently can burn an oxygen nucleus down to alpha particles by (p,α) and (n,α) reactions with a very large energy release. It is of interest to note that the elements up to sulphur are quite prominent in the spectra of old supernovae such as the Crab Nebula. In young supernovae the expansion velocities are much too high to render valid interpretation of the spectroscopic data to ascertain the most prominent elements. It appears that helium, oxygen, carbon, and possibly iron and

A.
$$^{14}N\ (\alpha,d)\ ^{16}O - 3.12\ MeV$$
$$^{14}N\ (d,\alpha)\ ^{12}C + 13.58\ MeV$$
$$\alpha + 2\ ^{14}N \rightarrow \alpha + ^{12}C + ^{16}O + 10.46\ MeV$$

B.
$$^{14}N(p,^{3}He)\ ^{12}C - 4.77\ MeV$$
$$^{14}N(^{3}He,p)\ ^{16}O + 15.23\ MeV$$
$$p + 2\ ^{14}N \rightarrow p + ^{12}C + ^{16}O + 10.46\ MeV$$

C.
$$^{14}N(n,t)\ ^{12}C - 4.01\ MeV$$
$$^{14}N(t,n)\ ^{16}O + 14.47\ MeV$$
$$n + 2\ ^{14}N \rightarrow n + ^{12}C + ^{16}O + 10.46\ MeV$$

FIG.18. Propagation nuclear-fusion chain reactions in nitrogen.

$^{20}Ne(\alpha,p)^{23}Na - 2.38\ MeV$ $\quad$ $^{16}O(n,\alpha)^{13}C - 2.20\ MeV$
$^{23}Na(\alpha,p)^{26}Mg + 1.84\ MeV$ $\quad$ $^{13}C(p,\alpha)^{10}B - 4.08\ MeV$
$^{26}Mg(\alpha,n)^{29}Si + 0.02\ MeV$ $\quad$ $^{10}B(p,\alpha)^{7}Be + 1.15\ MeV$
$^{29}Si(\alpha,n)^{32}S - 1.53\ MeV$ $\quad$ $^{7}Be(n,\alpha)\alpha + 18.99\ MeV$

$$^{20}Ne + ^{16}O + 4\alpha \rightarrow ^{32}S + 5\alpha + 11.81\ MeV$$

$$k = \sqrt[8]{1.25} = 1.03;\ \bar{E}_{max} = 5.91\ MeV$$

Delayed Reaction (^{7}Be Residue)

$$^{7}Be + e \rightarrow ^{7}Li + 0.86\ MeV \quad (53.6\ days)$$
$$^{7}Li(p,\gamma)^{8}Be \rightarrow 2\alpha + 17.34\ MeV$$

FIG.19. Nuclear-fusion chain-reaction burning of oxygen and neon nuclei.

other metals are now being identified[28] in the 1937 Type I supernova, IC-4128, which had a well defined 55 ± 1 day half life for its light decay[29]. In 1950 Lyle Borst suggested this light decay of 55 days was due to ^{7}Be radioactivity[30] which has a half life of 53.61 days[31]. He proposed the very endothermic $\alpha(\alpha,n)^{7}Be - 19.0$ MeV reaction to produce the ^{7}Be in a contracting star. Gryzinski[27] suggested ^{7}Be production by the $\alpha(^{3}He,\gamma)^{7}Be$ reaction and postulated that ionization of the L electron shell would extend the half life of 53.61 days by about 4%, thus making for almost a perfect fit of the long light decay curve after the initial burst of the Type I supernovae.

Figure 19 shows that ^{7}Be can be produced in chain reaction cycles involving Ne and O fusion reactions (also C, Mg or Si and O chain reaction burning can generate ^{7}Be). The ^{7}Be residue would be especially prominent if the alpha particles slow down too fast so that they can not produce sufficient neutrons via the $^{29}Si(\alpha,n)^{32}S$ reaction to destroy the ^{7}Be. For example, early expansion of the supernovae star would increase the stopping power of the alphas by the electrons because the Coulomb logarithm increases significantly as the density of the star decreases (Fig. 16). There are, of course, many other chain reaction combinations that can produce ^{7}Be; e.g., ^{16}O and ^{24}Mg, ^{16}O and ^{28}Si, etc.

The residual ^{7}Be would decay to ^{7}Li with a half life of about 55 days for doubly ionized Be in the core of the star where low energy proton reactions may then occur much more rapidly in the core remnant. For example, the ^{7}Li can be consumed by the highly exothermic reaction $^{7}Li + p \rightarrow {}^{8}Be^{*} \rightarrow 2\alpha + 17.3$ MeV. Thus, a much smaller amount of ^{7}Be would be required in the residual core than the figure of 0.007 of the supernovae mass postulated by Borst[30] to explain the 55 day afterglow of supernovae of Type I.

Figure 20 shows a possible nuclear fusion chain reaction involving two neon nuclei which are burned up and down respectively to α's and ^{32}S. In this case one also sees a multiplication of alpha chain centers and a net energy release for the closed cycle.

$$^{20}Ne(\alpha,p)^{23}Na - 2.38 \text{ MeV} \qquad | \qquad ^{20}Ne(n,\alpha)^{17}O - 0.61 \text{ MeV}$$
$$^{23}Na(\alpha,p)^{26}Mg + 1.84 \text{ MeV} \qquad | \qquad ^{17}O(p,\alpha)^{14}N + 1.20 \text{ MeV}$$
$$^{26}Mg(\alpha,n)^{29}Si + 0.02 \text{ MeV} \qquad | \qquad ^{14}N(n,\alpha)^{11}B - 0.15 \text{ MeV}$$
$$^{29}Si(\alpha,n)^{32}S - 1.53 \text{ MeV} \qquad | \qquad ^{11}B(p,\alpha)2\alpha + 8.67 \text{ MeV}$$

$$2\,^{20}Ne + 4\alpha \rightarrow {}^{32}S + 6\alpha + 7.06 \text{ MeV}$$
$$k = \sqrt[8]{1.5} = 1.05; \quad \bar{E}_{max} = 2.35 \text{ MeV}$$

or

$$2\,^{20}Ne + 3\alpha \rightarrow {}^{32}S + 5\alpha + 7.06 \text{ MeV}$$
$$k = \sqrt[8]{1.667} = 1.07; \quad \bar{E}_{max} = 2.35 \text{ MeV}$$

FIG. 20. Nuclear-fusion chain reaction in neon fuel.

$$^{16}O(\alpha,p)^{19}F - 8.12 \text{ MeV} \qquad | \qquad ^{16}O(n,\alpha)^{13}C - 2.20 \text{ MeV}$$
$$^{19}F(\alpha,p)^{22}Ne + 1.71 \text{ MeV} \qquad | \qquad ^{13}C(n,\alpha)^{10}Be - 3.85 \text{ MeV}$$
$$^{22}Ne(\alpha,n)^{25}Mg - 0.48 \text{ MeV} \qquad | \qquad ^{10}Be(p,\alpha)^{7}Li + 2.57 \text{ MeV}$$
$$^{25}Mg(\alpha,n)^{28}Si + 2.66 \text{ MeV} \qquad | \qquad ^{7}Li(p,\alpha)\alpha + 17.34 \text{ MeV}$$

$$2\,^{16}O + 3\alpha \rightarrow {}^{28}Si + 4\alpha + 9.63 \text{ MeV}$$
$$k = \sqrt[8]{1.333} = 1.04; \quad \bar{E}_{max} = 4.82 \text{ MeV}$$

FIG. 21. Nuclear-fusion chain reaction in oxygen fuel. Note highly endothermic reaction requiring, at least, 8.12 MeV energy in centre-of-mass system, thus preventing early explosion of pure oxygen systems. The theoretical multiplication factor is 1.333 for the complete cycle or an average of 1.04 per generation. $\bar{E}_{max}$ is the maximum propagation energy assuming negligible energy feed to degenerate electrons and complete consumption of fuel.

The various fusion chain reactions with C, N, O, and Ne suggest more direct ways of producing significant abundances of Li, Be, and B whose presence in the heavens is still apparently an astrophysical puzzle. Even a short chain length for the fusion chain reactions would ensure the production of relatively large quantities of these elements.

Figure 21 illustrates a nuclear fusion chain cycle for a hypothetical pure oxygen star. This cycle has a very highly endothermic step requiring a center of mass energy in excess of 8.12 MeV. Thus, oxygen-rich, as well as carbon-rich, stars do not appear to be nearly as favorable a nuclear environment for the production of multiplying chain reactions as do mixtures of oxygen and neon or carbon and neon. It appears that stars rich in carbon, oxygen, and neon or other medium heavy elements are most likely to permit nuclear fusion chain reactions to occur if there is a strong compression of the stellar material and the conditions for the compressional shock wave persist.

4. SUMMARY

Nuclear fusion chain reactions may have a potential impact on controlled fusion reactors and may give a more complete explanation of supernovae processes; however, much more basic research in science and technology will have to be done to elucidate the true role of fusion chain reactions. Of especial importance in this broad ranging research endeavor will be the accurate measurement of many nuclear cross sections involving both neutrons and charged particles as projectiles and fuels at least up to sulphur.

I hope I have stimulated in some small way the nuclear data community to the prospect for new and challenging research needs in the area of light element cross sections for controlled fusion and astrophysical applications. In closing, I paraphrase Louis Rosen, Director of the Los Alamos Meson Physics Facility who said at the National Particle Accelerator Conference in Chicago in 1971, "One barrier to the pursuit of this idea of fusion chain reactions in controlled fusion reactors is the grossly incomplete knowledge of reaction cross sections for light nuclei at low energies"[32]. The same kind of statement might also be made with regard to astrophysical explosions.

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DISCUSSION

G.A. KOLSTAD: This is a question addressed to Mr. Yankov, Mr. Chernilin and Mr. McNally: can you comment on the importance of

more and better nuclear data to the successful development of fusion reactors, i.e. is it (1) trivial, (2) important, or (3) critical?

J.R. McNALLY: The light particle reactions are generally quite well known except for cross-sections above about 1 MeV. There is a recent Batelle Northwest Laboratory report which summarizes the data over the years on these reactions. In the case of ^{6}Li D-fueled fusion reactors, if such are possible, the data are in general very poor and many cross-sections are urgently needed.

W.C. WOLKENHAUER: The Batelle Northwest report that Dr. McNally referred to is BNWL-1604 (1971). Another pertinent report which supplements this information is BNWL-1685 (1972).

G.B. YANKOV: In the matter of neutron cross-sections, it seems to me, in accordance with the priorities recommended for the list of nuclear data requirements for fusion reactors, that the first place belongs to cross-sections and other characteristics of non-elastic interactions of neutrons with the structural materials of the blanket, especially for the neutron energy range bordering on 14 MeV.

Yu.F. CHERNILIN (Chairman): As regards the DT reaction, I think some experimental studies are needed relating to the measurement of nuclear data in the energy region mentioned by Mr. Yankov, because one cannot always trust calculations. Of course, calculations must still be made but nevertheless certain hot-spots should be measured. The most important nuclear data requirement, I think, is in the matter of radiation damage. I fully share the views expressed by Mr. McNally regarding reactions other than the DT reaction.

Section IX

EVALUATED NEUTRON DATA FILES

Chairman

R. JOLY (France)

INTERACTION BETWEEN THE NATIONAL NEUTRON CROSS SECTION CENTER, THE CROSS SECTION EVALUATION WORKING GROUP, AND THE USER COMMUNITY

O. OZER
National Neutron Cross Section Center,
Brookhaven National Laboratory,
Upton, N.Y., United States of America

Abstract

INTERACTION BETWEEN THE NATIONAL NEUTRON CROSS SECTION CENTER, THE CROSS SECTION EVALUATION WORKING GROUP, AND THE USER COMMUNITY.

To a great extent the success of the ENDF/B effort has been the special relationship between the "National Neutron Cross Section Center" (NNCSC), the "Cross Section Evaluation Working Group" (CSEWG), and the user community. The CSEWG was organized by the USAEC in 1966 in order to assess data evaluation needs and capability among its contractors. Since then it has provided a forum for discussion and co-ordination of evaluation activities. Its present objectives are: (a) To formulate the scope and contents of the reference library (ENDF/B); (b) To establish formats and procedures for the use of the library; (c) To perform differential and integral testing of data for consideration in ENDF/B; (d) To provide recommendations on new experiments, evaluation and codes that are needed. The CSEWG consists of a group of representatives from national laboratories, industry and universities. Attendance is by laboratory rather than on an individual basis in order to ensure continuity of participation. The number of participants is purposely kept small. The group is divided into a number of subcommittees, each responsible for a specific task: The subcommittees are (1) Data Testing; (2) Codes and Formats; (3) Normalization and Standards; (4) Shielding; (5) Resonance; (6) Fission Products; (7) Error Quantities; (8) Nuclear Model Codes; (9) Non-Neutron Data. The primary interaction with ENDF/B is provided within CSEWG by the first two subcommittees: The Data Testing Subcommittee co-ordinates the microscopic and integral testing procedures, reviews sets considered for ENDF/B and provides NNCSC with guidelines for preparation of the data file. The Codes and Formats Subcommittee has the responsibility for the specification of formats for ENDF/B data including proposed modifications or additions. Twice a year executive meetings are held at BNL at which NNCSC acts as secretariat providing a chairman, meeting facilities, personnel, preparation and distribution of material and other services. The CSEWG guidelines are followed by NNCSC in the preparation and distribution of the ENDF/B data. The NNCSC acts as an interface to the user community outside of CSEWG by answering requests for data and channelling to CSEWG requests for more evaluation.

The primary base of evaluated nuclear data in use in the United States is produced by the cooperative efforts of two groups: The Cross Section Evaluation Working Group (CSEWG) and the National Neutron Cross Section Center (NNCSC), which administrates the former.

The coordination of tasks and activities by these two groups has resulted in considerable savings of time and effort at laboratories throughout the U.S. due to elimination of duplications and the standardization of reference files and formats. The most outstanding product of the interaction between these two groups, however, is the Evaluated Nuclear Data File-B[1] (ENDF/B), the third version of which has been distributed in February 1972, and which, at present, contains some 238 different sets of data [2]. The file has been generated entirely in agreement with basic cross section measurements, available at the time of the evaluation. Although results from carefully performed integral benchmark calculations were used in some cases to

influence the selection of one experimental set over another, the file is intended to serve as a "user's" file and not merely as a starting point to be thrown away after an "adjusted" set has been created.

The CSEWG-NNCSC interaction with each other, the user community, and the funding agencies, as well as the procedures followed in the generation of the ENDF/B files, will be examined in this paper.

1. OBJECTIVES OF CSEWG

CSEWG was organised in 1966, by the Division of Reaction Development and Technology of the USAEC, for the purpose of assessing data evaluation needs and capabilities among its contractors and similar organizations. Since that time, CSEWG has provided a forum where nuclear data problems can be discussed, solutions proposed, and evaluation and testing activities coordinated.

A prominent acheivement of CSEWG has been the significant cooperation that it was able to bring about among the members of the three groups most directly concerned with nuclear data: the experimentalists, the evaluators, and the users.

At present the objectives of CSEWG are:

a) To formulate the scope and contents of the reference library (ENDF/B).
b) To establish formats and procedures for the use of the library.
c) To perform differential and integral testing of the data for consideration in ENDF/B.
d) To provide recommendations on new experiments, evaluations, or codes that are needed.

2. COMPOSITION AND FUNCTIONS OF CSEWG

CSEWG consists of a group of working level representatives from national laboratories, industrial organizations, and academic institutions that are formally committed towards the achievement of CSEWG objectives. Participation is by laboratory rather than on a individual basis, in order to insure the continuity of activities.

Formal laboratory participation can be established by direct communication from the funding agency or laboratory management to the chairman of CSEWG. (A list of current participating laboratories is shown in Table I.) To this group of evaluation and data testing experts, the NNCSC, by request of the USAEC, acts as a secretariat; providing a chairman, meeting facilities, personnel, preparation and distribution of material, preliminary data, and other services as required. CSEWG, in return, provides the NNCSC with guidelines for the preparation of ENDF/B; and the funding agencies with recommendations for new evaluations, measurements, or codes.

The NNCSC provides the interface to the user community by distributing the finalized and approved data files and their documentation. User requests for new or additional evaluations are received by the NNCSC and channeled to CSEWG through the chairman. (Fig. 1)

3. ADMINISTRATION OF CSEWG

Decisions regarding CSEWG objectives are reached at the CSEWG executive meetings. These meetings are held twice a year at the Brookhaven National Laboratory. Although wide participation in the overall CSEWG effort is

TABLE I. INSTITUTIONS PARTICIPATING IN CSEWG ACTIVITIES

CSEWG - Laboratory

Aerojet Nuclear Co. (ANC)
Argonne National Laboratory (ANL)
Atomic Power Development Assoc. (APDA)
Atomics International (AI)
Bettis Atomic Power Laboratory (BAPL)
Babcock & Wilcox (B&W)
Battelle Northwest (BNW)
Combustion Engineering (CE)
Defense Nuclear Agency (DNA)
General Electric Co. (GE)
Gulf Radiation Technology (GRT)
Hanford Engineering Development La. (HEDL)
Knolls Atomic Power Lab. (KAPL)
Lawrence Livermore Lab. (LLL)
Los Alamos Scientific Lab. (LASL)
National Bureau of Standards (NBS)
Nuclear Fuel Systems (NFS)
Oak Ridge National Lab. (ORNL)
Rensselaer Polytechnic Inst. (RPI)
Savannah River Lab. (SRL)
U. S. Atomic Energy Comm. (USAEC)
Westinghouse Electric Corp. (W)

Other

Atomic Energy of Canada Ltd. (AECL)
Columbia University
Mathematical Application Group, Inc. (MAGI)
Radiation Shielding Information Center (RSIC)
Stanford University

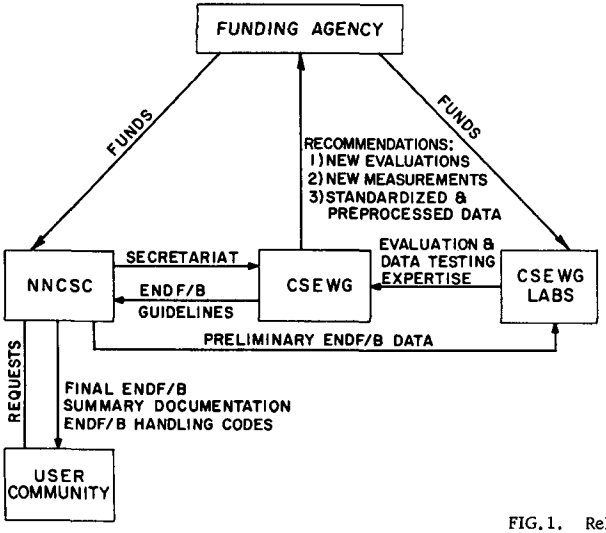

FIG. 1. Relationship between NNCSC-CSEWG, the funding agencies and the user community.

encouraged, attendance at the executive meetings is limited to one or two representatives from each institution in order to assure the representation of each laboratory without sacrificing the advantages of effective face to face discussion offered by a small group.

Much of the ground work necessary in order to reach decisions or CSEWG objectives, is done either by permanent specialized subcommittees or temporary "ad-hoc" committees in between executive meetings. These "ad-hoc" committees are appointed as needed and generally have a wider participation. Experts from outside of CSEWG may be invited. In general, no attempt is made to include all experts on a matter, but only as many as is necessary to reasonably deal with the problem at hand.

At present, CSEWG consists of the following subcommittees:

1) Data Testing
2) Codes and Formats
3) Normalization and Standards
4) Shielding
5) Resonance Region
6) Fission Products
7) Error Quantities
8) Nuclear Model Codes
9) Non Neutron Data.

In carrying out their assigned tasks and responsibilities, the subcommittees must closely interact with one another. From this point of view, the two most central committees are the Data Testing and Codes and Formats Subcommittees. Decisions regarding the balance between the physics and user needs in the areas of fast, thermal, shielding, and fusion applications are made in these two central committees.

4. THE DATA TESTING SUBCOMMITTEE

The primary purpose of the Data Testing Subcommittee is to provide CSEWG with a formal mechanism for the testing of nuclear data. Evaluated data sets being considered for inclusion in ENDF/B must undergo a series of tests. The first level of testing is done at the NNCSC in a more or less mechanical manner. Data sets submitted to the NNCSC are tested for correctness of structure and formats[3] to insure their processability by furhter codes. Next, increasingly rigorous physics consistency checks are conducted [4,5]. Obvious errors, when present, are reported to the original evaluator and corrected. Finally the data set is put into a standard form and a package containing the results of all checking codes, as well as a set of detailed plots is produced and submitted to the Data Testing Subcommittee. Data sets reaching this stage are assigned, by the Subcommittee, to one or more reviewers for a detailed "microscopic" review. The purpose of this review (referred to as Phase I review) is to determine the adequacy of the evaluation with respect to the most current experimental evidence. The Data Testing Subcommittee is responsible for the coordination of all Phase I reviews, as well as the recommendation for the acceptance, modification, or rejection of the preliminary sets. Data sets approved by the Subcommittee become officially part of the ENDF/B Library as category I data.

A further responsibility of the Data Testing Subcommittee is to determine the validity of the ENDF/B data in certain specific areas of application important to the overall national nuclear effort.

This objective is achieved by coordinating the Phase II integral and differential testing of the data in certain well defined "benchmark" situations. This phase of the testing is carried out in close cooperation with other subcommittees specifically responsible for a particular area of application. The major areas of Phase II testing are the following:

a) Fast neutron data
b) Thermal neutron data

c) Dosimetry
d) Shielding
e) Fission products.

The results of the Phase II testing are evaluated and published by the Data Testing Subcommittee in a timely manner, and are used in making recommendations for further data measurements, evaluations, or integral experiments when the need for these has been established.

5. THE CODES AND FORMATS SUBCOMMITTEE

The second objective of CSEWG, the establishment of formats and procedures for the use of the ENDF/B Library, is the responsibility of the Codes and Formats Subcommittee.

This responsibility includes the consideration of all proposed format additions and modifications and consequently it involves close cooperation with other subcommittees. Although any member of CSEWG may propose a format modification, in general, these proposals are made by the subcommittee most directly involved with the type of data that would be affected by the format modification (i.e., proposals affecting the formats and use of the gamma ray production are made by the Shielding Subcommittee).

After considering a proposal, the Codes and Formats Subcommittee may approve it, reject it, or return it to the originator with a request for further documentation or testing. Often suggestions for an alternate approach are made.

An approved format modification generally results in the following CSEWG tasks:

1) Updating of the Formats and Procedures Manual[1] by the NNCSC.
2) Updating of all affected computer programs to recognize the new formats by their responsible programmers.
3) Translation of the old library into the new format by the NNCSC.
4) Requesting of evaluators to conform to the new formats.

Because of the magnitude of the above tasks, proposals most likely to be accepted are those which do not involve extensive modifications in existing computer programs, or the translation of large amounts of data sets.

In addition to the ENDF/B formats and procedures, the Codes and Formats Subcommittee has responsibility for:

a) Improving and maintaining the integrity of codes which directly use the ENDF/B data.
b) Making recommendations for the development of additional processing or data manipulation codes.

The "Tedium Isotopes Test"[7] constitutes a good example of some of the effort going into the maintenance and improvement of existing codes. The "Tedium Isotopes" are a set of fictitious materials created in the ENDF/B format for the purpose of testing processing codes. In general, these isotopes contain a single salient and potentially troublesome characteristic (e.g., single narrow p-wave resonance, a large resonance located at a group boundary, etc.). The intercomparison of results from different codes processing the Tedium materials has resulted in the discovery and correction of numerous theoretical inaccuracies in algorithms or simple coding errors.

In the area of recommendations for new processing codes, the Subcommittee is presently cooperating in the specification of a group library preparation

code capable of producing a single, ENDF/B based, library of group constants that could be used for thermal reactor and shielding applications as well as for fast reactors. The production of such a library is planned for the near future.

6. OTHER SUBCOMMITTEES

In close cooperation with the first two subcommittees are a number of more specialized groups. These are:

6.1. The Normalization and Standards Subcommittee

A number of ENDF/B Cross sections are defined as standards. These include the thermal cross sections of all primary fissile nuclei as well as all cross sections classed as measurement standards by the U.S. Nuclear Data Committee.

The Normalization and Standards Subcommittee has the responsibility for continual review of these cross sections. Any modification of their ENDF/B files must have the concurrence of the Subcommittee. In addition, the Subcommittee makes recommendations on the values and shapes of thermal cross sections and resonance integrals of all ENDF/B materials and is in the process of developing a file of neutron dosimetry cross sections. The Phase I review and Phase II testing of the dosimetry file is being done under the direction of the Data Testing Subcommittee.

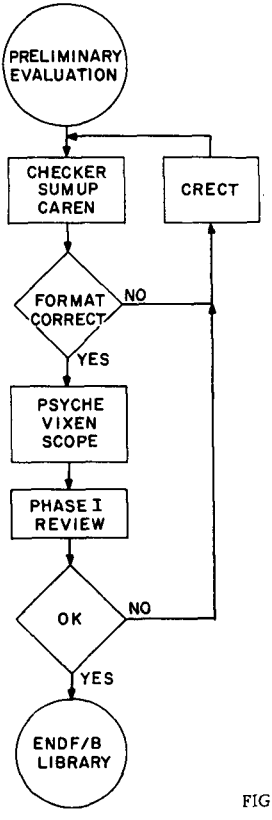

FIG. 2. Checking procedures for ENDF/B evaluations.

6.2. The Shielding Subcommittee

Checking the validity of the shielding data files in ENDF/B, determining their deficiencies, improving and correcting them is the primary responsibility of the Shielding Subcommittee. This is done through Phase I review and Phase II testing in cooperation with and under the direction of, the Data Testing Subcommittee (Fig. 2).

The interaction with the Codes and Formats Subcommittee consists of:

1) Suggesting formats and procedures for the shielding data files.
2) Reviewing the adequacy of codes used in processing of data for shielding calculations, including determination of needs or deficiencies through data testing and "Tedium" like code testing programs, recommending corrective actions.

Because of the nature of the data involved, the Shielding Subcommittee must interact strongly with other data centers in addition to the NNCSC (e.g., the Radiation Shielding Information Center at Oak Ridge National Laboratory, and the Photonuclear Data Center at the National Bureau of Standards) and with the shielding community in general.

6.3. Resonance Region Subcommittee

This committee is primarily concerned with problems relating to the different formalisms in use in the resonance region; their interpretation, formats, and procedures.

6.4. Fission Product Subcommittee

The evaluation, review, and testing of materials produced in fission is the primary responsibility of this Subcommittee, in cooperation with the Data Testing and Normalization and Standards Subcommittees.

6.5. Nuclear Model Codes Subcommittee

Nuclear model codes are used to supplement experimental evidence in producing the ENDF/B files. The use of these highly complex codes may often result in discrepancies due to numerical or physical errors. The scope of the Nuclear Model Codes Subcommittee is to keep the CSEWG members aware of codes reflecting those discrepancies as well as of new areas of code development.

6.6. New Subcommittees

The final two subcommittees are indicative of the trends presently being considered for the future versions of ENDF/B. The purpose of the Error Quantities Subcommittee is to study and make recommendations on the means and procedures for including errors or confidence limits in the ENDF/B files to be used in sensitivity calculations; while the Non Neutron Data Subcommittee is responsible for recommending means of including charged particle cross sections of interest to shielding and fusion calculations.

7. CONCLUSION

The cooperative NNCSC-CSEWG effort can be viewed as a successful example for the generation of a data base designed to satisfy the needs of a wide user community. The success of this effort can be seen in the widespread use of the ENDF/B Library as a reference file. The generation of a evaluated file, based entirely on the best experimental information

available, yet accurate in the calculation of integral experiments, could only be made possible through the close cooperation of evaluators with experimenters and users through the CSEWG organization.

This effort is expected to continue in the future in order to achieve the following goals:

7.1. Improve the accuracy of the existing files

In particular, ^{235}U capture and fission, ^{239}Pu capture, the standard cross sections, and the number of neutrons emitted in fission will be reviewed in the light of recent experimental evidence. The existing files are known to give reasonably good agreement with integral experiments for the case of fast assemblies. The next version of ENDF/B is expected to give improved agreement with thermal assemblies.

7.2. Expand the number and type of evaluations in the library

The next version of the library should contain evaluations for additional materials and reaction types. Shielding data files will be provided for additional materials.

7.3. Expand the usefulness of the library in new areas of application

This can be done by providing error files, charged particle interaction data, and reactions of interest to controlled thermonuclear fusion work.

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DISCUSSION

F. FRÖHNER: I was interested to hear that error information will in future be included in the ENDF library. Which version of ENDF will contain error information for the first time?

O. OZER: Version IV will contain error information, either as a separate file or as part of the evaluation.

J.J. SCHMIDT: Is there any plan to include evaluated data on neutron capture gamma ray spectra in ENDF/B version IV?

O. OZER: I am not aware of any such plans.

U. FARINELLI: Could you say anything about the recent CSEWG efforts in the field of detector cross-sections?

O. OZER: A file of dosimetry cross-sections is being prepared by the Normalization and Standards Subcommittee, which has responsibility for the Phase I review and Phase II testing of the file.

R. PANNETIER: Is there a possibility of access to the evaluation reports, which certainly contain essential information on the confidence intervals that can be assigned to ENDF/B data for each energy region and for each characteristic?

O. OZER: All ENDF/B evaluations are eventually documented in a formal report. These reports contain information on confidence intervals assigned.

J.Y. BARRE: At the ANS Conference at Kiamesha Lake in September 1972, the reactor physicists concluded that ENDF/B Version 3 gave correct results for reactivities but that there were very considerable discrepancies as regards spectral indices and hence that there were unexplained compensations in this version. In view of what you have said, has anything changed since that date?

O. OZER: Yes, these discrepancies were taken into account in the re-evaluation of the isotopes ^{235}U, ^{238}U, ^{239}Pu, ^{240}Pu and ^{241}Pu.

J.Y. BARRE: Does this mean that the nuclear data used for ^{235}U, ^{238}U, ^{239}Pu, ^{240}Pu and ^{241}Pu were determined by integral experiment?

O. OZER: No, integral results were only used to determine possible problem areas and influence the selection of experimental data sets.

P. RIBON: From one edition of ENDF to another you revise a number of evaluations. What mechanisms do you use for determining the revisions to be made? How do you determine the distribution of tasks and are there complications in connection with the execution of these tasks?

O. OZER: Revisions are made on the basis of recommendations of the Data Testing Subcommittee, which are in turn based on results of Phase II testing. There is no formal mechanism for assigning tasks: evaluations are generally assigned to laboratories that have a particular interest in the subject or are equipped to do the evaluations.

PRELIMINARY COMPARATIVE ANALYSIS OF AMERICAN AND GERMAN NUCLEAR DATA FILES FOR FAST-FISSION-REACTOR CALCULATIONS*

S. YIFTAH, Y. GUR, M. SEGEV, L. GITTER
Soreq, Nuclear Research Centre,
Yavne, Israel

Abstract

PRELIMINARY COMPARATIVE ANALYSIS OF AMERICAN AND GERMAN NUCLEAR DATA FILES FOR FAST-FISSION-REACTOR CALCULATIONS.

The American ENDF/B III, the German KEDAK, the British UKNDL and the Russian evaluated nuclear data files today serve as the main sources of the nuclear data for the calculation of fission reactors. Specifically, the files serve as the major sources of nuclear input for the calculations in the framework of the various big and expensive fast-reactor programs. Each country, naturally, uses its evaluated file and the question that has been asked for some time is the following: are the physics parameters of the fast reactors being calculated and designed a function of the specific evaluated data file that is used in the calculations? In other words, would these physics parameters remain the same were the calculations to use a different evaluated data file as input? To shed some light on this problem, a study has been undertaken where the comparative analysis of the evaluated files is performed on several levels, i.e. the basic-microscopic-data level, the multigroup-cross-section-set level and the physics-parameters-of-specific-systems level. In this paper, typical 1000-1200 MW(e) fast-reactor systems of about 6000-7000 litre core modelling – the target fast reactors being designed today by the major countries having an active fast reactor program – are calculated using the evaluated nuclear data files ENDF/B III and KEDAK as nuclear input. Identical techniques are used to obtain multigroup sets from the files. Also the reactor physics codes used are the same. The differences in results can, therefore, be attributed directly to the differences in the nuclear data input. Practical conclusions and recommendations are presented.

1. INTRODUCTION

The Nuclear Community has been aware for some time of persisting discrepancies in experimental data basic to the prediction of fast reactor parameters. In order to assess the impact of these discrepancies on the accuracy of reactor computations many so-called "sensitivity studies" have been carried out. In a sensitivity study one first estimates, by scanning various sources of experimental data, a likely degree of uncertainty in a given basic nuclear datum, then inserts the estimated uncertainty in reactor codes to calculate the counterpart estimated uncertainty in reactor parameters and finally, assuming certain engineering goals of accuracy in reactor parameter prediction, concludes by establishing whether the basic datum has, or has not, sufficient current accuracy. A recent excellent study of the above nature is the ANL-7851 report by H.H. Hummel[1] .

The complexity of basic data for reactor calculations, as well as the above mentioned discrepancies in such data from different experiments, rendered careful sorting and extensive filing an indespensible link between experiment and calculation. Evaluation of basic nuclear data[2] came to be

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recognized as a tool for the ellimination of experimental discrepancies and a means for providing the reactor analyst with the best updated nuclear input to his codes. A complete evaluation and filing, covering all neutronic data, is a major task, undertaken so far by the major fast reactor communities, namely the American[3], Russian, British and German[4].

Since these files have become the nearly exclusive source of neutronic data for the reactor analyst, it is worthwhile to study the differences between the files. Unlike sensitivity studies which deal with likely discrepancies in data, the comparison of files is a more pragmatic approach, dealing with existing discrepancies in nuclear input to calculations and design.

The philosophy of a comparative analysis of the different evaluated data files of the various countries has been discussed before[5]. A preliminary graphical analysis of the fission, capture and elastic scattering cross sections of ^{239}Pu, ^{240}Pu, ^{241}Pu and ^{242}Pu in the energy range 0.1 to 10 Mev taken from the ENDF/B-I and II, KEDAK and UKNDL files and plotted by the computer's plotter has been performed and presented[6] to the IAEA panel on Neutron Data Evaluation, Vienna, August 1971. Also a preliminary comparison of the ^{239}Pu and ^{235}U α ($=\sigma_{n,\gamma}/\sigma_{n,f}$) of the ENDF/B-III file and the Obninsk absolute measurements of Kononov et al.[7] has been performed[8].

Presently we compare the American file ENDF/B-III[3] with the German file KEDAK[4]. The comparison is centered on the main component elements of typically envisaged 1000 MWe fast reactor systems, namely ^{239}Pu, ^{238}U, Na. The method and procedure of the present comparison are applicable to other elements as well.

2. METHOD OF ANALYSIS

A complete direct comparative inspection of data files to detect differences has two shortcomings. It is inefficient, scanning the entire complex of data; it is non-selective, counting relevant and irrelevant differences alike.

Instead of a complete direct inspection we have adopted the following procedure for detection of data differences. First, the point-wise cross sections of the files are averaged to group constants with a weighting flux characteristic of large dilute systems. Secondly, a few typical 1000 MWe systems are chosen and the microscopic group cross sections admixtured correspondingly to macroscopic group cross sections for these systems. Third, reactor parameters are calculated by a simple spherical diffusion code. The comparison of the files now follows the above steps in reverse. The analysis of differences in reactor parameters point out elements, energy ranges, and types of cross sections, in which the group constant sets may differ. A follow-up inspection of the group cross sections then leads to a positive identification of elements, energy ranges and types of cross sections in which the group constant sets do significantly differ. Since the averaging flux is the same for all compared group sets, the significant differences found in group cross sections are attributed directly to significant differences in basic data of the files themselves.

3. COMPARISON BETWEEN KEDAK AND ENDF/B-III

We have applied the procedure described in the previous section, to a comparison of the KEDAK[4] and ENDF/B-III[3] files.

Two dilute systems, typical of the envisaged 1000MWe reactors, were chosen for the comparison. These are systems (8) and (9) of the list of systems in the intercomparison study reported in an Argonne conference[9] in 1965. The volume ratios of fuel /sodium/S.S. in the cores of these systems are 31/56/13 respectively. The two systems were blanketed alike. Table I describes the systems. The differential data of ENDF/B-III and KEDAK were retrieved from tapes and averaged to group constants on a SNEAK flux[10] (which is typical of large dilute systems) by a set of retrieval and averaging codes NANICK[11], developed at the Soreq Nuclear Research Center. The 26 group energy structure of Bondarenko et al[12] was adopted.

Keff for systems 8 and 9 were calculated at 300°K with macroscopic group sets corresponding to the two microscopic sets averaged from KEDAK and ENDF/B-III. All references to calculations done with these sets will be denoted by KDK and NDF respectively. The results for Keff are given in Table II. The systems were voided of Na and the calculation of Keff repeated. The results for Na voiding are given in Table III. We have not proceeded to doppler effect differences at this point as it became clear from Table II and III that there is vast disagreement between KEDAK and ENDF/B-III overshadowing the smaller effects of temperature on Keff.

Also at this point we could conclude from Table II that some or all of the main parameters which determine the Keff of a large system, namely $\bar{\nu}$(Pu239), σ_f(Pu239), σ_c(Pu239), σ_c(U238), may differ significantly from one file to the other. Table III implied a large difference of the Na cross section at some energy range, the most likely candidate being the group containing the big Na resonance at about 3 kev. Indeed Table IV shows the cross section in this group, as well as the removal cross section to the next group to differ appreciably from one file to the other. As the differences of Table II must have an appreciable component due to the different Na cross sections, the effect of U and Pu cross sections on Keff can be better estimated by comparing Keff of the Na-voided systems. Table V shows this comparison, together with estimates for the relative contribution to the difference in Keff emanating separately from $\bar{\nu}$(Pu239), σ_f(Pu239), σ_c(Pu 239), and σ_c(U238).

TABLE V figures for the estimates of the separate contributions to the difference in Keff are based on a perturbation expression for $\delta k/k$ in Weinberg and Wigner[13], generalized to the multigroup case and evaluated at center-core.

$$\frac{K_{NDF}-K_{KDK}}{K_{KDK}} = \frac{\sum_g \delta(\nu_g\Sigma_{f,g})\phi_g - \sum_g \delta(\Sigma_{a,g})\phi_g}{\sum_g \nu_g\Sigma_{f,g}\phi_g} \qquad (1)$$

where δ signifies a difference (NDF-KDK), ϕ_g is the group flux, and the summation extends over all groups.

Separating the effects of $\bar{\nu}$(Pu239), σ_f(Pu239), σ_c(Pu239) and σ_c(U238) in Eq. (1) we obtain

$$\frac{K_{NDF} - K_{KDK}}{K_{KDK}} = \delta\,\nu(Pu) + \delta_{fiss}(Pu) + \delta_{cap}(Pu) + \delta_{cap}(U) \qquad (2)$$

where

$$\delta\,\nu(Pu) = \frac{\sum_g (\delta\nu_g^{Pu})\sigma_{f,g}^{Pu}\phi_g}{\sum_g \nu_g^{Pu}\sigma_{f,g}^{Pu}\phi_g} \qquad (3)$$

TABLE I. ATOM DENSITIES ($\times 10^{-24}$) FOR SYSTEMS 8 AND 9

System	Core						Blanket				
	Na	^{239}Pu	^{238}U	^{240}Pu	S.S.	O	Na	^{239}Pu	^{238}U	S.S.	O
8	0.0123	0.00072	0.00648	0.00036	0.011	0.0144	0.0069	0.00012	0.012	0.011	0.024
9	0.0123	0.00090	0.00630	0.00045	0.011	0.0144	0.0069	0.00012	0.012	0.011	0.024

TABLE II. KEFF VALUES

System	K_{KDK}	K_{NDF}	$\frac{K_{NDF} - K_{KDK}}{K_{KDK}}$
8	0.930	1.035	+0.11
9	0.962	1.052	+0.09

TABLE III. $\delta k/K$ FOR TOTAL Na-VOIDING

System	KEDAK	ENDF/B-III
8	0.072	0.027
9	0.042	0.001

TABLE IV. CROSS-SECTION OF Na (barns)

File	Cross-section	
	σ_t (group 13) (huge resonance)	σ (group 13→group 14)
KEDAK	139	11.8
ENDF/B-III	92	7.8

TABLE V. SPLIT-UP OF CONTRIBUTIONS TO $(K_{NDF} - K_{KDK})/K_{KDK}$ (for 0% Na)

System	Effect of				Total	Value from successive K
	σ_f^{Pu} (NDF) $-\sigma_f^{Pu}$ (KDK)	σ_c^{U} (NDF) $-\sigma_c^{U}$ (KDK)	σ_c^{Pu}(NDF) $-\sigma_c^{U}$ (KDK)	$\bar{\nu}^{Pu}$ (NDF) $-\bar{\nu}^{Pu}$ (KDK)		
8	+0.048	+0.028	-0.016	-0.003	+0.057	+0.059
9	+0.043	+0.021	-0.010	-0.003	+0.051	+0.049

$$\delta_{fiss}(Pu) = \frac{\sum_g (\delta\Sigma^{Pu}_{f,g})\nu^{Pu}_g \phi_g}{\sum_g \nu^{Pu}_g \sigma^{Pu}_{f,g}\phi_g} \quad (4)$$

$$\delta_{cap}(Pu) = -\frac{\sum_g (\delta\Sigma^{Pu}_{a,g})\phi_g}{\sum_g \nu^{Pu}_g \sigma^{Pu}_{f,g}\phi_g} \quad (5)$$

$$\delta_{cap}(U) = -\frac{N_U}{N_{Pu}} \frac{\sum_g (\delta\Sigma^{U}_{a,g})\phi_g}{\sum_g \nu^{Pu}_g \sigma^{Pu}_{f,g}\phi_g} \quad (6)$$

where N_U/N_{Pu} is the ratio of atomic density of U to Pu in the system.

The overall $\delta k/k$ by the perturbation formula (2) matches quite closely the values of $\delta k/k$ by two complete Keff calculations. We therefore conclude that the difference ($K_{NDF} - K_{KDK}$) is contributed (besides the difference in the big Na resonance cross section) mainly by differences in group cross sections for σ_f(Pu239), σ_c(Pu239) and σ_c(U238) and in group values for $\bar{\nu}$(Pu239) as one goes from KEDAK to ENDF/B-III. In Table VI we show the relative difference in these cross sections for groups 3-14 which cover the range from 1 kev to 3 Mev. This is a range containing approximately 90% of the neutrons of the systems.

TABLE VI. RELATIVE DIFFERENCES (NDF-KDK)/KDK IN GROUP CONSTANTS OF ^{239}Pu AND ^{238}U

Group number		3	4	5	6	7	8	9	10	11	12	13	14
lower energy (keV)		2500	1400	800	400	200	100	46.5	21.5	10	4.65	2.15	1
relative differences in	σ_f (^{239}Pu)	0.03	0.03	0.02	0	0.01	0.01	0.06	0.06	0.14	0.08	0.11	0.32
	σ_c (^{238}U)	-0.15	-0.12	-0.15	-0.09	-0.08	-0.11	-0.06	0.05	-0.01	-0.07	0.05	-0.18
	σ_c (^{239}Pu)	-0.72	-0.55	-0.44	-0.15	0.15	0.15	0.22	0.21	0.36	0.12	-0.03	0.15
	$\bar{\nu}$ (^{239}Pu)	0	-0.003	-0.003	-0.003	-0.003	-0.003	-0.003	-0.003	-0.003	-0.003	-0.003	-0.003

In scanning Table VI it becomes evident that nearly all group differences in σ_f(Pu239) and σ_c(U238), as one goes from KEDAK to ENDF/B-III, happen to be in one direction, namely that of rendering ENDF/B-III a more reactive group set than KEDAK. The opposite effect of differences in σ_c(Pu 239) and $\bar{\nu}$(Pu 239), as evidenced by Table VI, is of a lower,albeit not negligible, magnitude.

A further separation of the terms in Eq. (2) to contributions from individual groups reveals that most of the effect on δk/K stems, apparently, from the very large differences in group cross sections in the range of 0.5 to 5 kev.

4. SUMMARY AND PRELIMINARY CONCLUSIONS

In his recent sensitivity study[1], Hummel has concluded that pessimistic assumptions about discrepancies in the measured σ_f(Pu239), σ_c(Pu239) and σ_c(U238) influence Keff of a large dilute system by the amount of several percent for each of the three cross sections.

As the present study shows,the evaluation effort which led, independently but with a few years difference, to the KEDAK and the ENDF/B-III files has not come up with better agreement in the calculated Keff of a large dilute system. We therefore conclude that what Hummel consider "pessimistic discrepancies" may reflect real discrepancies which persist even through evaluation.

Additional uncertainty in Keff, and a substantial difference in the Na void effect, is caused by different values for the cross sections of the big Na resonance in the two files.

Each of the above discrepancies in basic data, namely in σ_f(Pu239), σ_c(U238), σ_c(Pu239), and σ_{res}(Na), is alone a source for a few percent uncertainty in Keff. It so happens that the effects of discrepant σ_f(Pu239) and σ_c(U238), as one goes from KEDAK to ENDF/B-III, add up (see Table V), even after some reduction by an opposite effect from the discrepant σ_c(Pu 239), to an impressive 5% difference in Keff. We therefore conclude that the present accuracy of measured Pu and U cross sections is much too low to meet the reasonable goal of a 1% error limit[1] in calculated Keff.

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A SYSTEMATIC TEST OF THE ENDF-B/3 EVALUATED CROSS-SECTION LIBRARY ON "CLEAN" CRITICAL ASSEMBLIES

J.J. WAGSCHAL, Atara YAARI
Racah Institute of Physics,
The Hebrew University of Jerusalem, Israel

Abstract

A SYSTEMATIC TEST OF THE ENDF-B/3 EVALUATED CROSS-SECTION LIBRARY ON "CLEAN" CRITICAL ASSEMBLIES.

The use of integral data to test or even to modify neutron cross-sections has been popular for almost a decade. This article describes recent tests performed on the new ENDF-B/3 evaluated neutron cross-sections compilation. Fifteen "clean" measurements of critical masses of bare and reflected, plutonium and uranium spheres served as integral data. The specifications of most of these critical assemblies have been reevaluated and published in 1969. The quoted experimental errors in the critical masses of these assemblies were translated individually for each system into an uncertainty in the effective multiplication factor k_{eff}. The effective multiplication factors k_{eff} of the critical assemblies were calculated in a forty-group transport S_8 approximation, and the proper adjustments needed to account for anisotropic scattering and for the finite-differences scheme were carefully calculated for each assembly. All calculated k_{eff} fell outside the quoted uncertainties. A least-squares and perturbation scheme for the use of integral data in cross-section adjustments was applied to these critical masses and to the ENDF-B/3 ^{235}U, ^{238}U and ^{239}Pu cross-sections. The new effective multiplication factors k_{eff} are now well within the quoted experimental errors, and the adjustments needed in the cross-section evaluation are minor. The numerical results are presented and discussed.

1. Introduction.

One of the most severe tests of any new cross section library is the calculation of critical masses of various fast neutron assemblies. The high precision of the critical mass determination in simple geometry systems, led to the development of methods that use "clean" critical experiments for the modification of neutron cross sections. Such methods, which were reported already in 1964 [1] [2], became more popular and respectable yet more controversial in recent years[3][4][5][6]. In his 1971 review paper, Pendlebury[7] states: "At the present time one cannot have the mean curve through the (cross section) measurements giving the mean curve through the experimental k_{eff}". In the present paper we test the ^{235}U, ^{238}U, ^{239}Pu ENDF-B/3 evaluated nuclear data files and check whether this statement is still valid or not.

In section 2 the "clean" critical assemblies are described in detail. Special attention is given to the quoted experimental uncertainties. In section 3 we describe the method of analysis, and the accuracy of the calculations is discussed. Experimental and calculated effective multiplication factors k_{eff} are compared in section 4. A cross section modification scheme is applied and the results are presented in section 5. We conclude with a discussion of the results in section 6.

Table I. CRITICAL SPECIFICATIONS OF BARE AND NATURAL URANIUM REFLECTED PLUTONIUM SYSTEMS.

Assembly	1	2	4	7	11
Core:					
Radius R cm	6.3849	6.6595	4.5332	5.0419	4.5679
Density ρ g/cm^3	15.61	15.73	15.53	15.77_8	15.74
Ga at. fraction	.0341	.0338	.0367	.0335	$.0341_4$
^{239}Pu at. fraction	.9195	.7381	.9138	.9161	$.9528_3$
^{240}Pu at. fraction	.0435	.1942	.0466	.0474	$.0130_3$
^{241}Pu at. fraction	.0029	.0300	.0029	.0030	-
^{242}Pu at. fraction	-	.0039	-	-	-
Reflector:					
Thickness R cm	-	-	19.6088	4.1275	11.6911
Density ρ g/cm^3	-	-	19.0	18.92	18.9
^{235}U at. fraction	-	-	.0072	.0072	.0072
^{238}U at. fraction	-	-	.9928	.9928	.9928
Reference	[8]	[8]	[8]	[8],[10]	[9],[12]

2. "Clean" critical assemblies.

Our "clean" critical assemblies are bare and natural-uranium-reflected plutonium and oralloy spheres. Additional critical assemblies are **plutonium** cores in oralloy shells. The specifications of most of the critical assemblies are from the 1969 reevaluation of the Los Alamos fast neutron systems[8]. Additional systems have been selected from older publications [9][10][11][12]. The critical specifications of all assemblies are first listed as they appear in the literature. The densities and compositions are also given in a form suitable for input to neutron transport calculations in Tables I,II,III.

Table II. CRITICAL SPECIFICATIONS OF BARE AND NATURAL URANIUM REFLECTED ORALLOY SYSTEMS.

Assembly	5	6	12	13	14
Core:					
Radius R cm	8.7407	6.1156	6.3097	7.7548	6.9769
Density ρ g/cm^3	18.74	18.62	18.76	18.70	18.67
^{234}U at. fraction	.0102	-	-	-	-
^{235}U at. fraction	.9377	.9332	.9397	.9398	.9406
^{238}U at. fraction	.0521	.0668	.0603	.0602	.0594
Reflector:					
Thickness ΔR cm	-	18.0086	9.9695	1.7348	4.4247
Density ρ g/cm^3	-	19.0	18.9	19.0	18.67
^{235}U at. fraction	-	.0072	.0072	.0072	.0072
^{238}U at. fraction	-	.9928	.9928	.9928	.9928
Reference	[8]	[8]	[12]	[8]	[8]

Table III. CRITICAL SPECIFICATIONS OF PLUTONIUM CORES IN ORALLOY SHELLS.

Assembly	3	8	9	10	15
Core:					
Radius R cm	5.0419	3.1544	2.7051	2.7051	2.7051
Density ρ g/cm^3	15.778	19.22	19.48	19.42	19.43
Ga at. fraction	.0335	-	-	-	-
^{239}Pu at. fraction	.9161	.9500	.9756	.9497	.8047
^{240}Pu at. fraction	.0474	.0470	.0234	.0473	.1610
^{241}Pu at. fraction	.0030	.0030	.0010	.0030	.0292
^{242}Pu at. fraction	-	-	-	-	.0051
Nickel:					
Coating ΔR cm	-	-	.0254	.0254	.0254
Density ρ g/cm^3	-	-	4.17	4.99	5.98
Oralloy shells:					
Thickness ΔR cm	1.6637	4.1935	2.3495	2.3495	2.3495
Density ρ g/cm^3	18.80	18.80	18.63	18.63	18.63
Thickness ΔR cm	-	-	.1270	.1270	.1270
Density ρ g/cm^3	-	-	17.24	17.24	17.24
Thickness ΔR cm	-	-	.0253	.0641	.2023
Density ρ g/cm^3	-	-	17.424	17.424	17.424
^{235}U at. fraction	.9328	.9328	.9328	.9328	.9328
^{238}U at. fraction	.0672	.0672	.0672	.0672	.0672
Reflector:					
Thickness ΔR cm	-	-	18.8977	18.8589	18.7452
Density ρ g/cm^3			18.80	18.80	18.80
^{235}U at. fraction	-	-	.0072	.0072	.0072
^{238}U at. fraction	-	-	.9928	.9928	.9928
Reference	[8]	[9]	[11]	[11]	[11]

The critical specifications of the assemblies as given in the literature are as follows:

Ass 1. (Jezebel)

Pu (4.5 at.% ^{240}Pu, 0.3 at.% ^{241}Pu), 1.02 wt.% Ga.
m_c(alloy) = 17.02 kg ± 0.6% at ρ(alloy) = 15.61 g/cm^3 .

Ass 2. (Dirty Jezebel)

Pu(20.1 at.% ^{240}Pu, 3.1 at.% ^{241}Pu, 0.4 at.% ^{242}Pu), 1.01 wt.% Ga.
m_c(alloy) = 19.46 kg ± 0.8% at ρ (alloy) = 15.73 g/cm^3.

Ass 3.

Sphere of 8.471 kg Pu alloy, 4.9 at.% ^{240}Pu, 0.31 at.% ^{241}Pu, 1.0 wt.% Ga, ρ(alloy) 15.77_8 g/cm^3. Critical when surrounded intimately by U(93.2), ρ(U) = 18.80 g/cm^3 , at a thickness of 0.655 in. ± 1%.

Ass 4. (Flattop 49, Popsy)

6.06 ± 0.03 kg Pu alloy, 4.8 wt.% ^{240}Pu, 0.30 wt.% ^{241}Pu, 1.10 wt.% Ga, at the alloy density 15.53 g/cm^3 (reflector thickness 7.72 in.)
The reflector is normal uranium at a density of 19.0 g/cm^3.

Ass 5. (Godiva)

52.42 kg ± 0.3% sphere of U(93.71) at an average density of 18.74 g/cm^3 . The ^{234}U content is 1.02 wt.%, no ^{236}U.

Ass 6. (Flattop 25, Topsy)

17.84 ± 0.04 kg U(93.24) at the uranium density 18.62 g/cm^3. (reflector thickness 7.09 in.)
The reflector is normal uranium at a density of 19.0 g/cm^3.

Ass 7.

The core of this assembly is the same core as of assembly 3. The reflector is natural uranium at a density of 18.92 g/cm^3. The critical thickness 1.625 in. ±1%.

Ass 8.

Sphere of Pu(100%), 4.7% ^{240}Pu, $\rho(Pu)$ = 19.22g/cm^3, diameter 2.484 in., m(Pu) = 2.52$_7$kg.
Critical when surrounded by U(93.17), $\rho(U)$ = 18.8g/cm^3,at a thickness of 1.651 in. The critical ^{235}U mass of the reflector 26.8 $\pm$ 0.1 kg.

Ass 9.

A Pu sphere is surrounded by concentric U(93.2) shells and reflected by a thick natural uranium reflector.
The outer diameter of the Pu sphere is 2.150 in., it has a 10 mil nickel coating. The Pu mass is 1615.45 g, the nickel mass is 10.89g. The Pu isotopic composition is: .9556 ^{239}Pu, .0234 ^{240}Pu, .0010^{241}Pu. The outer diameter of the first oralloy shell is 4.000 in. and the mass is 8642 g. A second concentric oralloy shell has an outer diameter of 4.100 in. and mass 728g. An oralloy cap (part of a third concentric shell of outer diameter 4.240 in. and mass 1092 g) of mass 151 g is added. The rest of the third shell is natural uranium. The outer diameter of the reflector is 19.0 in. and the natural uranium density 18.80 g/cm^3.

Ass 10.

This system is similar to assembly 9. The same critical assembly machine was used with the following changes:
The Pu mass is 1610.30 g and the isotopic composition is: .9497 ^{239}Pu, .0473 ^{240}Pu, .0030 ^{241}Pu.The nickel mass is 11.76 g. The first two oralloy shells are the same as in assemblly 9. An oralloy ring of mass 385g was used as part of the third shell. The rest of the third shell was natural uranium. The system was reflected by the same natural uranium reflector.

Ass 11.

6.284 kg Pu sphere, 1.35 at.% ^{240}Pu, 1.02 wt.% Ga, at the density 15.74 g/cm^3. Critical when surrounded by a natural uranium reflector of thickness 4.603 in. at a density of 18.9g/cm^3.

Ass 12.

In close-fitting 3.925-in.-thick normal uranium(ρ=18.9g/cm^3), the critical mass of a U(93.90) sphere atρ(U)=18.76 g/cm^{3}is 19.74kg U $\pm$ 0.5%.

Ass 13.

In close-fitting 0.683-in.-thick normal uranium (ρ= 19.00g/cm^3), the critical mass of a U(93.91) sphere at ρ(U)=18.70g/cm^{3}is 36.53kg U$\pm$0.5%.

Ass 14.

In close-fitting 1.742-in.-thick normal uranium (ρ=18.67g/cm^{3}because of gaps in reflector), the critical mass of a U(93.99) sphere at ρ(U)=18.67g/cm^3 is 26.56 kg U $\pm$ 0.5%.

Ass 15.

This system is similar to assembly 9. The Pu mass is 1611.19 g and the isotopic composition: .8047 ^{239}Pu, .161 ^{240}Pu, .0292 ^{241}Pu, .0051 ^{242}Pu.
The nickel mass is 14.10 g. All three oralloy shells are used and the critical mass of the oralloy was estimated by extropolation of the 1/M vs mass curve as 10,618 g.

3. Method of analysis.

The ENDF-B/3 library was used both for cross section data and for nuclear model information. The isotopes used and their ENDF material numbers are listed in Table IV. The gallium data was compiled from BNL-325[13].

Table IV. ENDF-B/3 MATERIAL NUMBERS.

N_i	^{234}U	^{235}U	^{238}U	^{239}Pu	^{240}Pu	^{241}Pu	^{242}Pu
1123	1043	1157	1158	1159	1105	1106	1161

The fission spectrum in ENDF is both material dependent and energy dependent. Our SNG code is capable of using different fission matrices in different spatial regions. For each region the fission matrix is prepared in the following way.

$$\tau^{(f)}_{g\leftarrow g'} = \sum_m N_m \frac{\int_{E_{g-1}}^{E_g} \nu_m(E')\sigma^f_m(E')f_m(E'\rightarrow E)dE}{\qquad\qquad\qquad\qquad\qquad\qquad} g'$$

In this expression:

$\tau^{(f)}_{g\leftarrow g'}$ is the fission matrix element describing neutrons born in energy group g as a result of fission caused by a neutron in energy group g'.

N_m is the number of nuclei of material m in unit volume.

$\nu_m(E')$ is the number of secondary neutrons born by fission of material m caused by a neutron of energy E'.

$\sigma^f_m(E')$ is the m^{th} fission cross section.

$f_m(E'->E)$ is the m^{th} fission spectrum for fission caused by a neutron of energy E'.

E_{g-1}, E_g are the lower and upper energy boundaries of energy group g.

$\overline{\qquad}\, g'$ denotes an average (with a proper weight function) over energy group g'.

Other group average cross sections and scattering transfer matrices are prepared in a conventional way [14] [15].

The uncertainties of the integral measurments are stated in the literature in terms of uncertainties in the mass of the critical assemblies or in terms of their dimensions. From the literature it is not always clear how to interpret the stated mass uncertainty. One possibility is to interpret the stated error Δm as a mass added onto the surface of the appropriate material. A second possibility is to interpret Δm as mass added by a homogeneous increase of the density of the appropriate

material. In our analysis we need the experimental uncertainties stated in terms of an error in the effective multiplication factor k_{eff}. When the experimental uncertainties are stated in terms of reflector thickness the translation to an error in k_{eff} is unique. We denote this uncertainty by Δk_R. We use the same symbol (Δk_R) for the uncertainty using the first interpretation of Δm (viz. assuming the mass error to be an increment in the surface mass). The uncertainty in k_{eff} obtained by using the second interpretation of Δm is denoted by Δk_ρ.

The translation of the experimental errors to terms of uncertainties in k_{eff} is obtained by a series of four group S_8 transport approximation calculations. The unit used for k_{eff}^{-1} and for Δk_R and Δk_ρ is one part in a thousand (e.g., $\Delta k = 1.5$ means an uncertainty of 0.0015 in k_{eff}).

The experimental uncertainties and their translation to Δk_R , Δk_ρ are presented in Table V.

The small values of Δk which correspond to the quoted experimental uncertainties, dictate the high accuracy needed in the calculations. The effect of the number of energy groups as well as the effect of the number of spatial points have been checked to be negligible. Corrections due to the finite order of the S_N calculations and due to the use of the P_o-transport approximation were established and taken into account for each assembly individually.

The major part of the calculations was performed using the S_8, P_o-transport approximation with forty energy groups. This rather high number of energy groups for fast neutron systems should eliminate inaccuracies arising from the choice of the cross section weight function. The cross section weight function used is the old Cranberg fission spectrum (i.e., $N(E)=0.453\ e^{-E/0.965}\ \sinh\sqrt{2.29E}$). The effective multiplication factors of six assemblies (a bare plutonium sphere, a bare oralloy sphere, thick-natural-uranium-reflected plutonium and oralloy cores, a thin oralloy shell surrounding a plutonium core, and a thin-natural-uranium-reflected plutonium core) were also determined using a much higher number of energy groups. Except for the two thick-uranium-reflected systems the deviations in k_{eff} are less than 0.1, and

Table V. EXPERIMENTAL UNCERTAINTIES OF CRITICAL SPECIFICATIONS AND CORRESPONDING Δk_{eff} EQUIVALENT.

No.	Δk_R	Δk_ρ	Quoted experimental error
1	1.7	5.2	0.6% of mass
2	2.3	6.9	0.8% of mass
3	1.5	-	1% of reflector thickness
4	1.5	3.4	30g Pu alloy
5	.8	2.5	0.3% of mass
6	.6	1.3	40g U(93.71)
7	1.0	-	1% of reflector thickness
8	1.5	-	
9	1.5	-	
10	1.5	-	
11	1.5	-	
12	1.3	3.2	0.5% of mass
13	1.3	3.8	0.5% of mass
14	1.3	3.4	0.5% of mass
15	1.5	-	

even for these two assemblies the difference in k_{eff} between the calculations with the high number of energy groups and the forty group calculations is less than 0.3.

The corrections due to the use of the transport approximation, the S_8 approximation, and the finite number of spatial points were calculated for each assembly. The correction calculations were performed using four energy groups. The accuracy of these corrections as well as the accuracy of the Δk equivalent of the experimental uncertainties were checked by running a few forty group and twenty group calculations. The differences between the respective four and forty group calculations of the corrections were less than 3%.

Additional tests were performed to verify that all the corrections are additive to within the same 3% of the corrections.

The correction from S_8 to S_{64}, which we consider as S_∞ according to the convergence pattern, varies from 1.4 for the bare oralloy system to over 3.0 for the plutonium spheres with thick uranium reflectors. The difference $\Delta k(S_N)=k_{S8} - k_{S64}$ is given in Table VI. The corrections are listed for three types of systems according to increasing reflector thickness. The variation of the corrections with the change of the core materials and with the reflector thickness is as expected:

a. Higher corrections for plutonium systems.
b. The corrections increase as function of reflector thickness, reach a maximum and then fall off slightly.

Table VI. CORRECTIONS DUE TO THE $S_8 - S_\infty$ DIFFERENCE AND DUE TO ANISOTROPIC SCATTERING.

Plutonium cores			Oralloy cores			Plutonium cores in Oralloy shells		
No.	$\Delta k(S_N)$	Δk(anis)	No.	$\Delta k(S_N)$	Δk(anis)	No.	$\Delta k(S_N)$	Δk(anis)
1	2.0	3.4	5	1.4	2.9	3	2.1	4.8
2	2.0	3.5	13	1.5	4.8	8	2.6	5.4
7	2.6	8.7	14	1.7	6.2	9	3.2	8.1
11	3.1	9.2	12	2.0	6.6	10	3.2	8.0
4	3.0	8.9	6	2.0	6.4	15	3.0	7.8

A similar pattern of corrections is noticed for the anisotropic scattering. The angular dependence of the elastic scattering source term of the Boltzmann transport equation is Legendre expanded and the series truncated after L terms. The convergence was tested up to L=9 and was found to be quite sufficient. For all systems truncation after L=5 is accurate to at least .05 in k_{eff} (i.e. $|k_{L=9} - k_{L=5}| \leq .00005$). The corrections $\Delta k(\text{anis}) = k_{tr} - k_{L=9}$ are given in Table VI. This correction is the most important one. It varies from 2.9 for a

bare oralloy system to more than 9.0 for thick natural-uranium-reflected plutonium systems.

A sufficient number of spatial points was taken so that doubling it gave an average change of k_{eff} of less than 0.15.

4. Effective multiplication factors k_{eff}.

The effective multiplication factors of the fifteen fast neutron critical assemblies were calculated and the proper corrections were taken into account. For a good nuclear data file the deviations from $k_{eff}=1.$ should be less than the experimental error, provided the critical specifications are

Table VII. THE DEVIATION OF k_{eff} CALCULATED WITH ENDF-B/3 FROM THE EXPERIMENTAL VALUE IN UNITS OF EXPERIMENTAL ERROR

Assembly	Δk_R	$\frac{k_{eff}-1.}{\Delta k_R}$
1	1.7	2.329
2	2.3	-1.022
3	1.5	3.140
4	1.5	4.360
5	.8	6.450
6	.6	10.533
7	1.0	6.080
8	1.5	4.387
9	1.5	5.294
10	1.5	5.035
11	1.5	9.318
12	1.3	5.475
13	1.3	2.792
14	1.3	3.185
15	1.5	3.829

correct. In Table VII the deviations of the calculated k_{eff} from unity are given in units of the uncertainties Δk_R. All deviations are greater than the assumed experimental uncertainties Δk_R. This interpretation of the experimental error might be considered a little too stringent. Even if we take Δk_ρ as the assumed experimental error the calculated k_{eff} of only three systems Ass 1, Ass 2 and Ass 13 are within the experimental uncertainty.

The calculated k_{eff} of all systems but one are higher than unity. The only system with the low calculated k_{eff} is the bare plutonium sphere containing twenty percent ^{240}Pu. Assembly 15 that also contains a high percentage of ^{240}Pu has a lower k_{eff} than the similar assemblies 9 and 10 (though it is still greater than unity).

The results presented here for the fast neutron systems suggest the necessity of a modification of the evaluated neutron data file in the fast neutron energy range. In the next section an attempt is made to assess the needed modification.

5. Cross section adjustment procedure.

The cross section modification scheme employed in this work has been discussed in detail in earlier papers [16] [17], and the actual procedure followed is briefly reviewed. The adjusted cross section of reaction r (fission, capture, inelastic, and number of secondary neutrons per fission), for material m(^{235}U, ^{238}U, ^{239}Pu, ^{240}Pu), in the energy range g ($E_{g-1} \leq E \leq E_g$) is written as $(1+ p^r_{mg})\sigma^r_{mg}$. Here p^r_{mg} is the fractional change of cross section σ^r_{mg} and is related to the desired change in k_{eff} of assembly i by

$$k^i_{eff}\ (p^r_{mg})-1= -\sum_{m,r,g} D^{ir}_{mg}\ p^r_{mg} +\varepsilon_i$$

The D^{ir}_{mg} is the sensitivity of the effective multiplication factor of assembly i to a change in the cross section typified by m,r,g. These quantities are calculated by a first order perturbation method. The residual deviation of k_{eff} from unity is denoted by ε.

The p^r_{mg} are obtained by a least squares procedure. The quantity minimized is:

$$\frac{1}{\Delta E_o} \sum_{m,r,g} \left(\frac{p^r_{mg}}{\beta^r_{mg}}\right)^2 (E_g - E_{g-1}) + \sum_i \left(\frac{k^i_{eff}-1}{\Delta k^i_R}\right)^2$$

In this expression β^r_{mg} is the fractional experimental uncertainty in σ^r_{mg}. The relative weight of the integral experiments in the least squares procedure is given by a free parameter ΔE_o.

Table VIII. DEVIATIONS OF k_{eff} FROM UNITY BEFORE AND AFTER A LEAST SQUARES PERTURBATION ADJUSTMENT OF THE CROSS SECTIONS.

Assembly	Δk_R	$\frac{k_{eff}-1.}{\Delta k_R}$	$\frac{k^{new}_{eff}-1.}{\Delta k_R}$
1	1.7	2.329	.303
2	2.3	-1.022	-.787
3	1.5	3.140	-.438
4	1.5	4.360	-.355
5	.8	6.450	.333
6	.6	10.533	-.255
7	1.0	6.080	.356
8	1.5	4.387	.411
9	1.5	5.294	.405
10	1.5	5.035	.290
11	1.5	9.318	4.190
12	1.3	5.475	.802
13	1.3	2.792	-.823
14	1.3	3.185	-.317
15	1.5	3.829	-.231

The sum over i can be taken over any desired number of integral experiments. Taking into account only a small number of integral experiments might still produce a set of cross section modifications p^r_{mg} which will bring $k_{eff}(p^r_{mg})-1$ (i.e., k_{eff} obtained using the modified cross sections) of many more critical assemblies to within Δk_R.

We determined the cross section modifications for ^{235}U, ^{238}U, ^{239}Pu, ^{240}Pu in forty energy ranges (identical to our group structure). The sensitivities of k_{eff} of all the assemblies to the cross sections were calculated by our perturbation code. The fractional uncertainties of the cross sections β^r_{mg} were taken from Küsters [18] and Greebler [19]. The cross section modifications obtained from the least squares procedure, taking into account all assemblies except Ass11, are within their respective experimental uncertainties. The original deviations of k_{eff} from unity as well as the residual deviations after the cross section modification are given in Table VIII in units of Δk_R.

6. Discussion.

Reexamination of Table VII leads to the following observations:

a. All k_{eff} calculated with the unmodified ENDF-B/3 file are higher than unity with the exception of Ass 2, dirty Jezebel.

b. Assembly 15, which contains 16 per cent ^{240}Pu, has a lower k_{eff} than the similar assemblies 9 and 10.

c. The deviation of k_{eff} from unity for all systems, including assemblies with thick uranium reflectors, is less than 8, with the exception of Ass 11 where the deviation is 14.

The last observation suggests a deficiency in the critical specifications of Ass 11. The case of the two assemblies with a high ^{240}Pu content (Ass 2, Ass 15) could lead to either an increase in the fission cross section and a decrease in the capture cross section of ^{240}Pu or to the conclusion that there is an error in the composition specifications. The general pattern of the high

values of k_{eff} for all plutonium and oralloy systems indicates too high fission cross sections. All these qualitative conclusions should be brought up by the systematic least squares perturbation scheme in a quantitative way.

Taking the fourteen assemblies into the adjustment procedure (all but Ass 11) the cross section modifications, although within their experimental uncertainties, are pronounced. The ^{240}Pu fission cross section is increased by up to 7.5% around 1 MeV and the capture cross section lowered by up to 4.0% around 200 keV. As to ^{238}U changes of up to 2% are noticed in the inelastic scattering cross section. The ^{235}U fission cross section is lowered by about 4.0% between 50 and 200 keV and the ^{235}U inelastic scattering cross section is lowered by 5% between 300 keV and 1 MeV. The ^{239}Pu fission cross section modification is less than 2.5% , however it is not monotonous. The ^{239}Pu inelastic scattering cross section is increased by 2% between 1.1 and 2.5 MeV.

Although these modifications are perfectly legitimate according to the rules of our game we have little confidence in them, because they partially depend on critical assemblies in the specifications of which we have less confidence.

The modifications of the cross sections obtained by taking into account only three assemblies, Ass 1, Ass 4 and Ass 3 (the bare plutonium sphere, the thick-uranium-reflected plutonium core and the plutonium sphere within a thin oralloy shell) are sufficient to bring the $k_{eff}(p^r_{mg})-1$ of thirteen assemblies to within 1.3 Δk_R (Δk_R was stringent translation of the experimental uncertainty). The two assemblies which are still way out are, as expected, Ass2 and Assll. Out of these thirteen assemblies eight are within the experimental uncertainty (1,3,4,5,6,7,12,15) and three are within $1.1\Delta k_R$(8,10,14). The cross section modifications are presented in Table IX. There is a negligible modification of the ^{238}U and ^{240}Pu cross sections.

Trying to include Ass 11 in the adjustment procedure does not give modified cross sections that bring $k_{eff}(p^r_{mg})-1$ of Ass 11 within Δk_R . On the contrary it has a bad effect on the other systems.

Table IX. SUGGESTED CROSS SECTION ADJUSTMENTS OBTAINED BY A LEAST SQUARES PERTURBATION SCHEME ON ASSEMBLIES 1,3,4.

Data	Energy	Adjustment
$^{235}U\ \sigma(n,f)$	50 to 200 keV	-1.0%
	200 to 400 keV	-1.2%
	400 to 750 keV	-1.0%
	750 to 1000 keV	-0.8%
	1 to 1.6 MeV	-0.5%
$^{235}U\ \sigma(n,n')$	300 keV to 1.0 MeV	-1.2%
$^{239}Pu\ \sigma(n,f)$	50 to 300 keV	-2.4%
	300 to 500 keV	-0.8%
	500 keV to 1 MeV	-0.5%
$^{239}Pu\ \bar{\nu}$	10 keV to 1.6 MeV	-0.1%

To conclude minor modifications of the ENDF-B/3 file [20] are sufficient to bring $k_{eff}(p^r_{mg})-1$ of most critical assemblies to within Δk_R . The critical specifications of a few systems should be rechecked.

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DISCUSSION

P. P. T. REUSS: Which cross-sections have you modified and how much?

Mrs. A. YAARI: In the last table of the full paper you will find a partial answer to your question. It gives the corrections for the case in which only three assemblies (1, 3, 4) are taken into account. There are also corrections in other cross-sections but these are smaller: e. g. ^{239}Pu (n, γ), 10-300 keV, 0.8%; elastic 50-300 keV, 0.3%. We also modified the fission spectrum, i. e. we lowered the average energy. When we take 14 assemblies into account the corrections are larger.

P. SILVENNOINEN: Some earlier cross-section sets recommended for some of your assemblies, e. g. those given by Hansen and Roach, produce negative values for some corrected transport cross-sections. How do your group cross-sections behave in this respect?

Mrs. A. YAARI: As we used an S_8-P_0 approximation to the Boltzmann equation the problem never arose.

THE ROLE OF INTEGRAL EXPERIMENTS IN THE PRODUCTION OF NUCLEAR DATA FOR REACTOR CORE AND SHIELD DESIGN AND FOR IRRADIATION EXPERIMENTS

U. FARINELLI
C.N.E.N., Centro Studi Nucleari della Casaccia,
Rome, Italy

Abstract

THE ROLE OF INTEGRAL EXPERIMENTS IN THE PRODUCTION OF NUCLEAR DATA FOR REACTOR CORE AND SHIELD DESIGN AND FOR IRRADIATION EXPERIMENTS.

The inadequacy of differential measurements is briefly reviewed. A general scheme of a "global approach" taking into account both differential and integral information is derived. The various philosophies that can be employed in designing integral experiments and in using their information are critically reviewed. The application of this type of approach to fast-reactor cross-sections and the bases for the feedback to nuclear data evaluations and for adjustment procedures are reviewed. The possibility of using these methods to uncover systematic errors is examined. The extension of these procedures to other areas is discussed; in particular, the interactions of calculational model and nuclear data in thermal reactor design are considered. In the case of shielding, the same problem is parallelled by the necessity of extending the sensitivity calculations. In the irradiation experiments and, in particular, for the corss-sections of the integral detectors used, the connection of nuclear data, integral experiments and damage functions is discussed, and the usual criterion of spectrum unfolding is shown to be too restrictive for practical purposes.

1. Introduction

Nuclear data used in reactor technology and other applications derive from three sources: theoretical calculations, differential measurements and integral experiments. Theoretical calculations, based on more or less sophisticated nuclear models, are widely used in evaluations, in interpolation and extrapolation of differential data and have a major role in providing us quantitative estimates where no reliable experimental information is available. Broadly speaking, there seems to be general agreement on when and how to make use of theoretical models, and what should be expected from them. The respective role of differential measurements and integral experiments and especially the way in which to put together the information from these two sources, on the other hand, is a fairly controversial subject.

It is our intention here to discuss shortly the problem of making the best use of the available information in rather general terms and then to review the situation in some fields of application. Rather than proposing a new model, we shall try to look at the various philosophies so far adopted in a unified way and to identify trends of development. Although most of the work in this field up to now concerns fast reactors, and in particular the interpretation of fast critical experiments, we should keep in mind

other fields of applications such as shielding, damage studies and fusion reactors, where problems of this kind are just being identified.

2. A General Model

Many physicists believe that the ideal approach to reactor design and other nuclear applications is, or should be, to use only detailed cross sections obtained from measurements performed with accelerators and accurate calculational methods that starting from these cross sections evaluate the integral quantities without introducing appreciable errors. Actually, as we shall see, all practical applications up to now rely more or less heavily on integral evidence, although this may some time be concealed. It is quite questionable whether an ideal approach based on differential measurements alone will ever be practicable. Although tremendous progress has been achieved in cross section measurements and evaluations, in code development and in computer technology to make "fundamental" calculations possible, the improvements achieved in terms of predicting reactor parameters are much slower and not uniform. More refined measurements have shown discrepancies with each other; this has called for evaluations to solve these discrepancies. But as evaluations are based very much on personal judgement, they often differ among each other. Recently we have heard of requests for new measurements in order to solve discrepancies among evaluations /1/! So this procedure of increasing the reliability of differential data does proceed, but more slowly than expected.

Cross sections evaluated on the basis of differential measurements alone perform rather poorly in predicting integral quantities.

An example is given in Table I, where the results obtained with the most recently generated "uncorrected" data file in the UK, FGL5U, is seen to be fully inadequate /2/ while adjusted cross sections perform nearly an order of magnitude better.

TABLE I

Discrepancies in calculated values for a typical fast reactor using the most recent evaluated, unadjusted U.K. data (fine group library FGL5U, 2000 groups, 1972)

k_{eff} for U and Pu fuelled systems	5% low
k_{∞} for null reactivity test zones up to	10% low
U-238 fission/Pu-239 fission	15% low
U-238 capture/Pu-239 fission	12% high
U-235 fission/Pu-239 fission	3% high

Even some old empirical multigroup libraries, although partly based on obsolete measurements, still give good results in par-

ticular fields, because they contain errors that compensate each other, or compensate for some errors introduced by the calculational method.

The amount of detail given by the differential libraries (especially the number of energy points) has in many cases far surpassed what is necessary and usable in practical applications, so that a preliminary rough condensation procedure has to be used to obtain the data in more "integral" form. Such is the case, for instance, of the cross section of iron in ENDF/B-3, represented by more than 11000 energy points /3/.

However, the main point against a purely differential approach is the waste that it makes of a rich and valuable source of pertinent information, integral experiments. It is quite possible that integral information is less meaningful if the use to be made of the cross section is, for instance, basic nuclear physics. Some misunderstanding in the discussions over the sources of nuclear data may be due to the incautious statement of some supporter of correlation procedures that the purpose of his exercise was to "improve cross sections". As Dragt rightly puts it, "the final aim of the analysis is to obtain estimates for integral quantities, the differential data having no interests in themselves (at least for reactor design purposes)" /4/.

We shall therefore put ourselves from the viewpoint of the user, who wants to predict the gross properties of a system with adequate accuracy, who wants if possible to estimate this accuracy and who looks for useful information from whatever source is available. Two more properties generally characterize this user: he works on a time schedule and has a finite budget.

In this context, the most reasonable and logical approach appears to be some sort of global approach in which both differential and integral information, all the available information, is taken into account, with appropriate allowance for the characteristics of each type of measurement, accuracies, statistical and possible systematic errors, correlations, normalizations and so on.

This is of course easier to say than to do. We shall discuss in the following a somewhat idealized general procedure, which is represented schematically in Fig.1 in the form of a flow diagram allowing for parallel operations and logical options. Most of the philosophies followed for the various problems in different countries can be identified as paths through this diagram.

According to our general point of view, the first step of our approach should be to identify the design tasks and their required accuracy, these tasks are generally different from the related physical quantities (next step in our procedure) which will be in general the immediate objective of our calculations. For instance, k_{eff} or conversion ratios or neutron spectra are common example of relevant physical quantities: but the design tasks that we shall finally solve will be, for instance, the enrichment that will yield a given core life or the damage life of a structural

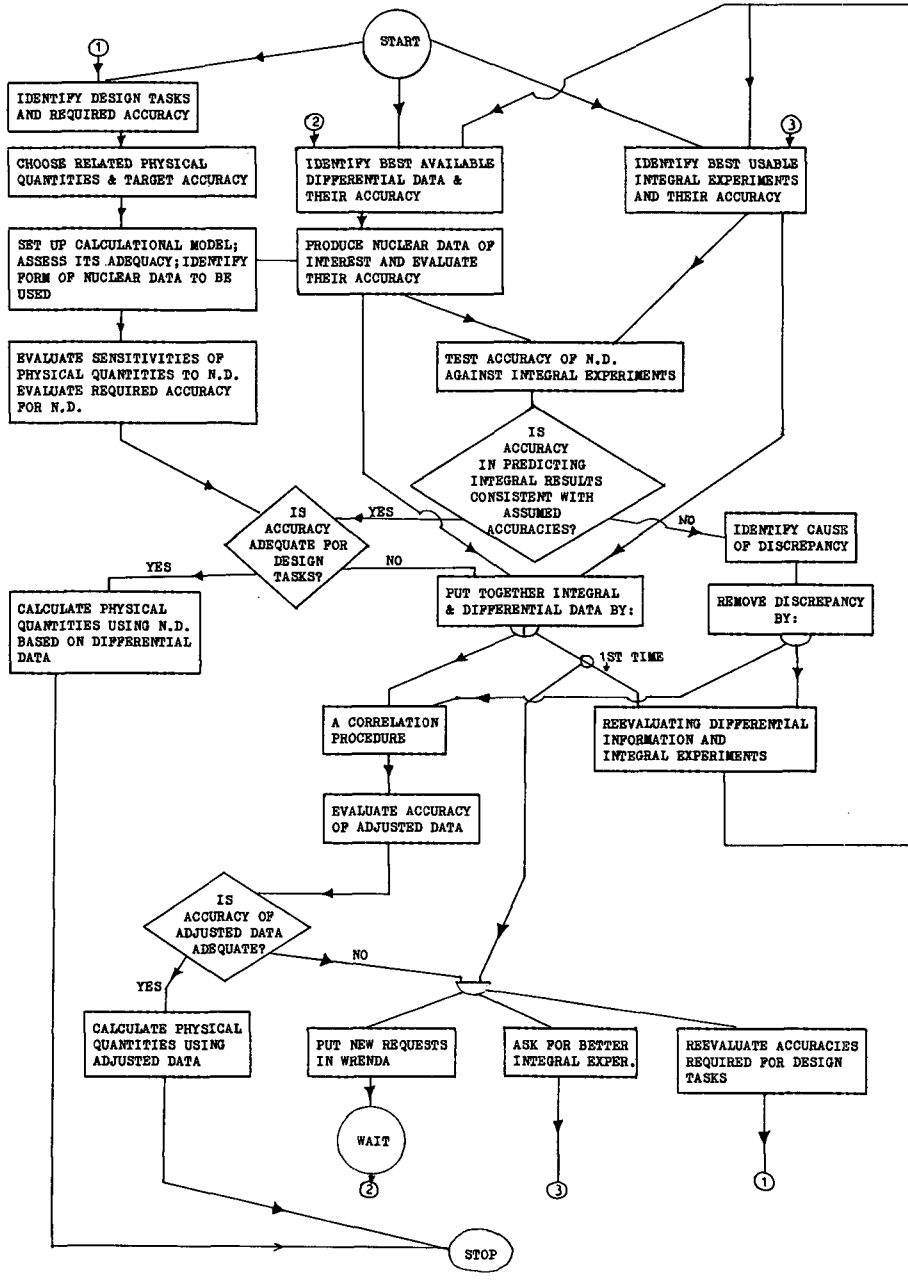

FIG. 1. Flow diagram for nuclear data processing.

component, which, at least in principle, could be obtained from different physical quantities.

The required accuracy for the design tasks, and therefore for the related physical quantities, is a function of many parameters: in a feasibility study or a conceptual study it may be determined by the uncertainty in other aspects of design, and requirements may not be strict; in the case of an actual, final design the evaluation is based on economical considerations and sometime on technological limits. For instance, a large uncertainty in the determination of the enrichment required for a given core life would involve the economic penalties associated with a higher inventory of fissile material and possibly with a more effective control system; on the other hand, it would be worthless to know this enrichment to a better accuracy that the fuel fabricator is able to comply with.

A complete economical balance would require, on the one hand, the cost one has to pay for a given error (or uncertainty) in the design data and, on the other, the cost of R & D needed to get the same accuracy. This is hardly possible in a general way, but some sort of a rough balance should be drawn in all cases, and it has been done in some.

The next step is to set up an adequate calculational model, which will in turn define the form of the nuclear data of interest. This choice has actually a great influence on balancing integral versus differential information; if a rather basic model is chosen, where nuclear data are in the form of detailed cross sections, then differential information is likely to play a more important role than in the case of a calculational model based on more integral quantities (think for instance to the role of effective resonance integrals, or of Fermi age in classical thermal reactor physics); in the latter case, which may involve uncertainties in the theoretical methods to deduce these nuclear data from differential measurements, direct integral experiments will tend to take the lead.

We then have to evaluate the sensitivities of the physical quantities to the nuclear data used for their calculation.

We usually define the sensitivity of a certain physical quantity (or design task) Q to a particular nuclear datum x as the variation of this quantity for a unit variation of the nuclear datum /5/:

$$S_x^Q = \frac{dQ}{dx}$$

The general problem of evaluating these sensitivities is by no means a simple one. Q may be a multiplication factor but it may also be a neutron lifetime, a breeding ratio, or the number of neutrons of energy higher than 1 MeV streaming out of a reactor shield; x may be the capture cross section of U-238 in a certain energy group, or a resonance self-shielding factor or any other form of nuclear data. Much progress has been made in the last years in this field, especially with the development of generaliz-

ed perturbation techniques (/5/, /6/) but certainly much remains to be done. From these sensitivities, one can infer the accuracy required for the various nuclear data. This is by no means a trivial task, especially if one takes into account the fact that errors in the cross sections cannot be treated as independent /7/.

If one has gone so far, one can now assess the adequacy of the differential and integral information in providing the required nuclear data. In doing so, it is customary and useful to check first the consistency of differential and integral information by calculating the results of integral experiments in the area of present interest by means of the "best" available cross sections. If there is consistency, the accuracy of the nuclear data obtained from differential data is compared with the requirements. In case these requirements are not met, one can try to put together integral and differential information in order to arrive at nuclear data which are better suited to the calculation of the particular physical quantities of interest. The ways of doing this can be very different. The two extremes are typified by the U.K. and U.S. approaches with fast reactor cross sections. In one case, a correlation procedure is used, in a more or less automated way, to "adjust" the differential data on the basis of the integral results, in such a way as to use information from all sources in a statistically correct way /8/, /9/. In the other, information from integral experiments is fed back to the evaluators as a guideline in a new choice of purely differential data /10/.

If, at this point, the requirements for nuclear data are not yet satisfied, one is again at a point of choice: one can file in a new request in the RENDA list to ask for better differential measurements (and this is in the best of cases a long-term solution); or he can try to have better or more ad-hoc integral experiments (which is in general a quicker procedure); or finally (as is often done) he can go back and look hard at his design requirements and see if he cannot loosen them somewhat.

Another situation that can arise is that there is inconsistency between differential data and integral experiments, in the sense that there are discrepancies between calculated and observed results sensibly larger than the uncertainties deriving from assumed errors in the cross sections and in the calculational methods. In this case the cause of discrepancy should be identified and then removed, either by going back to the differential measurements or by applying a correlation procedure.

Actually, the most widely expressed criticism of the adjustment procedures is based on the difficulty of assessing a priori the uncertainties associated with nuclear data evaluated from the differential information. It is a fact that it is more difficult to evaluate uncertainties in nuclear data than the nuclear data themselves. Also the distribution of uncertainties is generally assumed as Gaussian, which is reasonable if the errors are supposed to be of a statistical nature but it is not if systematic errors are involved. Then, there are correlations among the errors

in the different cross sections, according to the standards relative to which the measurements were performed, to the normalization criteria selected in the experiments or in the evaluation etc.

In the absence of detailed and reliable information on all these points, the correlation procedures tend to distribute the causes of discrepancies between measured and calculated cross sections among a very high number of nuclear data and thus to smear out the difficulties rather than to solve them. Such dilute corrections may have little physical significance and may conceal more real causes of discrepancies, which would make extrapolation of the adjusted data to different integral problems unreliable.

The present efforts in the area of correlation methods, (at least as far as fast reactors are concerned) try to overcome these criticisms along three lines.

First of all there is an effort to apply modern statistical theory consistently, in order to take into account in the most appropriate way all the information available and also to be able to estimate the reliability of the nuclear data thus obtained /9/, /4/, /11/, /12/.

Secondly, such methods are being investigated with regard to the possibility of identifying systematic errors in the cross sections and eventually correcting them (as we have hypothesized in our scheme).

Finally, available correlation methods are being applied with more insight into the ultimate sources of error, by taking into account the correlations between errors in the differential measurements, by preserving general shapes or characteristics of curves /13/, by tracking the origin of some cross sections back to specific nuclear models and adjust consistently the relative parameters /14/.

In other words, data adjustment methods should be considered as tools and not as rigid procedures. The possibility of changing the a priori uncertainties and of introducing supplementary conditions to describe the correlation among errors make these methods very flexible, and it is possible to verify or discard the hypothesis of a systematic error on any of the input data. The input conditions and the corresponding outputs should be scrutinized at all times and "physical judgement" applied to them. In this way adjustment methods become a powerful evaluation tool rather than empirical methods to deal with a limited range of specific situations.

3. Applications to Reactor Core Analysis

Most of what we have said in the previous paragraph is directly applicable to fast reactors. The application of a "Global Approach" to fast reactor core design is simpler and much more common than for other problems. This derives from the fact that the major source of uncertainties in most fast reactor calculations is considered to be nuclear data rather than calculational methods,

and also from the relative standardization of calculational methods, therefore of the form of nuclear data, in this area.

The first adjustment procedure, as far as I know, was adopted at Atomics International around 1960 in the frame of the Epithermal Thorium Reactor Program /15/. Methods for cross section adjustment were then rapidly developed in Italy /16/, /6/, the United Kingdom /8/, /9/, France /17/, Sweden, Israel, Japan and elsewhere. Use of integral experiments as benchmanrks to provide feedback to the evaluators of cross sections was widely employed in the same period in the USA in setting up the ENDF/B files /10/, in Germany and in other countries. Both differential and integral information is everywhere recognized to be necessary for the timely development of fast breeder reactors, although the way in which to take them into account varies widely from one place to another. In most cases, however, integral experiments both of benchmark and mockup type are carried out in support of specific fast reactor programs.

Concerning thermal reactors, the use of integral experiments in predicting their behaviour is as old as the reactors themselves. However there is a quite different relationship between these experiments and basic differential data with respect to the case of fast reactors.

The main reason for this is that the prediction of the integral parameters of thermal systems is affected at least as much by the approximations in the computational methods as by the uncertainties in the nuclear data.

Another important and related reason is that simple models are often assumed for the calculation of thermal reactors, (especially well thermalized systems) both concerning the neutron balance (like the four factor formula) and the neutron spectrum (the 1/E plus Maxwellian behaviour). These models define a number of quantities that have a simple physical meaning and that it is generally possible to measure directly in an integral experiment. Just think how much information of integral nature is included in a "classical" formula for k_{eff} such as:

$$k_{eff} = \frac{\epsilon p \eta f e^{-B^2\tau}}{1 + L^2B^2}$$

where p in turn is expressed as a function of the effective resonance integral and f is computed on data based on Westcott's formalism!

Of course, differential data are used for the predictions: but the respective role of differential and integral information is often reversed with respect to the case of fast systems. In particular, the main difficulty met in using the differential data is their reduction to the basic physical quantities that are used in the calculations. When a discrepancy is observed, it is by no means easy to decide whether it should be attributed to the nuclear data or to the calculational model. Whenever a reliable integral

experiment is available for a given quantity, its result is thus preferred to the corresponding result obtained by a calculation based on differential data, and this last is generally used as a check rather than a reference.

For instance, non-negligible discrepancies exist between the integral measurements and the differential calculations for such quantities as the Fermi age of fission neutrons in the most common moderators, and the resonance integrals of several important nuclides. The tendency in all these cases is to rely on the integral information.

The relatively smaller cost and greater ease of integral experiments in this field has made it possible to cover a wide range of elementary systems (either single rods or uniform lattices) of interest for thermal reactor calculations. Single parameters, as for instance the effective resonance integral of fuel rods as a function of their dimensions, and comprehensive sets of data for many systems are available and have in many cases been compiled in an easily usable form /18/, /19/.

The role of differential measurements has thus been a subsidiary one through most of the history of thermal reactors, and most thermal power reactors are designed as of today on the basis of semiempirical simplified methods, adjusted on critical experiments and especially on the data from operating reactors.

A tendency to use more basic methods and data has emerged from time to time in different laboratories and organizations, as a consequence of the availability of sophisticated transport methods for space dependent neutron thermalization and of evaluated differential cross sections in the thermal and epithermal energy range; however in most cases and for most reactor types basic cross sections display an unsatisfactory performance, diffusion calculations are better than transport calculations, kitchen recipes have the lead over a truly scientific approach.

The reasons for this situation are largely a question of time scale: once the feasibility of a large power reactor has been demonstrated, the development of other reactors of the same basic type is very much a problem of step-wise extrapolation, and no commercial pressure is put on the development of more basic methods. Removing the discrepancies that are still present is considered of a minor importance, once the full scale reactor has shown which is the physical answer. Even in well proven reactors such as light water reactors such discrepancies continue to exist for instance in the calculation of void coefficients, and no substantial effort goes into resolving them.

The only fields in which basic methods and cross sections are of common use are those of more advanced systems (like HTGR's) or of relatively new applications in other systems (as for instance new types of burnable poisons), and this only as long as adequate integral or operational information is not available.

One lesson should be learnt from these considerations on thermal reactors. Reactors with a commercial incentive will be

built whether or not we provide the refined nuclear data, whether based on differential or on integral information. This will cause at first some economic loss, but feasibility is no longer at stake. All our efforts to arrive at evaluated, consistent sets of data must be confronted with the time-scale of the actual technological development - a time-scale which is largely independent of our efforts.

4. Shielding

The situation of nuclear data for shielding is probably worse than for core design. Requirements for shielding differ from other requirements for several reasons: higher energy range, interest in cross section minima rather than in maxima, different materials, interest in the gamma rays generated by capture and inelastic scattering, importance of small angle scattering data, etc. Priority in measuring programs has been given so far to data for core design applications and only recently have specific differential measurements been started for shielding design purposes. As a consequence, present differential data are not considered adequate by shield designers.

Integral experiments have been extensively used in the past to support shield calculations. The most successful removal-diffusion methods, for instance /20/, have been based on removal cross sections obtained from experiment.

An assessment of the possible systematic role of integral and differential measurements is only at its beginning. The reasons are that a number of difficulties are present in this task. First of all, both cross sections and calculational methods produce relevant uncertainties in actual shield calculations. Although bulk shields dealt with by refined transport methods are more subject to errors due to cross sections and highly heterogeneous shields to the calculational methods and to the geometrical representation used, both sources of uncertainties are always present.

Second, sensitivity studies are not available except for special cases. The reasons for this are, in turn, the great variety of problems met in shield design, the high problem-dependence of the cross sections, the difficulty of applying generalized perturbation theory to shielding problems. Some of these difficulties have been dealt with recently /21, 22/. It seems probable that in the next future we shall witness a mushrooming of studies in this field.

Even before sensitivity studies will yield us a comprehensive view of the data requirements for shielding, the discussion on the possible role of integral experiments has reached its climax. The Summary Report of the ENEA/IAEA Specialist Meeting on Shielding (December 1970) /23/ states that "it is estimated in the U.K. that... data will need adjustments against benchmark experiments because the target accuracy is unlikely to be met in the next decade by differential measurements. This contrasts with the current

view in the USA where it is envisaged that all the important cross sections will be measured in the foreseeable future".

This contrast appeared somewhat attenuated at the recent Shielding Conference (Paris, October 1972) /24/ where Monte-Carlo calculations based on differential cross sections were considered by the U.K. representatives as an alternate approach in generating the "empirical" cross sections to be used in adjusted-diffusion calculations. The use of integral experiments for data adjustment was, however advocated by other participants, notably from France.

Benchmark type experiments are meanwhile carried out in several countries. A typical experiment consists in studying the propagation of neutrons from a well-known source in a large homogeneous block of a material of interest, and measuring the neutron spectra at various distances from the source. What will be the ultimate use of such experiments remains to be seen. The situation can be easily reconducted to our scheme of Fig.1. The integral experiments can be used as a check of the adequacy of the cross sections plus calculational model, or to identify possible discrepancies to feed back to the evaluators or finally to adjust the cross sections. Experiments of this kind are very sensitive to the features of the cross sections that are interesting for shielding purposes. The transmitted neutrons at or below the energies of cross section minima actually vary so much with small variations of these cross sections that the application of first order perturbation theory is generally not adequate. It is therefore tempting to use these experiments as direct sources of information. The applicability of results of such measurements to other systems (in particular those with strong heterogeneities) will have to be adequately scrutinized.

Which will be the actual solution adopted in the various countries will largely depend on the time scale imposed for the solution of new shielding problems on the one hand and on the production of differential data specifically intended for shielding on the other.

5. Radiation Damage and Activation Measurements

A similar situation is met with respect to radiation damage. Two parallel lines can be followed when studying, for instance, the damage life of a structural component. One of the main concerns, in this case, is the comparison of damage rates in different neutron spectra, so as to be able to predict material performance in a radiation field of characteristics different from those of the environment in which damage experiments where carried out. The differential approach here consists in assuming some basic model for the production of damage (such as for instance the Linhard model for steel /25/) and calculate on the basis of this model and of pointwise scattering cross sections for the nuclides involved a detailed damage function /26/. The integral approach consists in generating directly a few group damage function by considering some radiation damage experiments in different neutron spectra /27/.

These studies are relatively new (former investigations both of differential and integral type were based on much coarser approximations) and although some comparisons between results from the two methods have been carried out, it is too early to draw conclusions. However in principle the same situation as for core or shield neutronic calculations may arise in this field, and it is perfectly possible that adjustment procedures are developped to correct multigroup damage functions, originated from basic models, on the basis of the results of integral experiments.

A related problem is the one of activation detectors, which are widely used in many areas of reactor physics, but specially in order to obtain spectrum and fluence information to be used in connection with irradiation experiments.

A long tradition of integral experiments in different neutron spectra exists for these detectors; the differential information on them was until quite recently scarce or contradictory, so that integral experiments were essential for the normalization of cross sections and in some cases also for shape assessment.

A coordinated effort of differential measurements and evaluation has been started recently in several countries and at the IAEA /28/. We shall see in the near future whether this effort will meet the requirements of the users or whether further feedback from integral measurements will be needed.

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DISCUSSION

Z. SZATMÁRY: If you adjust cross-sections, there are several factors which affect the adjustment: errors in the cross-section measurement, errors in the calculational model, errors in the integral measurements and perhaps others. In your view, which of these is the major factor? Also, do you see any reasonable way of separating the effects of these factors?

U. FARINELLI: The relative importance of the three factors you mention depends very much on the problem considered. For instance, it is generally assumed that in fast reactor core calculations the uncertainty due to the cross-sections is predominant. In a typical shielding problem,

the uncertainty may be split equally between cross-sections and calculational methods (or rather, the representation of the system in the calculations). In such cases, benchmark experiments of very simple geometric description (such as neutron propagation in a large block of a single material) may be chosen to enhance the contribution of cross-sections with respect to calculations.

Concerning the second question, modern adjustment procedures try to take into account all the possible sources of error in a statistically correct way and especially to verify a posteriori the consistency of the adjustments performed, although in practice this is a difficult job.

W. W. HAVENS, Jr.: The history of reactor development shows that both differential and integral measurements are necessary. Discrepancies between the results of integral measurements and the same results calculated from differential measurements have lead to some of the most important improvements in the reactor field. The discrepancy requires both groups to re-examine their measurements carefully to determine the cause of the discrepancy. In many cases finding the cause of the discrepancy has led to major advances in the field as a whole.

U. FARINELLI: I fully agree with you.

W. B. LEWIS: Would Dr. Havens agree that it is the models rather than the measurements that have improved?

W. W. HAVENS, Jr.: In my opinion both the models and the measurements have improved considerably.

L. HJÄRNE: Drs. Farinelli, Lewis and Havens have touched upon an important topic of rather general nature. The evaluation of data is of course a very complex undertaking. The experimental data are evaluated, first, independently of the area of application and, secondly, with an orientation towards application (referred to also as "adjusted sets"). We certainly need both kinds. Between data sets for different purposes we might very well find different values for one and the same quantity. One reason for this can be found in the differences in importance of data for different applications. For different applications, different parameters are varied and different criteria (e. g. consistency criteria) are applied in order to minimize the chances of errors or penalties in the end results of the applications, i. e. when calculations are made with the data. This illustrates the need not only for application-independent evaluations but also for what I should like to call "repackaged" data compilations/evaluations. This is probably true for many kinds of data and their applications.

G. A. KOLSTAD: You have touched upon an important area of confusion in the nuclear data field. If the evaluation of nuclear data through the use of integral information does not lead to "better values" of the cross-sections, then it follows that one should not conclude that, when experiments begin to yield the same results as those given in such evaluated data files as ENDF, no further experiments are needed. "Best values" for cross-sections, e. g. nuclear standards, should therefore be based only on careful differential experiments. Do you agree?

U. FARINELLI: I agree with your conclusions. In particular, I believe that one important reason for having well-known standard differential cross-sections is to simplify analysis of the correlation of errors in other cross-sections.

G. A. KOLSTAD: Is it reasonable to develop a set of application-independent evaluated cross-sections?

U. FARINELLI: I think that a "universal" evaluated nuclear data file is certainly possible in principle but at least for the moment it is not practical.

R. NICKS: I should like to make some comments about nuclear cross-sections and shielding.

First, the complex geometries involved in most shielding problems can be handled by newly developed 2- and 3-dimensional transport codes, so that uncertainties due to geometry can be greatly reduced. More generally, the availability of sophisticated shielding codes leads to an escalation of data requirements. I am somewhat skeptical of the attribution of equal weights to nuclear data uncertainties and errors due to other sources.

Second, in shielding one needs differential cross-sections, and more particularly, gamma production cross-sections.

Third, for deep penetration problems, the accuracy required may be of the order of 1% for some isotopes. I wonder if this is feasible experimentally at the present time.

J. T. BARRE: It would be interesting to hear the views of users, i. e. people who really make reactor calculations, on this basic point. The speaker and I belong to the "integralist" party. Are there any other supporters in the hall?

G. CASINI: I should like to make a few remarks on the choice of integral experiments for nuclear data testing. In the past the "integralists" committed an error by failing to indicate which integral experiments are suitable for nuclear data testing. Many calculations relating to such experiments were unproductive because the geometrical configurations were too complicated or the experimental data supplied by laboratories were insufficient.

To help clarify the situation, I think a limited number of experiments should be designated as suitable for checking nuclear data in different fields of application. These are the so-called "basic, clean" or "benchmark" experiments. An example of this approach is the list of 12 benchmark experiments chosen by the United States for testing fast breeder cores. The effort should be that of trying to implement these types of tests, if necessary with a number of European experiments, and then of concentrating efforts on checking the data from these experiments.

Such endeavours should be concentrated mainly in new areas where the need for clean integral checking of nuclear data is becoming important, e. g. in reactor shielding and fusion blanket problems.

D. R. HARRIS: There seems to be some agreement, then, that integral and differential measurements and evaluations are mutually helpful. If this agreement is to have practical significance, the need for a balance of funded effort is indicated. There is one subject which has been discussed at this conference where such a balance has been lacking. I am referring to the matter of decay heat following potential loss-of-coolant accidents in reactors, for which integral measurements are very much needed. In this area the funded support for differential measured nuclear data in aid of nuclear physics has been disproportionately greater than the support for integral experiments in aid of applications.

P. RIBON: The "differentialists" are convinced of the value of adjustments as a means of obtaining a satisfactory description of integral experiments and a better design of projects. The problem is to what extent

account should be taken of these adjustments in making evaluations, i.e. to what extent their significance must be known from the point of view of nuclear data.

A number of adjustments have proved to be significant and have confirmed (or been confirmed by) recent differential data determinations. It would be useful to be able to compare the adjustments made by different laboratories, taking identical differential data as their starting point but working independently of one another (different programs, different types of experiment analysis, etc.).

U. FARINELLI: I believe that one should distinguish between two possible results of the application of adjustment procedures. In the case where all errors in the original cross-sections are assumed to be of a statistical nature, adjustment procedures tend to smear out the correction over a large number of data. Although the results are statistically correct, their value is limited essentially to applications, and they may have little significance from a "fundamental" point of view; in this case, the corrections are likely to depend on the particular objective of the adjustment and a comparison of different cases is probably not very meaningful. The other possibility is that adjustment procedures result in the identification of a discrepancy (i.e. many differences between calculated and measured integral parameters can be explained by a single change in a cross-section of a quantity much larger than the originally assumed error). This can be found by most adjustment procedures, but methods specially suited for the purpose are being developed. In this case, a comparison of the results obtained by the various groups is essential; furthermore, these indications should be fed back to the evaluators and originators of the differential measurements for the identification of possible causes of systematic errors.

H. GRUPPELAAR: I would merely point out that there are cases in which integral measurements are in fact the only measurements which are possible in actual practice. For instance, if targets are highly radioactive, differential cross-section measurements are hardly possible unless one is willing to apply the nuclear explosion technique. Another example is measurement of the cross-section of an isotope for which not enough enriched target material is available.

J.J. SCHMIDT: At present a number of adjustment procedures are being used by various reactor groups. I wonder whether Mr. Farinelli could comment on the extent to which nuclear physics knowledge and considerations are taken into account in the adjustments? There are examples in which adjustment has led to physically incorrect data, such as an increase, instead of a decrease, in a capture cross-section when passing an inelastic scattering threshold. He himself referred to these examples when he mentioned correlation between nuclear data values at different energies.

U. FARINELLI: Most adjustment codes have the capability of accepting supplementary conditions, such as correlation among different corrections, preservation of certain features of the original data etc. As I point out in the paper, these codes should be regarded as flexible tools rather than as fixed routines. The problem now is how much fundamental information (such as that deriving from nuclear physics) is actually incorporated in the adjustment procedure. There is evidence that the quantity (and quality) of such information used is rapidly in-

creasing. I know of recent adjustment work in which different confidence values have been assigned to the shape and to the normalization of the Pu-α value, or others (in progress), in which an excited level for inelastic scattering or a nuclear model for evaporated neutrons is specifically taken into account. All these things are possible and I think there is a definite trend towards introducing nuclear physics into these correlation procedures.

T. FUKETA: There have now been many fairly favourable comments on the adjustment of cross-section data by integral measurements. However, I think there is a similarity between the systematic error in the differential type of experiment and the effect of integralists' model-system calculations on the adjusted data. If this comment is valid, what we need to have are more independent adjusted data so that we can recognize and evaluate the scattering among those data, which the individual investigator might not foresee.

THE USE OF NUCLEAR DATA IN THE MONTE-CARLO CODES GEM AND MONK IN REACTOR PHYSICS AND CRITICALITY CALCULATIONS

J.G. MOORE
Safety and Reliability Directorate,
United Kingdom Atomic Energy Authority,
Culcheth, Nr. Warrington, Lancs, United Kingdom

Abstract

THE USE OF NUCLEAR DATA IN THE MONTE-CARLO CODES GEM AND MONK IN REACTOR PHYSICS AND CRITICALITY CALCULATIONS.

This paper gives a description of the nuclear data used in the Monte-Carlo codes GEM and MONK, and its use in reactor physics and criticality calculations. The nuclear data are based on the UK Nuclear Data Library and because of the importance of the total cross-section in the calculation, no approximations are made to the original library data. The representation of total cross-section, therefore, is in the form of a table of point values at specific energies, unlike most codes which have group representation. Group constants however are used for the relative probabilities of the partial cross-sections.

There are two main approximations in the use of nuclear data in GEM and MONK: there is a single energy group below 0.1 eV and unresolved resonances are not considered. Calculations on critical systems have shown that the multiplication factor, k, usually lies within about 5 per cent of unity. It is possible that this spread in the multiplication factor could be due to these approximations. Calculation of reaction rates in low-power reactors has shown agreement within about 10 per cent of experimental results. It is concluded that, in spite of the approximations in the nuclear data, the use of the UK Nuclear Data Library gives reasonable agreement with experiment. It is recommended at present, however, that sufficiently large safety factors should be incorporated in the predictions of critical mass until the nature of the discrepancies is understood. Future work is aimed at removing the approximations in the nuclear data.

INTRODUCTION

The potential of the Monte Carlo method has long been appreciated but its implementation has not always been feasible, usually because of limitations of available computer facilities. These limitations have usually manifested themselves either as computer storage problems or excessive computer time required to achieve the desired accuracy. The Monte Carlo method attempts to solve the neutron transport problem with little approximation to the nuclear data or geometry and it is inevitable that this produces a computer storage problem.

A Monte Carlo method may be required to check an approximate method of solution to the transport equation as used in diffusion and transport codes. It has been used in some instances where great detail is required in the nuclear data such as in the calculation of the Doppler effect in nuclear reactors [1]. In the UK, Monte Carlo techniques have been most frequently used in the solution of criticality problems. In many reactor physics calculations the effects of approximations in geometrical and in nuclear data are relatively well understood whereas in criticality work the diversity of chemical and metallurgical plant gives only limited opportunity to build up sufficient experience to enable approximations to be made with confidence. It is for this reason that Monte Carlo techniques

are often used in the criticality field. The use of Monte Carlo techniques have so far found only limited application to reactor physics problems unless they have been written for a specific application. This is because reactor physicists are quite often interested in reactivity differences between different reactor configurations rather than the absolute values and it is difficult to achieve the required statistical accuracy by Monte Carlo methods. However, this difficulty can be overcome to some extent by the use of perturbation techniques. Monte Carlo could however be used where reaction rates are of interest since it is often feasible to calculate these to an acceptable accuracy.

In the Monte Carlo codes GEM [2,3] and MONK [4] the basic philosophy was to provide codes with as little approximations in their treatment of geometry and nuclear data. The errors due to geometrical arrangement can be made negligible in both codes though this is somewhat easier in MONK. At the time these codes were written the intention was to use the most detailed information that was available on nuclear data. In the UK this was represented by the UK Nuclear Data Library. The contents of the library as used in the codes are described in reference [5] and the format of the data in reference [6]. In effect the library is used without approximation with typically several thousand energy points for each nuclide. Two approximations had to be made owing to lack of data or means of prediction; this situation has now changed to some extent. At the time the codes were written there was no satisfactory method for sampling the thermal scattering laws, nor was there any data, so a single energy group for neutrons below 0.1 eV was employed in GEM and MONK. In addition the prediction of resonances in the unresolved resonance region was not well established so the actual values of the cross-sections from the library were used.

There are two benefits however from the use of the UK Nuclear Data Library in the way in which it is used in GEM and MONK. Firstly all calculations are done with the full library structure so there is no need to consider spectrum averaging in order to produce 'fewer groups'. Secondly there is no need as in most other codes for particular attention to be payed to resonance self shielding as this is automatically accounted for if the resonance shape is represented in the cross-section data.

The rest of the paper describes some of the more detailed aspects of the nuclear data used in GEM and MONK and some of the experience gained with these codes.

THE FORM OF NUCLEAR DATA USED IN GEM AND MONK

A detailed description of the way nuclear data is used in GEM and MONK may be found in References [2], [3] and [7]. The following sections will highlight the important features in these reports.

1. Thermal Data

In GEM and MONK it is assumed that above 0.1 eV the kinetic energy of the nuclei of the medium at a given temperature is negligible compared with the kinetic energy of the neutron. When the energy of the neutron is less than 0.1 eV the thermal motion of the nuclei should be considered in the interactions with neutrons. In GEM and MONK neutrons may be absorbed, scattered isotropically or cause fission. A single energy group is used with associated group constants. The current values are based on those given by Norton [8]. It is recognised that the use of Maxwellian

averaged values is an approximation as most thermal neutron density distributions are rarely similar to a Maxwellian distribution. Also since inelastic scattering is not allowed, molecular binding effects are largely ignored. In order to compensate for these approximations the scattering cross-section was calculated from the measured diffusion lengths for different materials and the Maxwellian averaged absorption cross-sections. This enabled binding effects to be taken into account approximately.

The nuclear data used in general corresponds to 300°K but for calculations at higher temperatures different sets of data incorporating Doppler broadening of resonances and modified thermal constants are required.

2. Microscopic Total Cross-section

Nuclear cross-sections and energies are given in pairs in the Nuclear Data Library such that linear interpolation on a log-log scale introduces acceptably small errors. GEM and MONK interpolate between the points given in the library and in this way the total cross-section is as accurate as the main library data.

TYPES OF COLLISION AND THEIR DETERMINATION

Six types of collision are possible:

(i) elastic scatter
(ii) absorption
(iii) fission
(iv) (n,n') or inelastic scatter
(v) (n,2n) reaction
(vi) (n,3n) reaction

The particular reaction is identified by sampling the ratio of the partial cross-section to the total cross-section. The ratio of the partial to the total cross-section does not vary with energy as much as the total cross-section does so it is a reasonable approximation to represent this ratio as a step function. The POND4 [7] program calculates the energy boundaries for each step function in order to minimise errors.

NUMBER OF SECONDARY NEUTRONS

The mean number of neutrons per fission, $\overline{\nu}$, is represented in the data library in a similar way to cross-section. Linear interpolation is used between points in the library. The number of secondary neutrons released in a fission, is an integral random variable whose expectation is equal to $\overline{\nu}$.

DETERMINATION OF SCATTER ANGLES

The angle of scatter, θ, is obtained by sampling the angular distribution given in the data file which is given as a frequency of probability against cosine of the scatter angle. In general when the distribution is anistropic there are two procedures which may be adopted. The first is the discrete scatter angle approximation: POND4 produces 32 equiprobable values of θ, one of which is chosen at random. The second method is to approximate $\cos\theta$ by a truncated cubic polynomial in Z, the random number, and has been found by trial to give a close representation of the angular distributions which occur.

CALCULATION OF SECONDARY ENERGIES

When it is not possible to calculate the emergent neutron energy from the scatter angle, secondary energy laws are provided by the Nuclear Data Library.

1. $E' = E_d$
2. $E' = k(E-E_d)$
3. E' is obtained from a probability distribution independent of E
4. E' is obtained from a probability distribution dependent on E
5. E' is obtained from a probability distribution dependent on E and $E'/\sqrt{E}$
6. E' is obtained from a probability distribution dependent on E and E'/E
7. E' is obtained from an evaporation spectrum given by

$$q(x) = ax \exp(-x\sqrt{a})$$

where $x = E'/\sqrt{E}$

A neutron of a given energy has a probability of obeying any of these laws so this probability has to be sampled in the first instance. When the type of law is decided the secondary energy, E', is sampled for the probability distribution given in the data library.

FISSION SPECTRUM

It is assumed in the computer codes that the same fission spectrum applies to all nuclides and is independent of incident energy i.e.

$$f(E)dE = 0.4527 \exp(-E/0.965) \sinh(\sqrt{2.29E})dE$$

the well known Rosen-Cranberg spectrum.

CRITICALITY CALCULATIONS

A good deal of the criticality assessment work which has been done in the UK in the past has been based on calculations done by GEM and MONK. The reasons for this have been that little approximation needs be made to the geometry and the nuclear data has been based on the UK Nuclear Data Library. Unlike many computer codes no approximations were made to the original library data by group reduction procedures. When such principles are adopted it is generally found that the group cross-sections invariably have to be adjusted to give reasonable agreement with critical experiments. In spite of this the group libraries produced are usually applicable over particular energy ranges and it is quite common in reactor physics work to find different group libraries for reactor systems of different neutron spectra. This procedure has obvious disadvantages in criticality work where one can be dealing with systems which span the whole energy spectrum. However GEM and MONK are available as codes with universal application because of the absence of such group reduction procedures. As we shall see later some improvements still need to be made.

Nevertheless although GEM and MONK have great potential it has always been the policy in the UK to supplement this by calculations on critical systems of as varied a nature as could be found in the literature, and by this means confidence in the predictions of criticality can be achieved. Most of the work done to date has been done on GEM since MONK is still a relatively new code. However since the Nuclear Data Library has been changed little since the GEM calculations have been done, the experience obtained from the nuclear data in GEM can be carried over to MONK. Some recent results on MONK are however included to confirm the general conclusions. The values of the multiplication factor, k, given in the following tables have a standard deviation of approximately 1% associated with them. The values of k calculated by GEM are based on a cycle of tracking known as boundary B tracking [9]. The values of k from MONK calculated in Tables III and IV are for boundary B tracking whereas those in Table V were obtained from a fission to fission cycle of tracking. It is not within the scope of this paper to discuss the relative merits of these tracking cycles. However for a critical system k will be unity by both methods. In the critical systems described in this paper k is usually

TABLE I. CRITICALITY CALCULATIONS DONE WITH GEM ON URANIUM SYSTEMS

	Core Description	Reflector Description		
U3	U(93.15) metal cylinder	None	ORNL	1.0190
U2(1)	U(93) metal sphere	Thick water	LASL	0.9674
U11	U(93.9) metal sphere	10 cm uranium	LASL	1.0006
AM1	U(38) metal cuboid	Thick polythene	AWRE	1.0075
US11	U(93.4) solution H/U = 55	Thick water	ORNL	1.0080
US21	U(93.4) solution H/U = 466	Thick water	ORNL	1.0130

TABLE II. CRITICALITY CALCULATIONS DONE WITH GEM ON PLUTONIUM SYSTEMS

	Core Description	Reflector Description		
SAP3	Pu metal sphere	None	AWRE	0.980
SAP5	Pu solution sphere H/Pu = 980	30 cm water	Hanford	1.011
SAP1	Pu sphere	12.65 cm water	AWRE	1.020
SAP10	Pu solution sphere H/Pu = 87	25 cm water	Hanford	1.026
SAP7	Pu solution sphere H/Pu = 140	$\frac{1}{2}$ in	Hanford	1.036
SAP4	Pu solution sphere H/Pu = 172	30 cm water	Hanford	1.024
SAP8	Pu solution sphere H/Pu = 1158	None	Hanford	0.976

TABLE III. CRITICALITY CALCULATIONS DONE WITH MONK ON PLUTONIUM SYSTEMS

Core Description	Reflector Description		
PuO_2/Polystyrene compacts H/Pu = 0.04	Plexiglas	Hanford	1.0208
PuO_2/Polystyrene compacts H/Pu = 15	"	"	1.0177
PuO_2/Polystyrene compacts H/Pu = 50	"	"	1.0372
JEZEBEL Pu sphere 4.5% Pu240	None	LASL	0.9741
JEZEBEL Pu sphere 20.1% Pu240	"	Hanford	0.9479
Pu nitrate slab H/Pu = 57 Pu240 = 4.6%	Water	"	1.0456
Pu nitrate solutions spheres H/Pu = 87 Pu240 = 4.6%	"	"	1.0491
Pu nitrate solutions spheres H/Pu = 244 Pu240 = 4.6%	"	"	1.0248

TABLE IV. CRITICALITY CALCULATIONS DONE WITH MONK ON URANIUM SYSTEMS

Core Description	Reflector Description		
Godiva U Sphere 93.6% U235	None	LASL	0.9880
2 x 2 x 2 Array Metal Cylinders 93.2% U235	Paraffin Wax	ORNL	0.9953
Uranyl Nitrate Solution H/U 40 93.2% U235	None	REP	1.0262

within 5% of unity, so that differences between the two tracking schemes will produce on average a systematic difference of about 2% on k. The purpose of this paper is to make deductions about the nuclear data from the general trends of the results rather than from the absolute values.

Tables I and II show some representative results obtained with GEM [10] on uranium and plutonium systems with different reflector conditions. The details of the experiments may be found in Reference [11]. It can be seen that in spite of the variety of systems, which include comprehensive range of hydrogen to fissile material ratio and reflector properties, that k lies mainly with $\pm 2\%$ or two standard deviations as one would expect from purely statistical arguments. However a few of the results deviate by up to 5% from unity and a few have even greater deviations. Tables III and IV give results by MONK on critical systems [12]. It can be seen that these results show the same general trends as the GEM results.

TABLE V. CALCULATIONS OF k OF VARIOUS CRITICAL CORES

Description of Core	Source	H/Fissile Ratio	k
^{239}Pu metal/water spheres water reflected	LA 3612	0	0.9471
"	"	1	0.9659
"	"	3	0.9645
"	"	10	0.9743
"	"	30	0.9978
"	"	30	1.0203
"	"	100	1.0460
"	"	100	1.0222
"	"	300	1.0279
"	"	500	1.0147
"	"	1000	0.9968
"	"	2000	1.0007
^{239}Pu metal/water spheres, unreflected	"	0	0.9798
"	"	30	1.0049
"	"	500	1.0084
^{235}U (93.5) metal/water spheres, water reflected	"	0	0.9659
"	"	9.96	0.9357
"	"	30	0.9999
"	"	100	0.9826
"	"	300	1.0070
"	"	1500	0.9837
SCAMP, Composition A, 25 cm Core	CRIT/OP /39	238	1.0521
30 cm Core	"	238	1.0615
37.5 cm Core	"	238	1.0557
50 cm Core	"	238	1.0468
SCAMP, Composition B, 30 cm Core	"	844	1.0140
37.5 cm Core	"	844	1.0221
50 cm Core	"	844	0.9943
SCAMP, Composition C, 37.5 cm Core	"	1453	1.0152
50 cm Core	"	1453	1.0234

These trends have also been confirmed by recent calculations done by Parker [13] using MONK for plutonium-water and uranium-water systems with varying reflector conditions. The results are shown in Table V. Parker has plotted k against hydrogen to fissile material ratio for the plutonium-water systems, [Figure 1]. The solid line represents the best fit to the data. It can be seen that MONK underestimates k by about 5% in the unmoderated region and overestimates k by a similar amount in the region of optimum moderation. For the overmoderated cores where the predominant

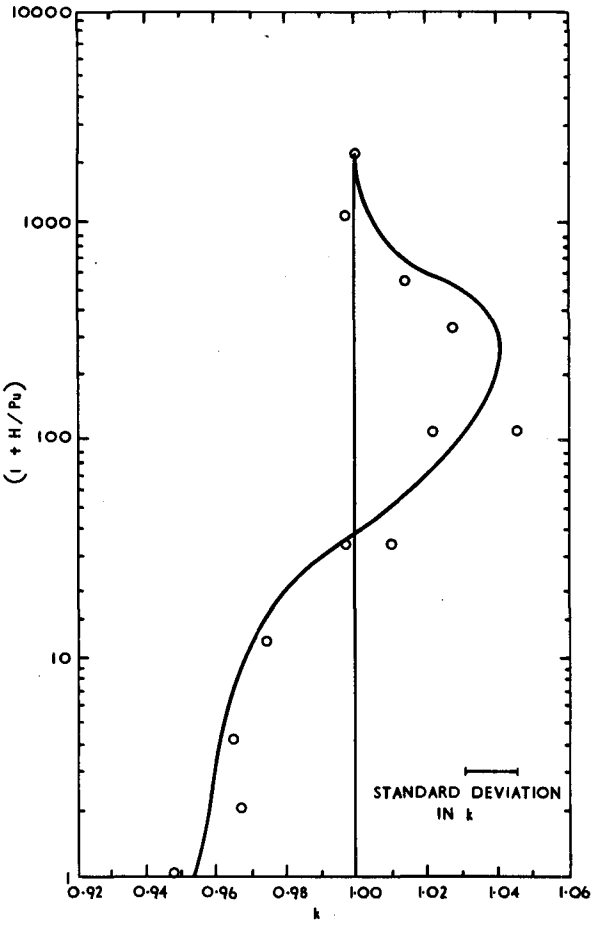

FIG. 1. Fully reflected plutonium/water systems, plot of k versus hydrogen/plutonium ratio.

effect is absorption by hydrogen k approaches unity. Figure 2 shows a similar trend for the variation of k against hydrogen to fissile material ratio for some unreflected plutonium-water systems, fully reflected 93.5% U235 water systems and the SCAMP results.

DISCUSSION OF CRITICALITY RESULTS

The deviations from unity which have occurred in these results cannot all be explained by statistical variations. In the past when such discrepancies have occurred a closer look at the experimental data or its interpretation has frequently provided plausible reasons for some of the discrepancies. However, there remain a few systems with discrepancies larger than can be reasonably explained by these means. The only remaining possibility, apart from errors in the code, must be the nuclear data.

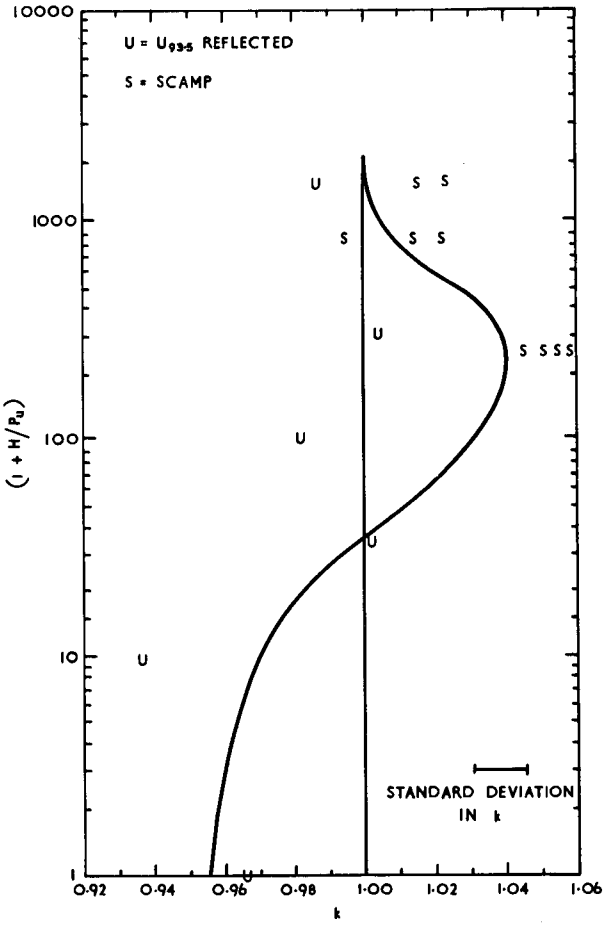

FIG. 2. Plot of k against hydrogen to fissile ratio.

There are two possibilities; either there are errors or omissions in the Nuclear Data Library, or the treatment of nuclear data in MONK introduces errors. Improvements in the Data Library can only be made as better experimental evidence becomes available and suitable evaluations done. In GEM and MONK it is possible that the omission of the prediction of unresolved resonances may account for some of the discrepancies but as yet no final deductions have been made. In the optimum moderation region it is possible that the single thermal group representation below 0.1 eV does not give an adequate representation of the neutron behaviour. This deficiency could be removed from MONK as data is now available in the WIMS data library [14] for the thermal scattering laws and the unresolved resonance region. This could easily be achieved by having group representation of the nuclear data in these regions in MONK and using the WIMS library data. A modified form of neutron tracking would need to be incorporated in MONK for the group representation. A version of MONKG has

already been written and tested which uses group data but without upscatters; with some modification this could be made to give upscatters in the thermal region [15] and give a better treatment of thermal neutron scattering.

The other main approximation in the nuclear data used in MONK is in the representation of the unresolved resonance region. The code GENEX [16] does provide an independent evaluation of the unresolved region but the amount of data produced is so large that it would create an impossible storage problem. The best compromise would probably be to use the 2000 fine group library [17] available at Winfrith which has basically a group structure based on GENEX and the Nuclear Data Library.

REACTOR PHYSICS CALCULATIONS

Monte Carlo methods are most frequently used in reactor physics calculations where it is necessary to simulate complexity in either the geometry or the nuclear data; or to check approximate solutions to the transport equation. Another field of important application is the calculation of perturbation effects e.g. the calculation of Doppler coefficient, control rod worths and small reactivity effects [18].

GEM and MONK have been used to their full capability in the calculation of flux distributions and reaction rates in complex geometries. Figure 3 shows a comparison of GEM calculations with experimentally measured reaction rates for an early core assembly of ZEBRA. The core had a typical fast reactor composition complete with breeder. The calculated results in GEM have been normalised to the measured reaction rate of U235 at the centre. The standard deviation associated with the theoretical curve is typically a few percent. As can be seen the agreement is reasonably good. Figures 4 and 5 show the comparison of reaction rates for other nuclides. Thus the nuclear data used in GEM appears adequate in a typical fast reactor spectrum.

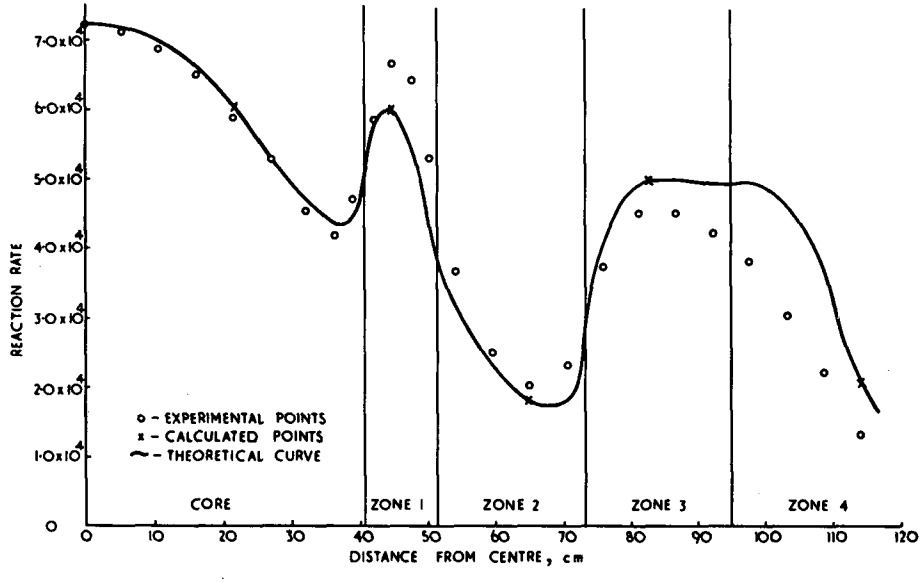

FIG. 3. Radial ^{235}U (nf) reaction rate.

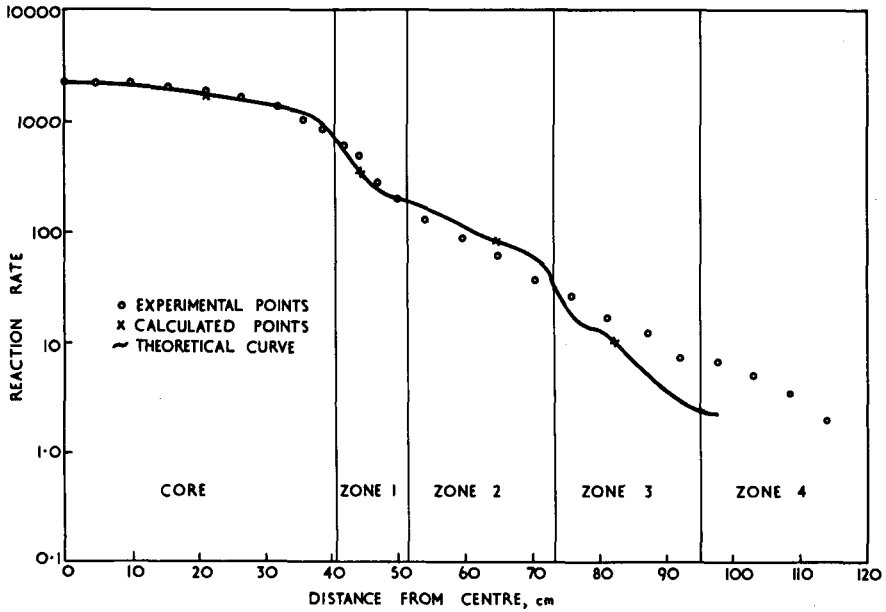

FIG. 4. Radial ^{238}U (nf) reaction rate.

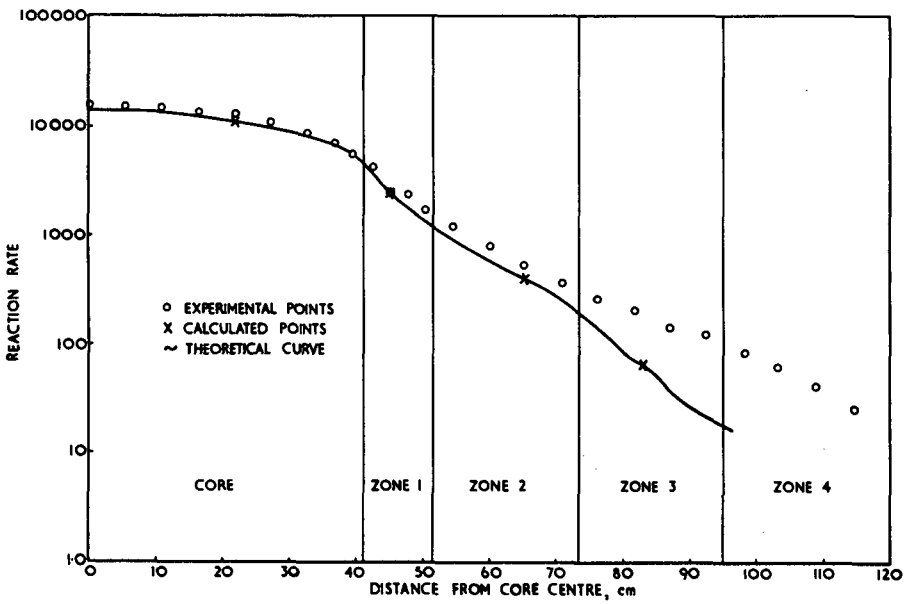

FIG. 5. Radial ^{237}Np (nf) reaction rate.

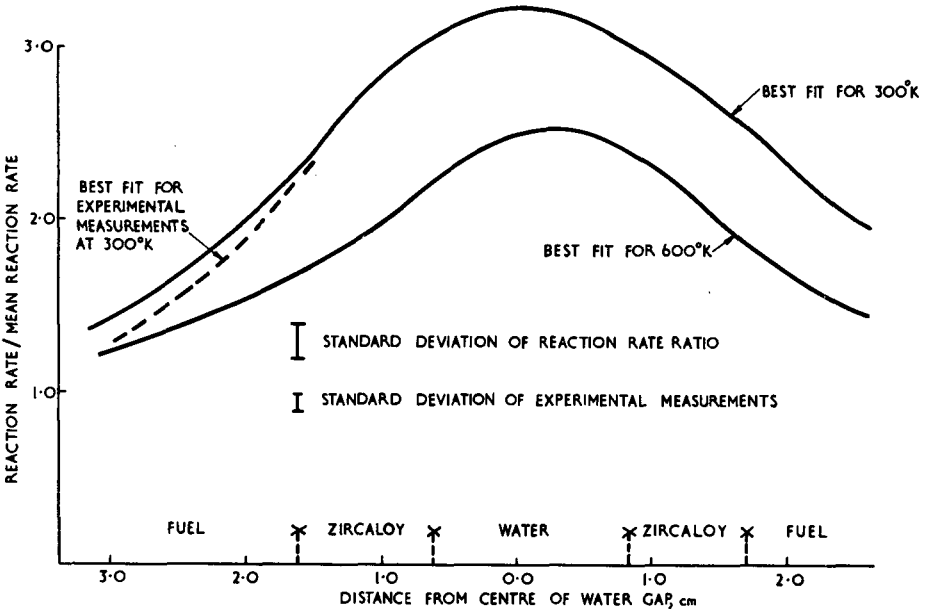

FIG. 6. Comparison of calculated and measured reaction rates.

To demonstrate the wide range of applicability of the nuclear data used, a MONK calculation was done with a spectrum typical of a light water moderated reactor. One of the problems of interest in such a reactor is the calculation of the power peaking in water gaps between the modules. Often the geometries are complex and because of the large flux gradients it is not feasible for methods based on diffusion theory to give predictions with very good accuracy. It is necessary, in order to get a reliable estimate of fluxes and reaction rates, to use a code based on Monte Carlo methods or transport theory. Figure 6 shows the results obtained with MONK in a typical water reactor at room temperature and the agreement with the experimentally measured U235 reaction rate in the fuel region is quite good. The standard deviation on the results however is about 10%. It is of interest to reactor designers to know the change in flux shape with temperature and Figure 6 shows the calculated reaction rates with the water density changed from 1.0 to 0.8 gm cm^{-3}. It was found that there was little change in the calculated U235 reaction rate at 300°K with this change in water density so a single curve is shown in Figure 6. This phenomenon has also been observed in some plastic moderated critical assemblies which are used to simulate coolant density variation. [19]

Figure 6 shows additionally the effect of changing the thermal constants to simulate a change in the thermal neutron spectrum. In order to test the reliability of the data at elevated temperatures it may be necessary to compare the results of calculations with experimental measurements from a high temperature test facility such as the Pressurised Test Reactor [20]. Meanwhile it is intended to compare the results of MONK with other computer codes at elevated temperatures.

CONCLUSIONS

The calculations on critical systems have shown that discrepancies as large as 5% can occur. It is likely that some of this variation is due to inaccuracies in the UK Nuclear Data Library and the representation of this data in the codes. The reaction rates are reasonably well calculated for fast reactor systems. The agreement between calculation and experiment lies within the statistical uncertainty for light water systems. However since most of the reaction rates occur in the thermal region the extrapolation to higher temperatures needs to be compared with the prediction of other methods. It is intended therefore that the thermal group representation and also the unresolved resonance region be improved. It is recommended therefore that in a safety assessment based on criticality calculations using GEM and MONK that sufficiently large safety factors be incorporated in the predictions of critical mass to compensate for uncertainties in the nuclear data.

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DISCUSSION

R.A. BONALUMI: I think reference might be made here to some adjustment work done by Chawla (India), I believe when he was at Winfrith. The adjustments were based on the WIMS calculational system and the UK Nuclear Data File. If I remember correctly, point-value cross-sections were varied (even for ^{235}U) and the fission spectrum was softened. The results were quite satisfactory in several respects, covering k_{eff}'s and reaction rate ratios equally well. In other words, no problem-dependence was visible, at least for H_2O and D_2O reactor lattices. Could Mr. Moore comment on this, i.e. on the existence of a very satisfactory adjusted data set based on the WIMS model? Could he indicate the range of Chawla's problems?

J.G. MOORE: Chawla considered only H_2O and D_2O moderated assemblies and, using the adjusted data set, obtained k_{eff} within about 1% of unity.

EFFECTS OF DIFFERENT NUCLEAR COMPUTER CODES AND DATA LIBRARIES ON THE EVALUATION OF THE CRITICAL HTR-EXPERIMENT CESAR II

W. SCHERER
Institut für Reaktorenentwicklung,
Kernforschungsanlage Jülich, GmbH,
Federal Republic of Germany

Abstract

EFFECTS OF DIFFERENT NUCLEAR COMPUTER CODES AND DATA LIBRARIES ON THE EVALUATION OF THE CRITICAL HTR-EXPERIMENT CESAR II.

In 1972, an extended experimental program on neutron physics of low-enriched HTR fuel balls has been performed in the graphite-moderated critical facility CESAR at CEN Cadarache/France. The experiments were carried out in the framework of a collaboration contract between KFA Jülich and CEA Cadarache. A large part of the measurements was designed to neutron flux mapping throughout the whole reactor. The core had in its centre a cavity filled with fuel balls, and several surrounding zones composed of hexagonal graphite blocks with various uranium fuel rods in order to achieve criticality and to adjust the HTR neutron spectrum in the centre of the reactor. The experimental results and some well-known computer codes have been compared by the author, showing that there are discrepancies not only between experiment and theory but also between the different codes and libraries. As an example (the whole comparison will be published later) the results of the GAMTEREX, MUPØ and ANISN spectrum calculations in connection with 2-D EXTERMINATØR reactor calculations, using the GAM-I, the THERMALIZATION, and the DRAGON-5 libraries are reported. As it was possible to combine different codes with the same library, the effects introduced by the different mathematical methods in solving the Boltzmann equation can be discussed separately. The results show that most of the discrepancies concerning the behaviour of neutron flux and reaction rates become severe near the boundaries of fuel zones with very different neutron spectra. The eigenvalue of the system differs from code to code by about 2% max. The effects of uncertainties in neutron data seem to be more important than the uncertainties of nuclear computer codes; thus greater attention should be paid to library problems in the future.

GENERAL REMARKS

In 1972 a comprehensive experimental programme on neutron physics of low-enriched HTR fuel balls was performed in the critical facility CESAR at the C.E.A. Nuclear Research Center Cadarache/France, in the frame of a collaboration contract.

It was the purpose of the programme to achieve information on the applicability of nuclear computer programmes for reactor lay-out, and to find the influence of different data-libraries on calculated results or at least to learn how to evaluate them. The low-enriched fuel cycle was chosen because that one was less studied in Germany than the uranium/thorium fuel cycle. In addition, our French partners had performed similar experiments with prismatic fuel elements. Consequently a comparison between fuel ball and prismatic fuel element was enabled from the nuclear point of view.

THE C E S A R - I I CORE

A big part of the experimental programme refers to the measurement of global reaction rates of the whole reactor (see Fig. 1). Because the fuel elements for the experiment were limited in their availability, the reactor had to be made critical by means of "classic CESAR elements". These are lightest-enriched (0.96 %) uranium rods embedded into hexagonal graphite blocks. However, compared with HTR fuel balls these driver elements have a completely different neutron spectrum; in order to stabilize the HTR eigenspectrum it was necessary to use several buffer and reference zones in the HTR fuel ball zone which finally led to the very heterogeneous core configurations shown in Fig. 2. A more detailed information might be taken from [1] and [2].

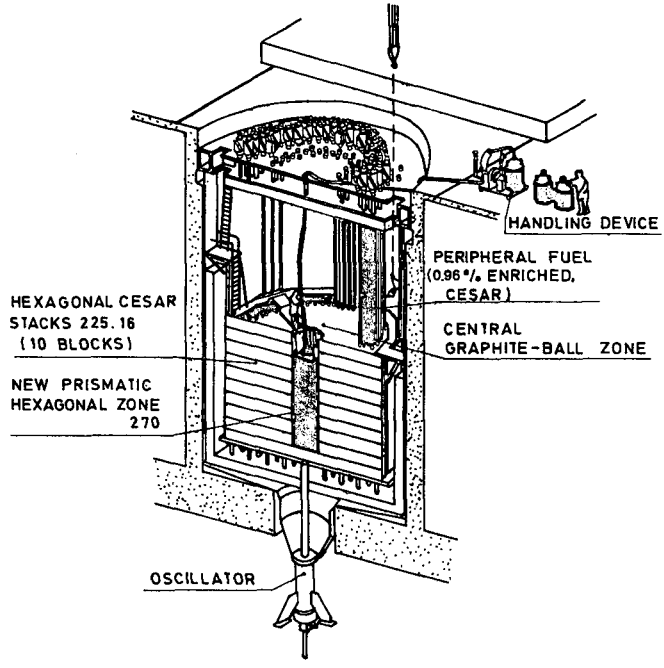

FIG.1. General view of CESAR reactor.

This core with its very many compositions and neutron spectra deviating from each other, on the one hand, does not represent an optimum of simplicity in reactor calculations; on the other hand, it permits an extensive testing of computer programmes and data libraries under strong conditions, thereby showing clearly the boundary conditions by which certain programmes could deliver reasonable results.

MEASUREMENTS

The measurements discussed here include the determination of reaction rates in both Mn and Au foils and the determination of cadmium ratios with those foils.

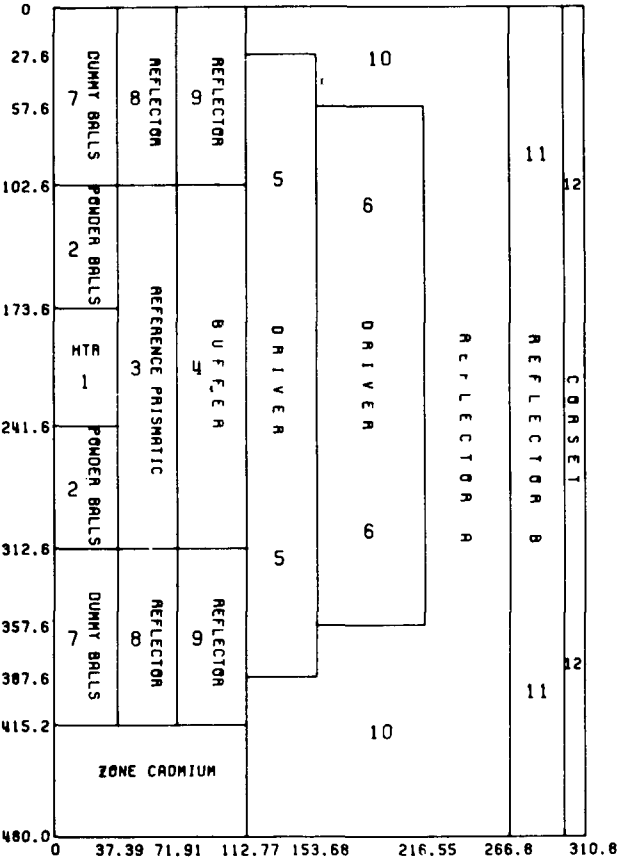

FIG.2. R-Z description of the CESAR reactor.

The measurements did not only refer to that part of the reactor filled with HTR fuel balls, but also to the outer zones of the critical facility which are present for spectral adaptation and achieving of criticality. Of particular interest is information from reflector boundaries and from transitions of reflector to fuel material, because here arise the most important problems in the calculation of the neutron spectrum.

CALCULATION MODEL AND PROGRAMMES

The mathematical evaluation of the measurements was performed in three steps. First, there were carried out multi-group spectral calculations for the different compositions of the core, and 4-group cross-sections were condensed. By recycling the buckling for several times, the spectral coupling of the different compositions were considered. At least the reaction rates for the mentioned detectors were determined by 4-group fluxes and by 4-group activation cross-sections condensed from spectrum calculations.

The following programmes are available for testing:

(a) APØLLØ
(b) GAMTEREX [3]
(c) GAMTERANEX [4]
(d) MUPØ-EX [5]

The APØLLØ programme represents a P_{ij} code developed by C.E.A. which was kindly made available to evaluate the CESAR-II experiment. The pertinent library is a 99-group library of UKAEA. GAMTEREX has been applied successfully in the KFA for many years to calculate the nuclear reactor lay-out. GAMTERANEX couples the GAM- and THERMØS-library with the well operating SN-code ANISN. MUPØ including the DRAGON-5 library is dependent on disadvantage factors from other calculations; a comparison, however, seems interesting because of the different library. The reactor calculation itself was performed in all cases with the two-dimensional diffusion code EXTERMINATOR-2.

RESULTS

The k_∞-values calculated by the different codes are given as first results in TABLE I. The differences are about 2 % except the buffer-zone where the discrepancy is around 7 %. It is interesting to recognize that the differences are not due to errors in the thermal spectra in general, because the $\eta \cdot f$-values as listed in TABLE II are in a much better agreement, especially for the buffer-zone.

TABLE I. VALUES OF k_∞

Zone	APØLLØ	MUPØ	GAMTEREX	GAMTERANEX
HTR balls	1.217	1.190	1.210	1.216
Powder balls	1.217	1.203	1.203	1.225
Prism. reference	1.328	1.333	1.342	1.353
Buffer	0.791	0.850	0.834	0.836
Driver (4)	1.152	1.177	1.151	1.147
Driver (5)	1.180	1.205	1.180	1.176
Driver (6)	1.152	1.179	1.147	1.148

TABLE II. VALUES OF $\eta \cdot f$

Zone	APØLLØ	MUPØ	GAMTEREX	GAMTERANEX
HTR balls	1.781	1.786	1.785	1.785
Powder balls	1.774	1.789	1.777	1.783
Prism. reference	1.752	1.763	1.758	1.759
Buffer	1.216	1.244	1.239	1.240
Driver (4)	1.266	1.303	1.267	1.267
Driver (5)	1.300	1.333	1.304	1.302
Driver (6)	1.266	1.306	1.268	1.268

TABLE III. VALUES OF k_{eff}

Programme	APØLLØ-EX	MUPØ-EX	GAMTEREX	GAMTERANEX
k_{eff}	0.9832	1.0003	0.9968	1.0064
Δk	- 1.7 %	0.0 %	- 0.3 %	0.6 %

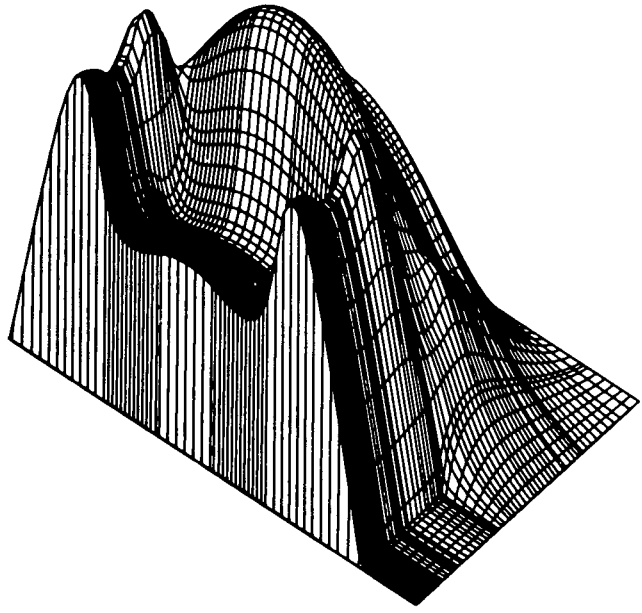

FIG.3. Thermal neutron flux; R-Z description.

Therefore it can be concluded that problems arise in calculating the resonance integral, and it should be taken care of both the methods and the resonance data.

The computer programmes have calculated deviating k_{eff}-values for the experimental critical facility. They are given in TABLE III.

Fig. 3 shows the thermal neutron flux in the reactor from which one can see the strong heterogeneity of the facility. The reaction rates measured by Au and Mn detectors show nearly the same shape.

Fig. 4 gives the Mn reaction rate in a radial section. The points connected by lines represent the calculation, the unconnected points the measurement. While the APØLLØ results are in good accordance with the experiment and also the MUPØ calculations could still be accpeted, GAMTEREX and GAMTERANEX show a relatively strong discrepancy in the region of the driver zones.

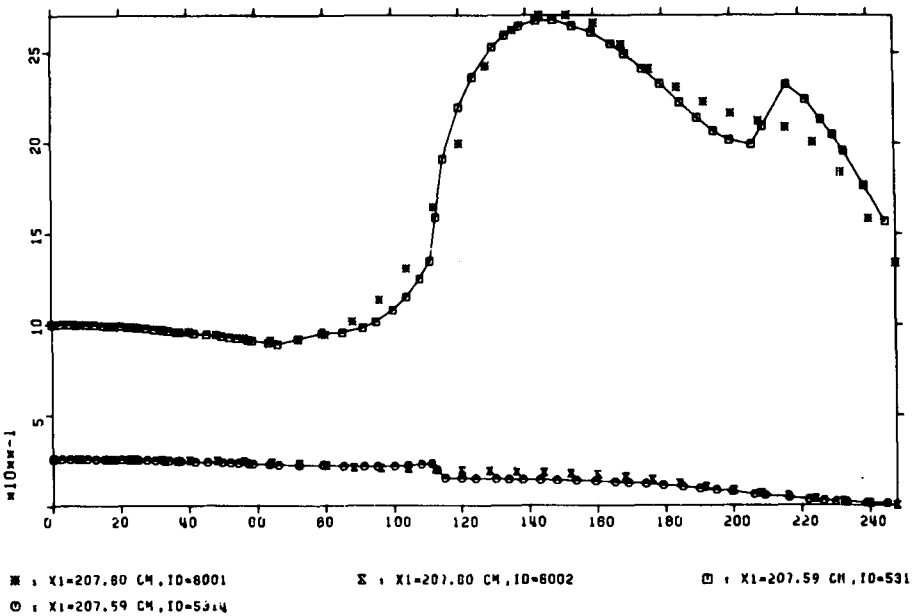

FIG. 4/1. Manganese radial without and with Cd; comparison between APØLLØ calculation and experiment.

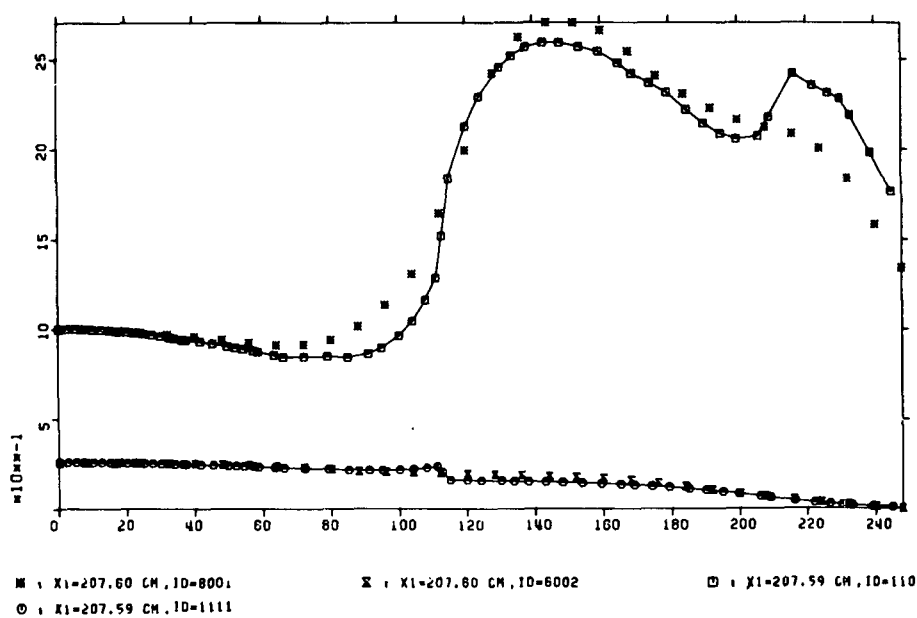

FIG. 4/2. Manganese radial without and with Cd; comparison between MUPØ-EX calculation and experiment.

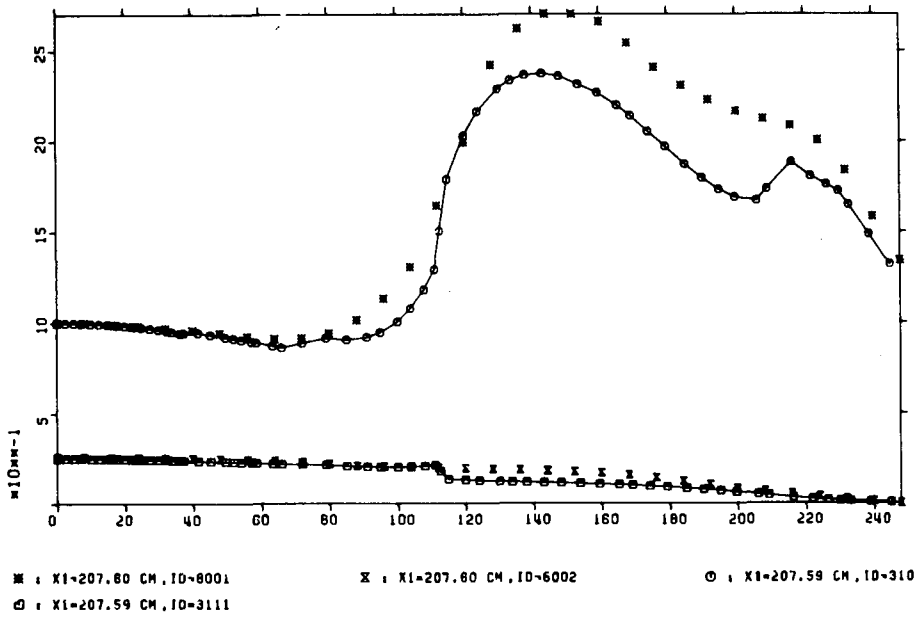

FIG. 4/3. Manganese radial without and with Cd; comparison between GAMTEREX calculation and experiment.

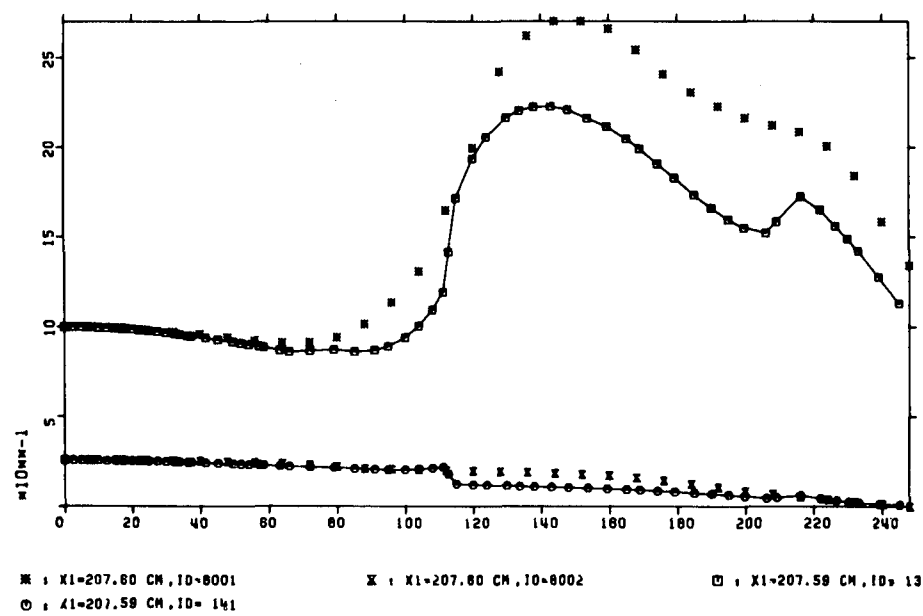

FIG. 4/4. Manganese radial without and with Cd; comparison between GAMTERANEX calculation and experiment.

An exact investigation resulted that these discrepancies stem from difficulties in the calculation of resonance absorption. For these two programmes the U-238 resonance integral was determined by means of ZUT-DGL. The transformation of the resonance reaction rate in a homogenized cross-section, however, is not perfectly performed in ZUT for extreme fuel elements like the CESAR driver element and buffer element. A correction is being carried out at the IRE.

Fig. 5 shows the axial section through the reactor core axis. It is shown the Mn reaction rate. (8008 = EXPERIMENT, 5313 = APØLLØ, 1101 = MUPØ, 1301 = GAMTEREX, 131 = GAMTERANEX.) Deviations from the prognosis in the reflector are easy to recognize. GAMTEREX and GAMTERANEX are much better in that case than the others, which could be explained by the quality of these programmes in absence of a resonance absorber.

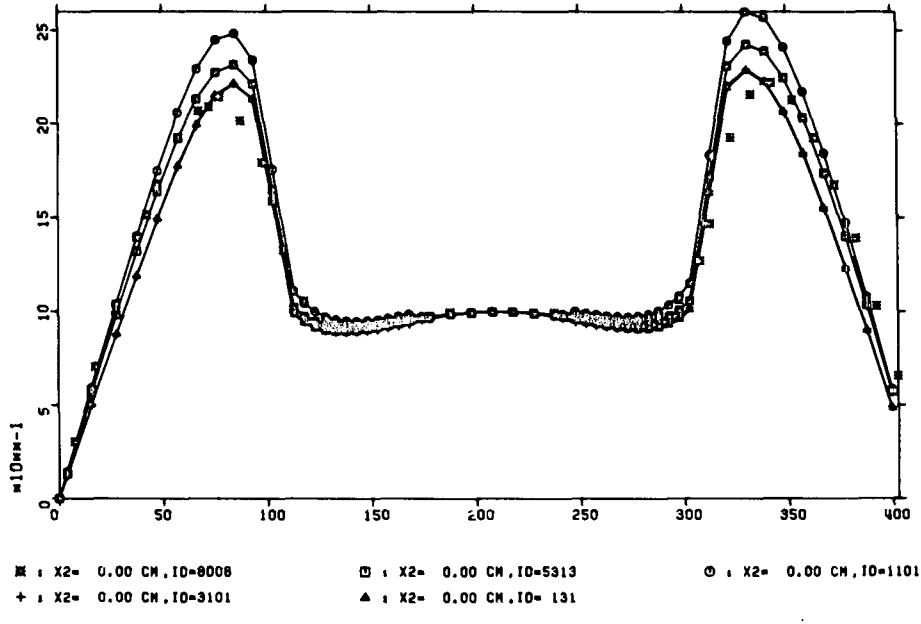

FIG. 5. Manganese axial; comparison between theory and experiment.

Fig. 6 shows results for gold detectors in radial direction. Also here, the highest discrepancy in the driver fields is shown for the GAMTEREX and GAMTERANEX programmes. However, from Fig. 7 we can see that this discrepancy stems from a failure in calculating the thermal flux. Fig. 7 shows the thermal neutron flux normed in the reactor center for all reactor programmes.

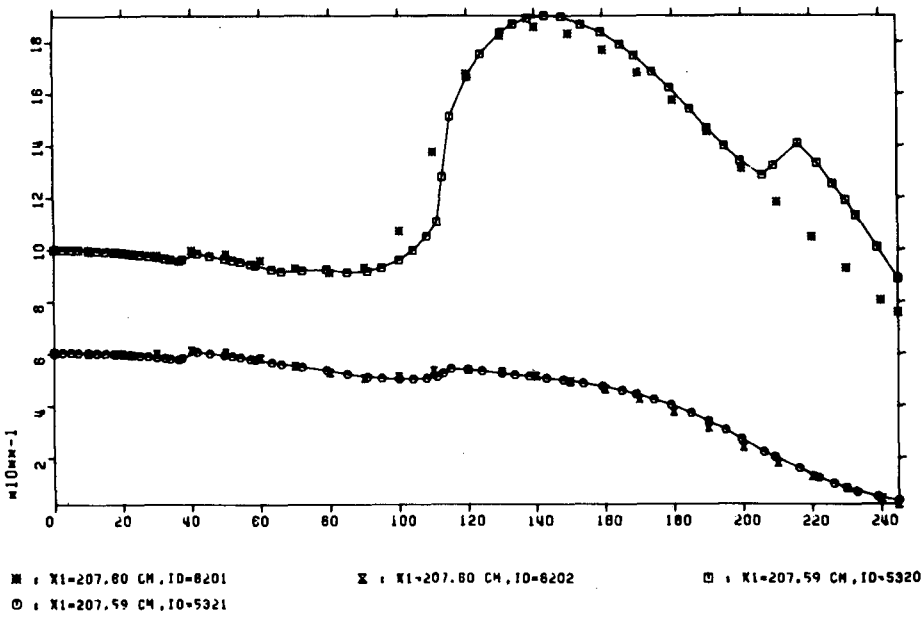

FIG.6/1. Gold radial without and with Cd; comparison between APØLLØ calculation and experiment.

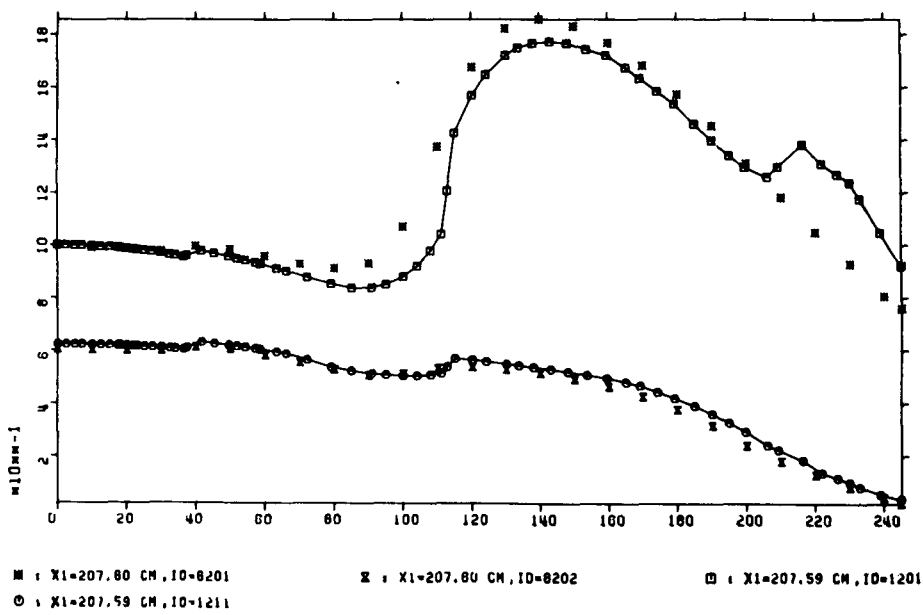

FIG.6/2. Gold radial without and with Cd; comparison between MUPØ-EX calculation and experiment.

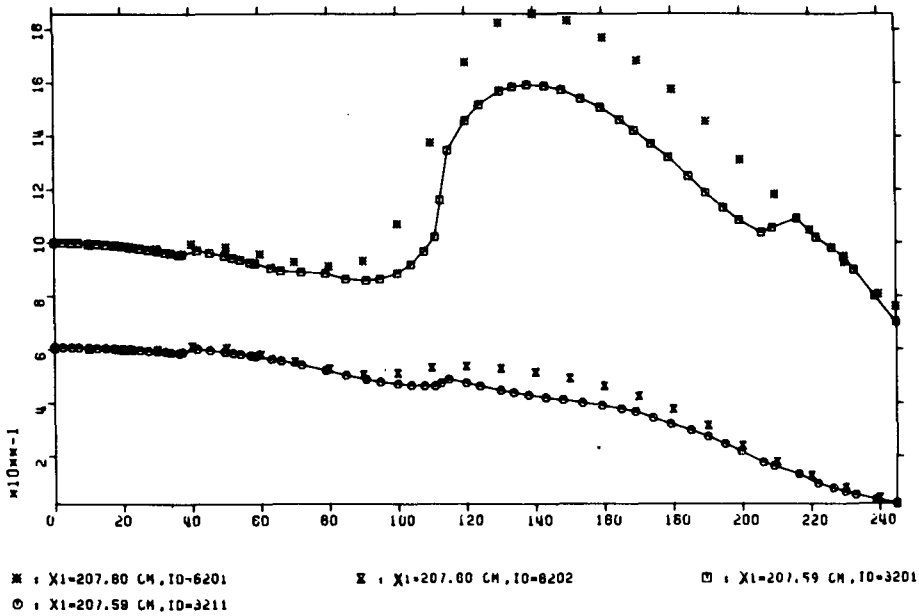

FIG. 6/3. Gold radial without and with Cd; comparison between GAMTEREX calculation and experiment.

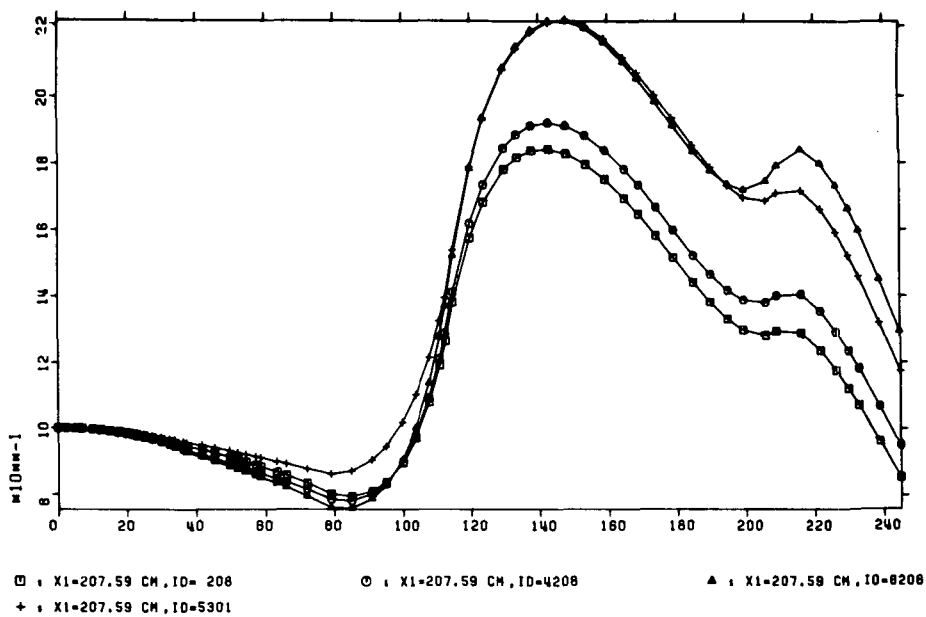

FIG. 7. CESAR II. Normalized thermal flux; radial. Theoretical values.

SUMMARY

The tested programmes essentially provide an exact record of the neutron-physical reactor behaviour. Deviations due to different calculation methods (SN, P_{ij}) are low compared with discrepancies due to the library which include above all the determination of group cross-section sets in the resonance region.

Deviations between experiment and theory do not significantly arise in typical HTR fuel elements. It is, however, necessary to improve the nuclear data libraries, as was demonstrated by the uncertainties of the calculated and measured values for the extremely heterogeneous CESAR-II fuel elements.

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Transl.:
A. Conrad-Wienands

WATER-MODERATED REACTOR ANALYSIS WITH ENDF/B DATA

S. JABBAWY, J. KARNI, W. ROTHENSTEIN, S. VELNER
Department of Nuclear Science,
Technion – Israel Institute of Technology,
Haifa, Israel

Abstract

WATER-MODERATED REACTOR ANALYSIS WITH ENDF/B DATA.

The clean thermal-reactor lattices with heavy and light water as moderator, originally studied by Honeck, Hellens et al., have been re-analysed with recent versions of ENDF/B data. A study based on the first ENDF/B compilation using the HAMMER code system at Brookhaven National Laboratory led to eigenvalues which were by one to two per cents too low. The later data sets, which necessitated modification of the multigroup library preparation code to handle the new formats and improved information, have not changed the results significantly. Attempts have been made to determine which part or parts of the analysis might account for the discrepancies which have been found.

A number of parameters have been compared with Monte-Carlo calculations which made use of the same basic data. Of these comparisons, the ones relating to the resonance and thermal reaction rates have been described previously. The values of the effective resonance integrals, as obtained by the Nordheim method in the HAMMER code, were decreased in accordance with the results then obtained. The THERMOS integral transport treatment of the unit lattice cell at thermal energies and subsequent Fourier-Transform leakage-spectrum calculations have been left unchanged. The Monte-Carlo estimates, treating the scattering anisotropy more fully, had resulted in very similar thermal reaction rates and slightly reduced leakage; for D_2O-lattices, the resulting increase in k_{eff} was not more than 0.2 to 0.4 per cents, and for H_2O lattices, the eigenvalues were changed even less.

Scattering anisotropy as specified in the ENDF/B data, together with the basic recommended cross-sections, have now also been included in a Monte-Carlo code for the epithermal energy region. The code has been used to verify the accuracy of the HAMMER code in which cell homogenization is effected by means of standard collision-probability methods, and the leakage spectrum is obtained by the MUFT B-1 procedure. At high energies, where the anisotropy of scattering becomes more important, the Monte-Carlo estimates led to a somewhat softer spectrum for the H_2O-moderated lattices. The leakage rates as obtained by the two methods are in reasonable agreement.

It may be concluded that the accuracy of rapid evaluations of the effective lattice resonance integrals from the basic ENDF/B data is the main cause of error in the lattice analysis.

Introduction

The analysis of neutron multiplication and space energy distribution of reaction rates in thermal reactor lattices provides a good means of testing the adequacy of the nuclear data used for reactor calculations. On the other hand the influence of the numerous approximations and simplifications introduced into computer codes to reduce machine times are by themselves a source of error which cannot be easily separated from the errors in the data themselves.

The present paper extends the clean thermal reactor analyses made previously [3] to ENDF/B-II data[1]. The calculations are based on the HAMMER code system [5]. In order to emphasize the assumptions and procedures used in the code for rapid computation the theoretical details have been brought together from the various relevant reports and papers and are presented in a consistent mathematical notation. The material is presented in the order in which the calculations are made in the code itself and should be borne in mind in the discussion of any effect which may give rise to discrepancies between experimental and calculated results.

Details of the ENDF/B-II multigroup libraries used are given and compared with the libraries used previously. Brief remarks are made about the data of the third version of ENDF/B. The systems analysed include clean heavy and light water moderated lattices. In the latter the calculations were extended to very tight lattices in which comparitively many fissions occur at epithermal energies. Some UO_2 fueled systems were also analysed.

In several parts of the energy spectrum comparisons have been made with Monte Carlo calculations based on special codes which use the same nuclear data as fully as possible. In particular resonance absorption remains the area in which the HAMMER system is most liable to error especially at very small moderator to fuel volume ratios in the light water slightly enriched Uranium lattices.

The Unit Cell Analysis Performed by the HAMMER Code System

The HAMMER code system [5] calculates the space energy distribution of the flux, current and reaction rates in the unit cell of a reactor lattice in one dimensional - and in particular cylindrical - geometry. The results are approximate solutions of the Boltzmann Equation. In the following the assumptions and simplifications made in the analysis will be described as these may be in part responsible for any discrepancies between calculation and experiment.

In the steady state transport equation

$$(\bar{\Omega}\cdot\nabla + \Sigma)f = q \qquad (1)$$

[1] For previous studies in this field, see Refs [1-4].

the total cross section Σ, the angular flux density f and the neutron emission density q are all functions of position $\bar{x}$, energy E and direction $\bar{\Omega}$.

The corresponding integral form of the transport equation is

$$f = \int_0^\infty ds\, e^{-\tilde{\Sigma s}}\, q(\bar{x} - s\bar{\Omega}, E, \bar{\Omega}) \tag{2}$$

where

$$\tilde{\Sigma s} = \int_0^s \Sigma(\bar{x} - \ell\bar{\Omega}, E)\, d\ell \tag{3}$$

is the optical distance between the point of emission of the neutrons and the point where the flux is evaluated.

Alternatively the integral transport equation for the flux density at $\bar{x}$, E and $\bar{\Omega}$ may be written in the form

$$f = \int \frac{d\bar{x}'}{s_1^2}\, e^{-\tilde{\Sigma s}_1}\, \delta(\bar{\Omega}' - \bar{\Omega})\, q(\bar{x}', E, \bar{\Omega}') \tag{4}$$

where s_1 and $\bar{\Omega}'$ are the magnitude and direction of the vector $\bar{x} - \bar{x}'$. The emission density at a specified location is

$$q(E,\bar{\Omega}) = \int dE' \int d\bar{\Omega}' [\Sigma_s(E \leftarrow E', \Omega \leftarrow \Omega') + \frac{\chi(E)}{4\pi} \nu(E')\Sigma_f(E')] f(E'\bar{\Omega}') + \frac{S(E)}{4\pi} \tag{5}$$

in which all the quantities refer to that location. If the double differential scattering cross section

$$\Sigma_s(E \leftarrow E', \bar{\Omega} \leftarrow \bar{\Omega}') = \frac{1}{2\pi} \sum_{\ell=0}^{\infty} \frac{2\ell+1}{2} \Sigma_{s\ell}(E \leftarrow E')\, P_\ell(\mu_o),\ \mu_o = (\bar{\Omega}' \cdot \bar{\Omega}) \tag{6}$$

is replaced by its isotropic term $\Sigma_{so}(E \leftarrow E')/(4\pi)$, the emission density becomes fully isotropic and an integral equation for the scalar flux $\phi = \phi(\bar{x},E)$ results from integration of Eq. (4) over all directions

$$\phi = \int \frac{d\bar{x}'}{4\pi s_1^2}\, e^{-\tilde{\Sigma s}_1}\, Q(\bar{x}'), \quad s_1 = |\bar{x} - \bar{x}'| \tag{7}$$

Here the total emission density at location $\bar{x}'$ refers to energy E and is given by

$$Q = \int d\bar{\Omega}\, q(E,\bar{\Omega}) = \int dE' [\Sigma_{so}(E \leftarrow E') + \chi(E)\, \nu(E')\, \Sigma_f(E')]\, \phi(E') + S(E) \qquad (8)$$

where all quantities refer to the point $\bar{x}'$.

In the cylindrical geometry of the HAMMER code the unit cell of the lattice is replaced by a cylindrised cell in which the volumes of all regions are preserved. The single coordinate r replaces the location $\bar{x}$, and a mesh of space points r_n each associated with volume ΔV_n is specified so that a number of values of n belong to each of the cell regions.

The calculations are divided into two parts, the first referring to thermal energies in which upscattering is taken into account and the second to epithermal energies. The order in which these two sections are treated is consistent with the assumption that after moderation the slowing down source into the thermal energy region is spatially flat in each region. The distribution of thermal fissions throughout the fuel region is then used as the fine structure of the fission source in the epithermal part of the code.

Both principal parts of the treatment use integral transport theory based on Eq. (7) with effectively isotropic sources to determine the space-energy distribution of the flux in the unit cell of an infinite lattice. After suitable homogenisation the transport equation (1) is used to estimate the effect of leakage on the energy spectrum. Finally the original space energy distribution of the flux in an infinite lattice is corrected for leakage, so that the resulting flux can be used for the calculation of any reaction rates desired.

The Procedure at Thermal Energies

Homogenisation

The spectrum calculations at thermal energies are based on the THERMOS code [6] in which 30 thermal energy groups are used. Here the energy variable is replaced by the neutron speed v expressed in units of 2200 m/sec :

$$v^2 = E/(kT_s) \qquad (9)$$

The discrete values v_i represent the central speeds of groups i and range from 0.1 to 5.285 with gradual increase of the group widths Δv_i. The cut-off of the thermal region is at the lower speed of group 30 (v = 5.0) which is not included in the averaging of thermal spectrum parameters.

The integral equation (7) for the scalar flux for isotropic neutron emission (8) is adapted to this group structure and applied to the discrete space points r_n. The flux $\phi(E)$, emission density $Q(E)$ and slowing down source $S(E)$ per unit energy at $E=E_i$ corresponding to the characteristic speed v_i of group i are replaced by the corresponding quantities $v_i N_{ni}$, Q_{ni} and S_{ni} per unit velocity in group i at space point n :

$$\phi(r_n,E_i) = N_{ni}/(2kT_s),\ Q(r_n,E_i) = Q_{ni}/(2v_i kT_s),\ S(r_n,E_i) = S_{ni}/(2v_i kT_s) \quad (11)$$

Similarly the scattering kernel $\Sigma_{so}(E \leftarrow E')dE'$ per unit final energy at $E=E_i$, and at $E'=E_j$, is replaced by the group kernel P^o_{nij} per unit final velocity at space point n :

$$\Sigma_{so}(r_n,\ E_i \leftarrow E_j)\ \Delta E_j = P^o_{nij}/v_i \quad (12)$$

so that

$$P^o_{nij} = (2kT_s)\ v_i\ v_j\ \Delta v_j\ \Sigma_{so}(r_n,\ E_i \leftarrow E_j) \quad (13)$$

The emission density equation becomes :

$$Q_{ni} = \sum_j P^o_{nij}\ N_{nj} + S_{ni} \quad (8')$$

and the integral transport equation

$$N_{ni} = \sum_{n'} T_{nn'i}\ \frac{Q_{n'i}}{v_i} \quad (7')$$

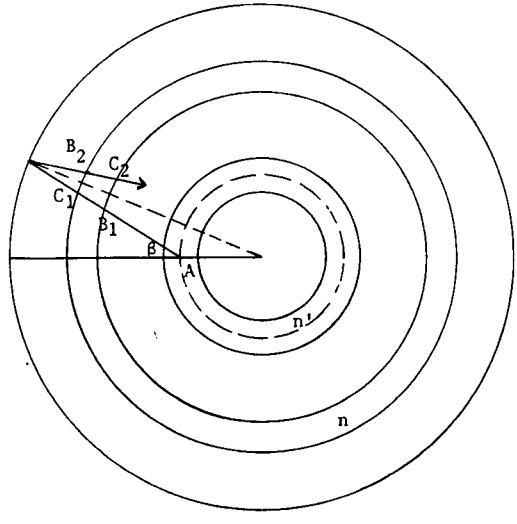

FIG.1. $\ell_p = AB_p$, $\ell'_p = AC_p$. p is the order number of the intersection of the neutron path with annulus n, if any.

The transport kernel $T_{nn'i}$ is obtained by integrating the transport equation (7) over the volume ΔV_n surrounding space point n. For constant fluxes in each subregion n the kernel becomes

$$T_{nn'i} = \frac{1}{\Delta V_n} \int_{\Delta V_n} d\bar{x} \int_{\Delta V_{n'}} d\bar{x}' \frac{e^{-\Sigma_i \tilde{s}_1}}{4\pi s_1^2}, \quad s_1 = |\bar{x}-\bar{x}'| \tag{14}$$

(see Fig. 1).

In the THERMOS procedure the integral over the initial annulus n' is approximated by the integrand appropriate to an initial location on the central cylinder of the annulus multiplied by $\Delta V_{n'}$. The kernel then becomes :

$$T_{nn'i} = \frac{\Delta V_{n'}}{\Sigma_{ni}\,\Delta V_n} \sum_P \int_0^{2\pi} \frac{d\beta}{4\pi} \cdot 2 \int_0^{\pi/2} \sin\theta \left[e^{-\tilde{\Sigma}\ell_P/\sin\theta} - e^{-\tilde{\Sigma}\ell'_P/\sin\theta}\right]$$

$$= \frac{\Delta V_{n'}}{\Sigma_{ni}\,\Delta V_n} \int_0^{2\pi} \frac{d\beta}{2\pi} \sum_P [K_{i2}(\tilde{\Sigma}_i \ell_P) - K_{i2}(\tilde{\Sigma}_i \ell'_P)] \tag{15}$$

where Σ_{ni} is the total cross section in annulus n in group i, and $\tilde{\Sigma}_i \ell_P$ and $\tilde{\Sigma}_i \ell'_P$ are optical distances in group i. The reflecting boundary condition at the outer cylinder of the unit cell can be altered effectively by an additional region of heavy scatterer which is taken into account in the spectrum calculations of the infinite lattice, but not in the averaging of the thermal parameters.

The scattering kernels of Eq. (13) are obtained from the Legendre moments of order zero of the scattering cross section, Eq. (6), for energies $E=E_i$ and $E'=E_j$. For the principal scatterer of the moderator of mass ratio A and free atom cross section σ_f the FLANGE code [7] calculates the moments of order ℓ, $\sigma_{s\ell}(E_i \leftarrow E_j)$ from the scattering function $S(\alpha,\beta,T)$ tabulated in the ENDF/B format [8] :

$$\sigma_s(E_i \leftarrow E_j, \bar{\Omega} \leftarrow \bar{\Omega}') = \sigma_f \left(\frac{A+1}{A}\right)^2 e^{-\beta/2} \sqrt{E_i/E_j}\, S(\alpha,\beta,T)/(4\pi kT) \tag{16}$$

where

$$\alpha = (E_i + E_j - 2\sqrt{E_i E_j})/(AkT), \quad \beta = (E_j - E_i)/(kT)$$

$$\sigma_{s\ell}(E_i \leftarrow E_j) = 2\pi \int \sigma_s(E_i \leftarrow E_j, \bar{\Omega} \leftarrow \bar{\Omega}')\, P_\ell(\mu_o) d\mu_o, \quad \mu_o = (\bar{\Omega}' \cdot \bar{\Omega}) \tag{17}$$

The diagonal elements $\sigma_{s\ell}(E_j \leftarrow E_j)$ are adjusted so that $\int \sigma_{s\ell}(E_i \leftarrow E_j) dE_i$ gives the ℓ'th moment of the scattering cross section $\sigma_{s\ell}(E_j)$. The integral transport treatment of the THERMOS code makes use of $\sigma_{so}(E_i \leftarrow E_j)$ only, so that the scalar flux can be calculated directly from the integral equation (7). If the kernel $\sigma_{s1}(E_i \leftarrow E_j)$ is also available the code replaces $\sigma_{so}(E_j)$ by $\sigma_{s,tr}(E_j) = \sigma_{so}(E_j) - \sigma_{s1}(E_j)$ and modifies $\sigma_t(E_j)$ accordingly.

For heavier isotopes the double differential cross section is taken from the gas model [9], which is also used to define the source into the thermal groups i. For one of the constituents K of the mixture in subregion n the microscopic slowing down source is

$$S_K(E_i) = \int_{E_o}^{\infty} \sigma_{so}^K (E_i \leftarrow E) \frac{dE}{E} \tag{18}$$

assuming a 1/E flux above the cutoff E_o of the highest thermal group.

$$\sigma_{so}^K (E_i \leftarrow E) = \frac{\sigma_f^K \theta^2}{2E} [e^{-(\epsilon_i - \epsilon)} \{\mathrm{erf}(\theta\sqrt{\epsilon} - \zeta\sqrt{\epsilon_i}) - \mathrm{erf}(\theta\sqrt{\epsilon} + \zeta\sqrt{\epsilon_i})\} + \mathrm{erf}(\theta\sqrt{\epsilon_i} + \zeta\sqrt{\epsilon}) + \mathrm{erf}(\theta\sqrt{\epsilon_i} - \zeta\sqrt{\epsilon})] \tag{19}$$

where

$$\theta = (A_K + 1)/(2\sqrt{A_K}) \;,\; \zeta = (A_K - 1)/(2\sqrt{A_K}) \;,\; \epsilon = E/(kT) \;,\; \epsilon_i = E_i/(kT)$$

The macroscopic scattering kernels P_{nij}^o and sources S_{ni}, derived from the thermal library data $\sigma_{s\ell}(E_i \leftarrow E_j)$ for the different isotopes and the sources $S_K(E_i)$, together with the transport kernel $T_{nn'i}$, Eq. (15), are the data for the iterative solution of Eqs. (7') and (8') for the spatial distribution of the neutron density N_{ni} per unit velocity in group i, or the flux per unit energy. As initial guess the Maxwell distribution $v_i^2 \exp(-v_i^2)$ is used.

The neutron density is used to homogenise the cross sections for the unit cell of the infinite lattice

$$\Sigma_i^x = \sum_n \Sigma_{ni}^x N_{ni} \Delta V_n / [\sum_n N_{ni} \Delta V_n] \tag{20}$$

$$P_{ij}^\ell = \sum_n P_{nij}^\ell N_{nj} \Delta V_n / [\sum_n N_{nj} \Delta V_n] \tag{21}$$

Leakage

The leakage calculations are based on the differential form of the transport equation (1). For leakage in the z direction only the equation is subjected to a Fourier Transform after which the critical eigenvalue B is assigned to the transform variable. This buckling makes the solution stationary. Denoting the transformed flux and emission density by $\tilde{f}$ and $\tilde{q}$

$$f(z,E,\mu) = 2\pi f(z,E,\bar{\Omega}) = \tilde{f}(E,\mu)e^{iBz} \quad , \quad q = \tilde{q}\, e^{iBz}, B = \sqrt{|B^2|} \tag{22}$$

and

$$(iB\mu + \Sigma)\,\tilde{f} = \tilde{q} \quad \text{at energy E} \tag{1'}$$

Expanding

$$\tilde{f}(E,\mu) = \sum_{\ell=0}^{\infty} \frac{2\ell+1}{2}\, i^{\ell}\, F_{\ell}(E)\, P_{\ell}(\mu) \tag{23}$$

and using a similar expansion for $\tilde{q}$ the P-1 equations are obtained in the usual manner at energy E

$$F_o = (\Sigma q_o + B q_1)/(\Sigma^2 + \tfrac{1}{3}B^2) \quad , \quad F_1 = (\Sigma q_1 - \tfrac{1}{3}B q_o)/(\Sigma^2 + \tfrac{1}{3}B^2) \tag{24}$$

If Eq. (1') is first divided by $(\Sigma + iB\mu)$ and the expansions are then made the B-1 equations result for energy E

$$F_o = b_{oo}\, q_o + b_{o1}\, q_1 \quad ; \quad F_1 = b_{1o}\, q_o + b_{11}\, q_1 \tag{25}$$

where $$b_{oo} = \frac{b}{\Sigma_t} \quad , \quad b_{o1} = \frac{3(1-b)}{B} \quad , \quad b_{1o} = -\frac{(1-b)}{B} \quad , \quad b_{11} = \frac{3(1-b)}{aB} \tag{26}$$

and $$a = \frac{B}{\Sigma_t} \quad , \quad b = \begin{cases} \dfrac{\tan^{-1} a}{a} & , \quad \text{if } B^2 > o \\ \dfrac{1}{2a}\, \ell n\, \dfrac{1+a}{1-a} & , \quad \text{if } B^2 < o \end{cases} \tag{27}$$

Since $q_o(E)$ and $q_1(E)$ depend on F_o and F_1 at other energies equations (24) or (25) are solved iteratively in their multigroup form.

One group thermal parameters at energies E_i corresponding to the speeds v_i are calculated from

$$\tilde{\Sigma}^x = \sum_i \Sigma_i^x \; F_o(E_i) \; \Delta E_i / \sum_i F_o(E_i) \; \Delta E_i \tag{28}$$

and the thermal diffusion constant is defined as

$$\tilde{D} = -\sum_i F_1(E_i) \; \Delta E_i / [B \sum_i F_o(E_i) \; \Delta E_i] \tag{29}$$

The previous neutron number density per unit speed (or flux per unit energy), N_{ni} in the infinite lattice is used as a spatial weighting function to determine the leakage corrected flux throughout the lattice :

$$N'_{ni} = N_{ni} \; F_o(E_i) / \sum_n N_{ni} \; \Delta V_n \tag{30}$$

These fluxes may ultimately be used to average any locally defined cross sections over a range of energy groups and geometrical regions in a manner which is equivalent to the two stage averaging process (infinite lattice spectrum and homogenised cell leakage spectrum) described above.

The Epithermal Procedure

Lattice Flux

Infinite lattice calculations based on the integral transport equation are carried out for the unit cell in a manner which is similar to the method used in the thermal groups. There are however some special features.

The transfer matrix in each of the epithermal groups is calculated in the HAMMER code by means of the method of random chords and cosine currents as distinct from the procedure described before. An advantage of this method is that the matrix is properly averaged over the region in which the neutron starts its flight, instead of choosing the average radius as the initial location. On the other hand calculation time for evaluating the transfer matrix elements is saved by the assumption that currents incident on the cylindrical surfaces of an annular region are proportional to the cosine of the angle of incidence.

Two probabilities P_n^o and P_n^i are calculated for a unit source of neutrons distributed uniformly and isotropically throughout the n^{th} annular region.

These are the probabilities of escape through the outer and inner surfaces respectively. They are given by

$$\int_0^{\pi/2} 2\sin\theta\, d\theta \int_{a_n^i}^{a_n^o} 2\pi r\, dr \int d\beta\, e^{-\Sigma_n x/\sin\theta} /(4\pi\Delta V_n) \qquad \text{(see Fig. 2).}$$

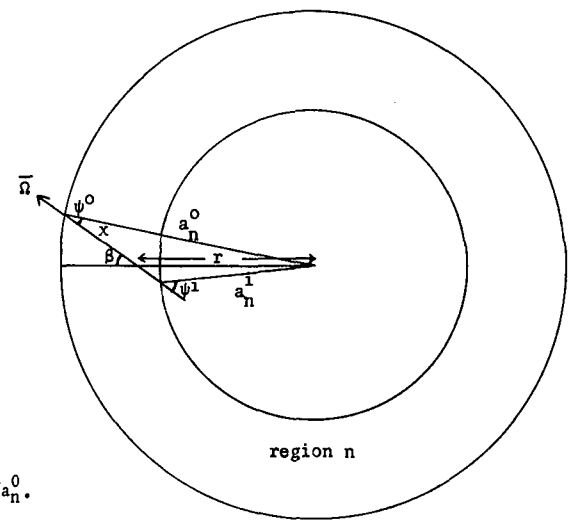

FIG.2. $K = a_n^i/a_n^0$.

The limits of β are determined by the criterion whether $\bar{\Omega}$, the neutron direction, will intersect the outer surface, for P_n^o, or the inner surface, for P_n^i. After introducing $rdr\,d\beta = a_n^o \cos\psi^o\, d\psi^o dx$, and performing the integration over x, the escape probability through the outer surface becomes :

$$P_n^o = \frac{a_n^o}{\Sigma_n \Delta V_n} \int_{\sin^{-1}K}^{\pi/2} 2d\psi^o \cos\psi^o \int_0^{\pi/2} d\theta \sin^2\theta (1-e^{-2\Sigma_n a_n^o/\cos\psi^o/\sin\theta})$$

$$+ \frac{a_n^o}{\Sigma_n \Delta V_n} \int_0^{\sin^{-1}K} 2d\psi^o \cos\psi^o \int_0^{\pi/2} d\theta \sin^2\theta [1-e^{-(\Sigma_n a_n^o \cos\psi^o/\sin\theta + \Sigma_n a_n^i \cos\psi^i/\sin\theta)}]$$

$$= \frac{2}{\pi a_n^o \Sigma_n (1-K^2)} \int_K^1 dt [\frac{\pi}{4} - K_{i3}(\Sigma_n a_n^o \sqrt{1-t^2})]$$

$$+ \frac{2}{\pi a_n^o \Sigma_n (1-K^2)} \int_0^K dt [\frac{\pi}{4} - K_{i3}(\Sigma_n a_n^o \sqrt{1-t^2} - \Sigma_n a_n^i \sqrt{K^2-t^2})] \qquad (31)$$

where $K = a_n^i/a_n^o$ and $a_n^o \sin\psi^o = a_n^i \sin\psi^i$.

Similarly the second term is found to be P_n^i . The appearance of $(\cos\psi^o \sin\theta)\ \sin\theta\ d\theta\ d\psi^o$ as well as the chord length in mean free paths in the exponential leads to the possibility of expressing the above two probabilities in terms of transmission probabilities G_n^{io} (from inner to outer), G_n^{oi} (from outer to inner) and G_n^{oo} (from outer to outer) for cosine currents impinging on the surfaces of the annulus :

$$P_n^o = \frac{\pi a_n^o}{2\Sigma_n \Delta V_n}(1 - G_n^o) \quad , \quad G_n^o = G_n^{oo} + G_n^{oi} \tag{32}$$

$$P_n^i = \frac{\pi a_n^i}{2\Sigma_n \Delta V_n}(1 - G_n^{io}) \quad , \quad a_n^i\, G_n^{io} = a_n^o\, G_n^{oi} \tag{33}$$

The transmission probabilities G_n^{oo} and G_n^{oi} are in fact the integrals over the K_{i3} functions appearing in Eq. (31) multiplied by $(4/\pi)$.

Assuming in the actual situation that the currents at the annular surfaces are cosine shaped, balance equations may be set up for (N-1) annular regions (n=2,3, ... N) surrounding the central cylinder n=1.

$$J_{n-1}^- = S_n P_n^i + J_n^- G_n^{oi} \tag{34}$$

$$J_n^+ = S_n P_n^o + J_{n-1}^+ G_n^{io} + J_n^- G_n^{oo} \tag{35}$$

for n=2,3, ... N , together with

$$J_1^+ = S_1 P_1^o + J_1^- G_1^{oo} \tag{36}$$

A reflecting boundary condition

$$J_N^+ = J_N^- \tag{37}$$

completes the 2N equations from which the currents J_N^- and J_N^+ are determined, once P_n^o has been determined for each region n from Eq. (31). The other probabilities all follow from Eqs.(32) and (33).

The final calculation of the transfer matrix is based on the solution of the above equations for a source $S_n = \Delta V_{n'}\ \delta_{nn'}$, in the annulus

n' only, and the calculation of the number of collisions ($\Sigma_n \phi_n \Delta V_n$) in region n from

$$\Sigma_n \phi_n \Delta V_n = \Delta V_{n'} \delta_{nn'} + J_n^- + J_{n-1}^+ - J_n^+ - J_{n-1}^- \tag{38}$$

The matrix element equivalent to Eq. (15) is then

$$T_{nn'} = \phi_n \tag{39}$$

The group structure for the epithermal calculations is the 54 group set of the MUFT code [10] with lethargy intervals of 0.25 in the region of the principal resonances just above the thermal energies and again in the region of the fission spectrum, and intervals $\Delta u = 0.50$ for the intermediate groups. The first group is at the top of the fission spectrum (with upper energy at 10 MeV). For group i the integral transport equations (7') and (8') may now be written together in the form

$$\phi_{ni} = \sum_{n'} T_{nn'i} \left(P_{n'i} \phi_{n'i} + S_{n'i}\right) \tag{40}$$

where the flux and source here refer to unit lethargy of group i. The kernel $P_{n'i}$ is analogous to Eq. (13) and expresses the in-group scattering cross section from the entire group i into unit lethargy of the same group. The source $S_{n'i}$ includes fission and down scattering. The definition of both quantities may be based on the Groeling-Goertzel [11] approximation for the elastic slowing down density

$$q(u) = \frac{\xi}{\lambda} \int_{-\infty}^{u} e^{-(u-u')/\lambda} \Sigma_s(u')\phi(u')du', \quad \xi = \langle(u-u')\rangle, \quad \xi\lambda = \frac{1}{2}\langle(u-u')^2\rangle \tag{41}$$

which is correct to a two term Taylor expansion of the scattering rate in the integrand near u'=u for A>1, and becomes exact for Hydrogen for which $q=\eta$ and $\xi=\lambda=1$.

For a group of width $\Delta u << \lambda$ the contribution to the integral of Eq. (41) due to downscattering from outside the group is given by

$$\frac{\xi\Sigma_s \phi \Delta u}{\lambda + \frac{\Delta u}{2}}$$

to a good approximation. The in-group scattering cross section is then

$$\Sigma_s \simeq \frac{\xi\Sigma_s}{\lambda + \frac{\Delta u}{2}} \qquad (\Delta u << \lambda) \tag{42}$$

where all the quantities are group averages.
On the other hand for heavy materials with $\Delta u >> \lambda$ the in-group scattering cross section is

$$\Sigma_s \simeq \frac{\xi\Sigma_s}{\Delta u} \qquad (\Delta u >> \lambda) \tag{43}$$

The two expressions become identical if one sets λ equal to $\frac{\Delta u}{2}$ for heavy isotopes.

The kernel $P_{n'i}$ is thus given by

$$P_{n'i} = \left[\Sigma_{so} - \Sigma_{s1} - \frac{2\ (\xi\Sigma_s')}{2\bar{\lambda} + \Delta u} - \frac{2\ \Sigma_s^H}{2 + \Delta u} \right]_{n'i} + \left[\Sigma_{in,i\to i}\right]_{n'} \tag{44}$$

where the average cross sections in group i and the inelastic in-group cross section are summations over the constituents of the mixture in the subregion near point n'. The prime of $\xi\Sigma_s'$ denotes summation over all isotopes other than Hydrogen (H) and their average $\bar{\lambda}$ is obtained using the individual values of $\xi\Sigma_s$ of these isotopes as weights. Hydrogen is treated entirely separately as shown.

The infinite lattice flux distributions are calculated from Eq. (40) starting with group i=1. Resonance capture calculations are made only at a subsequent stage, but before the next group is treated. Slowing down densities at the low lethargy of group i (indicated by superscript o) must therefore be corrected for resonance absorption in group (i-1), before they can be used to define the source $S_{n'i}$ of Eq. (40). The treatment is based on the differential equation derived from Eq. (41)

$$q + \lambda \frac{dq}{du} = \xi\ \Sigma_s\ \phi \tag{45}$$

Integrating over group (i-1) one obtains

$$q_i^o = \frac{2\bar{\lambda} - \Delta u_{i-1}}{2\bar{\lambda} + \Delta u_{i-1}}\ q_{i-1}^o\ (1 - R_{i-1}) + \frac{2(\xi\Sigma_s')_{i-1}\ \phi_{i-1}\ \Delta u_{i-1}}{2\bar{\lambda} + \Delta u_{i-1}} \tag{46}$$

where $q^o_{i-1}\ (1 - R_{i-1})$ is the corrected slowing down density at the low lethargy of group (i-1), R_{i-1} being the resonance absorption fraction in this group. The flux ϕ_{i-1} and $(\xi\Sigma'_s)_{i-1}$ are average values in the group. Eq. (46) refers to all constituents of the mixture other than Hydrogen, which is treated separately by a corresponding expression with q replaced by η, $\xi=\bar{\lambda}=1$, and Σ^H_s in place of $\xi\Sigma'_s$.

From Eq. (41) the slowing down density in group i becomes

$$q(u) = q^o_i \; e^{-(u-u_o)/\lambda} \tag{47}$$

where u_o is the low lethargy of the group. A similar approximation as the one used previously leads to the slowing down source into the entire group

$$q^o_i \; \frac{\Delta u_i}{\lambda + \frac{\Delta u_i}{2}} \qquad \text{for} \quad (\lambda >> \Delta u_i).$$

For groups which are very broad compared with λ the corresponding source into group i is q^o_i. These expressions become again identical if λ is replaced by $\frac{\Delta u_i}{2}$ for heavy isotopes. Consequently the source into unit lethargy of group i is

$$S_{n'i} = \left[\frac{2\, q^o_i}{2\bar{\lambda} + \Delta u_i} + \frac{2\, \eta^o_i}{2 + \Delta u_i}\right]_{n'} + \sum_{i'=1}^{i-1} \Sigma_{in,n',i'\to i}\, \phi_{n'i'} \Delta u_{i'}/\Delta u_i + \chi_i\, X_{n'} \tag{48}$$

where $\bar{\lambda}$ is again the average value for all constituents of the mixture near location n', except Hydrogen. χ_i is the average fission spectrum in group i and $X_{n'}$ the density of thermal fissions per unit volume near n'. It is calculated from the thermal spectrum calculations for the infinite lattice and replaced by a flat source in the fissionable regions if the THERMOS calculations have been omitted. $X_{n'}$ is normalised to unity over the unit cell.

For the calculation of the transfer matrix $T_{nn'i}$ the total cross section of each subregion is replaced by the transport cross section consistent with the procedure used for Eq. (44) :

$$(\Sigma_t)_{ni} = (\Sigma_{so} - \Sigma_{s1} + \Sigma_a + \Sigma_{in})_{ni} \tag{49}$$

Equation (40) is solved iteratively for the flux ϕ_{ni} per unit lethargy normalised to one fission neutron injected into the unit cell.

Resonance Absorption

The resonance absorption calculations are carried out according to the Nordheim treatment [12] for isolated resonances of the Single Level Breit Wigner Shape. In the groups containing resolved resonances of one of the constituent nuclides of the mixture its cross sections near one of these resonances are given by :

$$\sigma_c(E) = \sigma_o \frac{\Gamma_\gamma}{\Gamma}\sqrt{\frac{E_o}{E}}\,\psi(x,\theta) \quad , \quad \sigma_f(E) = \sigma_o \frac{\Gamma_f}{\Gamma}\sqrt{\frac{E_o}{E}}\,\psi(x,\theta)$$

$$\sigma_s^r(E) = \sigma_o \frac{\Gamma_n}{\Gamma}\psi(x,\theta) \quad , \quad \sigma_i^r(E) = \sqrt{g\sigma_p\sigma_o\frac{\Gamma_n}{\Gamma}}\,\chi(x,\theta)$$

$$\psi(x,\theta) = \frac{1}{\sqrt{4\pi\theta}}\int_{-\infty}^{\infty}\frac{e^{-(x-y)^2/(4\theta)}}{1+y^2}\,dy \quad , \quad \chi(x,\theta) = \frac{1}{\sqrt{4\pi\theta}}\int_{-\infty}^{\infty}2y\,\frac{e^{-(x-y)^2/(4\theta)}}{1+y^2}\,dy$$

$$\sigma_o = 4\pi\left(\frac{\hbar^2}{2mE_o}\right) g\,\frac{\Gamma_n}{\Gamma} \quad , \quad \Gamma = \Gamma_n + \Gamma_\gamma + \Gamma_f \quad , \quad g = \frac{2J+1}{2(2I+1)}$$

$$x = \frac{2}{\Gamma}(E-E_o) \quad , \quad \theta = \frac{\Delta^2}{\Gamma^2} \quad , \Delta^2 = \frac{4E_o kT}{A}$$

σ_p = potential scattering cross section, m = neutron mass

I = nuclide spin , J = compound nucleus spin

(50)

The resonance nuclide of mass m A_o may be mixed with up to two other nuclides of masses m A_1 and m A_2 , $(A_1<A_2)$, and constant cross sections in group i. The atom densities N_o, N_1 and N_2 are region averages over all subregions containing the resonance nuclide and scattering cross sections are normalised to N_o so that

$$\sigma_1 = (\sigma_s)_1 \cdot N_1/N_o \quad \text{and} \quad \sigma_2 = (\sigma_s)_2\, N_2/N_o \tag{51}$$

The total cross section normalised to N_o is then, by Eq. (50),

$$\sigma_t(u) = \sigma_a(u) + \sigma_s(u) + \sigma_1 + \sigma_2, \; \sigma_a(u) = \sigma_c(u) + \sigma_f(u) \;,$$

$$\sigma_s(u) = \sigma_s^r(u) + \sigma_i^r(u) + \sigma_p \tag{52}$$

The region flux per unit lethargy in group i is defined by

$$\Phi_o = \sum_n (\phi_{ni}\, \Delta V_n)/V_o \quad , \quad V_o = \sum_n \Delta V_n \tag{53}$$

where the summation includes all subregious containing the resonance nuclide and the fluxes ϕ_{ni} are calculated from Eq. (40). The collision density per unit lethargy in V_o is denoted by $N_o E\, \Psi(u)\Phi_o$ near the resonance and becomes asymptotically

$$N_o(\sigma_p + \sigma_1 + \sigma_2)\Phi_o = N_o E\, \Psi^{as}(u)\, \Phi_o \tag{54}$$

at energies E well above the resonance energy E_o. The function $\Psi(u)$ is calculated by numerical integration for lethargies in the vicinity of the resonance peak by numerical integration of its integral equation making use of collision and escape probabilities.

The escape probabilities are in general derived by the methods given in Eqs. (31-39). For transfer from V_o to a subregion n' outside it

$$P_{n'o}(u) = \Sigma_{n'}\, \Delta V_{n'}\, T_{n'o}(u) \tag{55}$$

where $T_{n'o}(u)$ is the average flux in n' due to unit source in V_o, distributed uniformly and isotropically, and is obtained from Eq. (39). The reverse probability is

$$P_{on'}(u) = N_o\, \sigma_t(u)\, V_o\, T_{on'}(u) \tag{56}$$

The quantity $T_{on'}(u)$ is in fact the kernel of Eq. (14) multiplied by $(1/\Delta V_{n'})$, the source density in subregion n', and satisfies the reciprocity relation

$$T_{on'}(u) = T_{n'o}(u) \tag{57}$$

The collision density in the subregions n' outside V_o is taken to be asymptotic at all lethargies including those near the resonance peak. Multiplying by the transfer probabilities $P_{on'}(u)$ the contribution to the collision density in region V_o from previous collisions in n' outside V_o becomes, using Eqs. (55-57)

$$\frac{1}{V_o} \sum_{n'} P_{on'}(u)\Sigma_{n'}\, \phi_{n'i}\, \Delta V_{n'} = \frac{1}{V_o} \sum_{n'} P_{n'o}(u) N_o\, V_o\, \sigma_t(u)\phi_{n'i} = N_o\, \sigma_t(u)\Phi_o\, P'_o(u) \tag{58}$$

where

$$\Phi_o \, P'_o(u) = \sum_{n'} P_{n'o}(u) \phi_{n'i} \tag{59}$$

The actual escape probability from V_o is

$$P_o(u) = \sum_{n'} P_{n'o}(u) \tag{60}$$

and becomes equal to $P'_o(u)$ for a flat flux. In fact for a flat flux, $\Phi_o = \phi_{n'i}$ for all n' in group i, a simple procedure is used to calculate $P_o(u)$ making use of the Dancoff factor C, which is the transmission probability through the subregions n' outside V_o

$$P_o(u) = \frac{P^o(u) \; (1-C)}{1 - C[1-N_o\sigma_t(u) \; \ell_o \; P^o(u)]} \tag{61}$$

where $P^o(u)$ is the escape probability through the outer surface of the isolated volume V_o, assumed to be the central region, and is calculated from Eq. (31). The lattice escape probability $P_o(u)$ of Eq. (61) makes use of the effective increase of the mean chord length ℓ_o, the diameter of the cylinder of volume V_o, by $(1-C)^{-1}$.

Using Eq. (58) the integral equation for $\Psi(u)$ becomes :

$$\Psi(u) = [1 - P_o(u)][T_a\Psi + T_1\Psi + T_2\Psi]_u + P'_o(u) \; \sigma_t(u)/E \tag{62}$$

where $\Psi(u) \to \Psi^{as}(u) = (\sigma_p + \sigma_1 + \sigma_2)/E$ for large $(E-E_o)$, — see Eq. 54 — and the probabilities $P_o(u)$ and $P'_o(u)$ are defined by Eqs. (59) and (60). The integral operators T_a and T_j (j = 1 or 2) operating on $\Psi(u')$ are

$$T_a = \frac{1}{1-\alpha_o} \int_{u-\ell n\frac{1}{\alpha_o}}^{u} du' \, \frac{\sigma_s(u')}{\sigma_t(u')} \quad , \quad T_j = \frac{1}{1-\alpha_j} \int_{u-\ell n\frac{1}{\alpha_j}}^{u} du' \, \frac{\sigma_{sj}}{\sigma_t(u')} \tag{63}$$

$\Psi(u)$ is calculated for a mesh of lethargy points near the resonance peak given approximately by

$$\varepsilon = \frac{2\Delta}{rE_o} \tag{64}$$

where Δ is the Doppler width defined in Eq. (50) and r is a quantity defined as

$$r = 5 + \frac{\Delta}{4\Gamma} \tag{65}$$

and has been found sufficiently large to cover the resonance shape well. In fact ϵ is adjusted slightly so that an even integral number (≤ 500) of mesh intervals cover the maximum collision lethargy range ($4/A_o$). Above an energy E_1 the asymptotic form $\Psi^{as}(u)$ of Eq. (54) is assumed to hold. This energy E_1 is chosen so that $(E_1 - E_o)$ is the larger of $(5 \cdot \frac{\Gamma}{2} \cdot \frac{\sigma_o}{\sigma_p}$ or $10\Delta)$ and is replaced by $\frac{4}{7} E_o$ if either quantity exceeds this value. Again E_1 is slightly adjusted to ensure that an even number of mesh intervals cover the energy range from E_1 to $[E_o - (E_1 - E_o)]$. Back values at the required number of mesh points - according to $4/(A_o\epsilon)$ - of $\Psi^{as}(u)$ are calculated at $E>E_1$ and make the successive evaluation of $\Psi(u)$ at all mesh points through the energy range near E_o possible. Slight adjustments of α_1 and α_2, as well as σ_1 and σ_2, are also made so that the ratios $\sigma_1/(1-\alpha_1)$ and $\sigma_2/(1-\alpha_2)$ are preserved, in order to ensure that the same mesh is applicable for the calculation of $T_1\Psi$ and $T_2\Psi$. Finally the resonance integral is obtained from

$$\int_{2E_o-E_1}^{E_1} \frac{\sigma_a(u)}{\sigma_t(u)} E\Psi(u)\,du \tag{66}$$

using numerical integration with the mesh points at which $\Psi(u)$ has been evaluated.

Three resonance wing corrections are applied to the resonance integral. The first extends the result to cover the range from E_1 to infinity and is a function of $z_1 = (E_1 - E_o)/E_o$. The second covers the energies from thermal cutoff E_c to the low limit of the effective range of the resonance $[E_o - (E_1 - E_o)]$. It is a function of z_2 which in general equals z_1 but is not allowed to exceed 4/7 and also is appropriately adjusted if $[E_o - (E_1 - E_o)] < E_c$. The third negative correction removes the 1/v effect of the resonance itself from E_c to infinity; it is based on the value of the Breit Wigner absorption cross section at $E_{th} = 0.0253$ eV. Assuming a 1/E flux for all these effects the total correction is

$$\left(\frac{1+3z_1}{z_1 y_1} - \frac{3}{2} \ln \frac{y_1+1}{y_1-1}\right) \frac{\sigma_o \Gamma\Gamma_\gamma}{4E_o^2} + \left[\frac{1-3z_2}{z_2 y_2} - \frac{1-3z_c}{z_c y_c} + \frac{3}{2} \ln\left(\frac{1+y_2}{1-y_2} \cdot \frac{1-y_c}{1+y_c}\right)\right] \frac{\sigma_o \Gamma\Gamma_\gamma}{4E_o^2}$$

$$- 2\sqrt{\frac{E_o}{E_{th}}} \cdot \frac{\sigma_o \Gamma\Gamma_\gamma}{4(E_o - E_{th})^2} \tag{67}$$

where $y_1 = \sqrt{1+z_1}$, $y_2 = \sqrt{1-z_2}$, $z_c = (E_o-E_c)/E_o$, $y_c = \sqrt{1-z_c}$ and σ_o is the peak resonance cross section of Eq. (50).

In the energy groups i in which the resonances are unresolved the resonance integral for a single resonance is obtained from Eqs. (66) and (62), in which the integral operators T_a, T_1 and T_2 of Eq. (63) are replaced by their Narrow Resonance equivalents σ_p/E, σ_1/E and σ_2/E respectively. The range of integration now covers all lethargies u.

$$I = \int \frac{\sigma_a(u)}{\sigma_t(u)} [\{1 - P_o(u)\}(\sigma_p + \sigma_1 + \sigma_2) + P'_o(u)\ \sigma_t(u)]du \qquad (68)$$

For a group flux which is nearly flat $P'_o(u) \simeq P_o(u)$ from Eq. (59). Its simplest rational approximation is

$$P_o(u) \simeq [1 + \frac{N_o\ \ell_o\ \sigma_t(u)}{1-C}]^{-1} \qquad (69)$$

where C is the Dancoff factor and ℓ_o the mean chord length. Introducing this expression into Eq. (68) in the absence of interference scattering

$$I = \frac{\sigma_B \Gamma \gamma}{E}\ J(\theta,\beta) \quad , \quad J(\theta,\beta) = \int_o^\infty \frac{\psi(x,\theta)}{\beta+\psi(x,\theta)}dx \quad , \quad x = \frac{2(E-E_o)}{\Gamma} \qquad (70)$$

where $\sigma_B = \sigma_p + \sigma_1 + \sigma_2 + \frac{(1-C)}{N_o \ell_o}$ and $\beta = \sigma_B/\sigma_o$

The geometrical cross section $(1-C)/(N_o\ell_o)$ is actually replaced by $(\sigma_p + \sigma_1 + \sigma_2)\ P'_o/(1-P_o)$, calculated in the absence of resonances, which allows approximately for the non flatness of the group flux, Eq. (59). Finally the resonance integral of Eq. (70) is averaged over the distribution of neutron widths and "summed" over the $\Delta E_i/D$ resonances occurring in the group i. (D is the average spacing between resonances).

$$I_i = \frac{1}{D} \int_{\Delta E_i} dE \int_o^\infty du\ \frac{e^{-u/2}}{\sqrt{2\pi u}}\ \frac{\sigma_B \Gamma \gamma}{E}\ J(\theta,\beta) \quad , \quad \Gamma_n = u<\Gamma_n^o>\sqrt{E} \qquad (72)$$

The integrals are evaluated numerically using a suitable integration mesh, and the procedure may be repeated for the separate s or p wave resonance sequences.

For each resolved resonance integral in group i, Eqs. (66) and (67), or the unresolved resonance integral of each sequence, Eq. (72), the

resonance absorption probability A_k for the nuclide under consideration is given by

$$A_k = \sum_r [1 - \exp\{-(I_i \Phi_o V_o)_k / \sum_n (q_i^o + \eta_i^o)_n \Delta V_n\}] \quad (73)$$

The flux Φ_o in group i and the volume V_o containing isotope k are given in Eq. (53), the slowing down density q_i^o in subregion n is given by Eq. (46), η_i^o is the corresponding expression for Hydrogen, and the summation $\sum_n$ covers the entire unit cell. The final summation over r covers all resolved resonances or unresolved sequences in group i and the group resonance escape probability is

$$(1 - R_i) = \prod_k (1 - A_k) \quad (74)$$

where R_i is the resonance absorption fraction appearing in Eq. (46) for the previous group.

Leakage

The leakage calculations are again based on the transport equation Eq. (1) with the Fourier Transform treatment of Eqs. (22-27), after homogenising the epithermal cross sections using the fluxes ϕ_{ni} obtained from Eq. (40). The lethargy variable is used throughout. The source of moment zero of Eqs. (24 and (25) is now replaced by the average fission source χ in group i and an average slowing down source ω into i due to inelastic scattering. The contribution due to elastic scattering is $\Sigma_s(u)\phi(u) - d\eta/du - dq/du$ where η and q are the slowing down densities due to Hydrogen and heavier isotopes. The latter satisfies the Groeling-Goertzel equation given in Eq. (45) and applied to the mixture of nuclides. The quantity η satisfies the corresponding exact differential equation (Selengut-Goertzel) with the Hydrogen scattering cross section Σ_s^H and $\xi=\lambda=1$. The first moment of the elastic slowing down source is denoted by ρ for Hydrogen and satisfies an exact differential equation, while the spproximate $\Sigma'_{s1}(u)\ J(u)$ is used for the other materials with combined first moment of the scattering cross section $\Sigma'_{s1}(u)$. Introducing all these effects into Eq. (25) with $F = F_o$ and $J = -F_1$ in group i, the MUFT equations (10) are obtained after rearrangement in the B-1 approximation.

$$BJ + (\Sigma_a + \Sigma_{in})F + E = -\frac{dq}{du} - \frac{d\eta}{du} + \omega + \chi \quad (75)$$

$$\frac{d\eta}{du} = -\eta + \Sigma_s^H F \quad (76)$$

$$\overline{\lambda}\,\frac{dq}{du} = -q + \xi\Sigma_s' F \tag{77}$$

$$(\gamma\Sigma_t - \Sigma_{s1})J = -\frac{2}{3}\frac{d\rho}{du} + \frac{1}{3}BF \tag{78}$$

$$\frac{2}{3}\frac{d\rho}{du} = -\rho + \frac{2}{3}\Sigma_s^H J \tag{79}$$

Here

$$\gamma = \frac{a^2}{3}\,\frac{(\tan^{-1}a)/a}{[1-(\tan^{-1}a)/a]}\,, \quad a = \frac{B}{\Sigma_t} \tag{80}$$

with $\tan^{-1}a$ replaced by $0.5\ \ell n[(1+a)/(1-a)]$ when $B^2<o$. The (P-1) approximation, analogous to Eq. (24) at thermal energies, is identical to Eqs. (75-79) with γ replaced by unity. If $\overline{\lambda}$ is replaced by zero the standard age approximation results and omission of $d\rho/du$ leads the basic Selengut-Goertzel method for Hydrogen. The resonance absorption rate is E.

The solution of Eqs. (75-80) is obtained after their integration over the energy group. Average group fluxes, currents and cross sections are used for this integration. The values of the slowing down densities are η_o, q_o and ρ_o at the low lethargy and η, q and ρ at the high lethargy. The integrated resonance absorption rate is

$$\int_{\Delta u_i} E\,du = (\eta_o + q_o)R \tag{81}$$

where R is the sum of the absorption fractions A_k of Eq. (73) for all resonance isotopes k. The five integrated equations resulting from Eqs. (75-79) have five unknowns in each group, η_o, q_o and ρ_o being known from the calculations in the previous group.

Few Group Parameters and Multiplication Factor

The 54 MUFT groups are ultimately combined into three broad groups, the first above 0.86 MeV, where fast fission is significant, the second covering the intermediate energies down to 5.5 keV and the third including the principal resonance absorption down to thermal energies. The thermal groups combined into one form the fourth broad group. For the three epithermal groups the quantities calculated are Σ_a, Σ_f, $\nu\Sigma_f$, D and Σ_R. The three first cross sections are defined so that reaction rates are preserved

$$\Sigma_x = \sum_i (\Sigma_x)_i\, F_i\, \Delta u_i / [\sum_i F_i\, \Delta u_i] \tag{82}$$

where F_i is the average flux obtained for group i in the previous MUFT calculations for the homogenised cell from Eqs. (75-79), and the summation covers all fine groups in a broad group. The diffusion constant is obtained from

$$D = \frac{1}{B} \sum_i J_i \, \Delta u_i / [\sum_i F_i \, \Delta u_i] \tag{83}$$

Finally for the first broad group the removal cross section - down scattering into the next group - is defined to be consistent with

$$(\Sigma_a + \Sigma_R + DB^2)_1 [\sum_i F_i \, \Delta u_i]_1 = [\sum_i \chi_i \, \Delta u_i]_1 = C_1 \tag{84}$$

while for the other two groups $(\Sigma_R)[\sum_i F_i \, \Delta u_i]$ for the previous group is added to the integrated fission spectrum of Eq. (84).

The final four group data are used to obtain k_{eff}, k_∞ and B_m^2, the material buckling, by rewriting the broad group equations in the form

$$(\Sigma_a + \Sigma_R + DB^2)_I \, \Phi_I = C_I + (\Sigma_R \Phi)_{I-1} \quad , \quad I = 1,2,3,4 \tag{85}$$

where the last term is omitted for I=1, and also C_4=o. For the input buckling the fluxes Φ_I calculated from Eq. (85) are clearly the same as the broad group fluxed $[\sum_i F_i \, \Delta u_i]$ of Eq. (84). Multiplying them by the broad group values of $(\nu\Sigma_f)_I$ the multiplication constant is obtained

$$k_{eff} = C_1[M_1+P_1M_2+P_1P_2M_3+P_1P_2P_3M_4] + C_2[M_2+P_2M_3+P_2P_3M_4] + C_3[M_3+P_3M_4] \tag{86}$$

where

$$M_I = [\nu\Sigma_f/(\Sigma_a + \Sigma_R + DB^2)]_I \quad , \quad P_I = [\Sigma_R/(\Sigma_a + \Sigma_R + DB^2)]_I \quad , \quad (\Sigma_R)_4 = 0 \tag{87}$$

With B^2=o in Eq. (87) the value of k_∞ results from Eq. (86), while for $B^2=B_m^2$ the eigenvalue k_{eff} = 1.

As in the case of the calculations at thermal energies the fluxes obtained from the integrated form of Eqs. (75-79) for the homogenised cell, and those from Eq. (40) for the infinite lattice may be combined as in Eq. (30) to give the leakage corrected flux throughout the unit cell of the lattice in all epithermal groups i.

ENDF/B-II Multigroup Cross Section Libraries

Thermal Library

The thermal cross section library for the isotopes encountered in the water moderated lattices was obtained by group averaging the values of σ_a, σ_f and σ_s. The slowing down source was obtained in accordance with Eq. (18).

The U-238 capture cross section in ENDF/B-II is given in considerably greater detail than in the first version of the data, and is slightly lower below 0.2 eV. The 2200 m/sec value is 2.72 b compared with 2.75 b previously. At energies above 0.2 eV the second version of ENDF has very slightly increased σ_c. For U-235 the value of ν at thermal energies is 2.423 compared with 2.43 previously. All the data for U-235 below 1 eV are given as smooth cross sections, whereas they were a combination of resonance and smooth data in ENDF/B-I. The 2200 m/sec values of σ_f and σ_c are 580.2 and 98.3 b respectively, so that $\alpha = 0.1694$ at that energy.

In the first version of ENDF/B the value of α was constant up to nearly 0.1 eV, above which the effect of the resonance at 0.29 eV becomes noticable. In ENDF/B-II α is lower by about 2 percent at 0.0253 eV and varies very slightly at energies below 0.1 eV.

The capture cross sections of the moderator isotopes are 0.332 b for Hydrogen, 5.1×10^{-4} b for Deuterium, and 1.4×10^{-4} b for Oxygen at 2200 m/sec, the latter value having been ignored in ENDF/B-I. For these nuclides and the cladding materials, Aluminium and the constituents of stainless steel, the appropriate thermal group averages of σ_c were used from ENDF/B-II. Thermal scattering kernels, Eqs. (16) and (17), were obtained with the FLANGE code [7] for H and D in light and heavy water respectively. They were based on the Haywood [13] scattering function $S(\alpha,\beta,T)$. For Oxygen and Aluminium gas kernels were used, while other materials were treated as heavy scatterers.

The overall effect of the changes between the first two versions of ENDF/B on the thermal lattice parameters was very small. In a typical D_2O moderated lattice with 1.0 cm radius U-metal rods and 7.0 cm pitch the one thermal group value of η changed from 1.3054 to 1.3038 when comparing ENDF/B-I and II. Similarity f changed from 0.9732 to 0.9727, so that ηf and consequently the multiplication factor was reduced by 0.2 percent. It should be pointed out also that the inclusion of a

scattering ring outside the cylindrised unit lattice cell for the spectrum calculations produced a negligible effect on ηf (less than 0.1 percent) even for very tight light water moderated slightly enriched Uranium lattices and a large rod diameter.

Epithermal Library

The differences of format [14] of the nuclear data in ENDF/B-I and II necessitated considerable modifications of the ETOM code [15] which produces the multigroup cross sections for the epithermal calculations. The modified code [16] can convert p wave resolved resonances, or all resonances of structural materials, to smooth cross sections; it can handle energy-dependent unresolved resonance parameters and convert these to smooth cross sections for all resonance sequences or for the non-s wave resonances only; and finally it can deal with the inelastic scattering cross sections for excitation of discrete levels and the continuum, as given in ENDF/B-II File 3, instead of the secondary neutron energy distributions given in ENDF/B-I File 5.

The library data for the materials encountered in the lattices studied were obtained for the 54 MUFT groups using a 1/E weighting function joined to a fission spectrum at 68 keV.

For U-238 the nuclear data are given as smooth cross sections below 5 eV in both the first two versions of ENDF/B. This energy is sufficiently close to the large resonance at 6.67 eV to be still within the range of the detailed resonance integral calculations of Eq. (66). Consequently the resolved energy range must be extended effectively somewhat below 5 eV in order to ensure that shielding effects are properly accounted for in the case of the most important resonance. It should be mentioned also that the wing corrections, Eq. (67), remove the entire 1/v effect of the resolved resonances above the thermal cutoff E_c from the capture resonance integral. The corresponding average group cross section for a 1/E flux must therefore be added to the smooth capture cross section in each epithermal group. This correction is based on σ_c = 2.51 barns at 0.0253 eV, which is the combined 2200 m/sec cross section of all resonances listed in ENDF/B-II obtained with the Breit-Wigner formula. In ENDF-I these smooth capture group cross sections were based on the actual σ_c = 2.73 barns at 2200 m/sec, since in this compilation (contrary to ENDF/B-II) a resonance with peak at negative energies was included. In addition to the 1/v effect,

smooth corrections to the resonance capture cross section are given in the second version of the data and not in the first. ENDF/B-I contains also smooth cross sections in place of the p wave resonances in the resolved resonance range, while in ENDF/B-II the p wave resolved resonance parameters themselves appear. As the latter are weak they were converted to smooth cross sections with the modified ETOM code [16]. The various effects at the energies of the resolved resonances are summarised in Table I. The contribution of the 1/v tail is clearly considerable in situations in which the effective resonance integral is only of the order of 10 barns. In addition these capture integrals, treated as unshielded, are larger in the second version of the nuclear data.

TABLE I. CAPTURE INTEGRALS (BARNS) OF ^{238}U BETWEEN 5 AND 3920 eV.

	ENDF/B-I	ENDF/B-II
1/v tail of σ_c	1.08	1.00
Smooth file 3 corrections	-	0.57
Resolved p-wave resonances	0.40	0.26
Total	1.48	1.83

In the unresolved energy region energy dependent parameters are given for U-238 for both the s and the p wave sequences in ENDF/B-II. The smooth corrections are very small. In Table II the approximate parameters are shown (ignoring their small energy dependence in ENDF/B-II), as well as the unshielded capture integrals and the shielded values for a lattice of 1.0 cm radius natural Uranium rods in D_2O at 7 cm pitch. The shielding effect is clearly significant. The unresolved resonance integral for p wave resonances exceeds that for the s wave sequence. Consequently in the U-238 epithermal library the unresolved p wave resonance parameters were included so that these were treated as in the case of the s waves by Eq. (72) to obtain the resonance integral.

For U-235 the same resolved resonance parameters appear in both the first two versions of the ENDF/B data. On the other hand instead of starting at 10^{-3} eV, the unresolved resonance region starts only at 1 eV in ENDF/B-II, all cross sections being smooth data below this energy.

TABLE II. APPROXIMATE UNRESOLVED RESONANCE PARAMETERS (eV) AND CAPTURE INTEGRALS (BARNS) FOR ^{238}U

		ENDF/B-I	ENDF/B-II
$\ell = 0, \quad J = \frac{1}{2}$	D	18.5	20.1
	Γ_n^o	0.00174	0.00182
	Γ_γ	0.0246	0.0235
$\ell = 1, \quad J = \frac{1}{2}$	D	18.5	20.1
	Γ_n^o	0.00292	0.0040
	Γ_γ	0.0246	0.0235
$\ell = 1, \quad J = \frac{3}{2}$	D	9.25	11.0
	Γ_n^o	0.00146	0.0022
	Γ_γ	0.0246	0.0235
s-wave capture integral	Unshielded	1.26	1.10
	Shielded*	0.52	0.45
p-wave capture integral	Unshielded	1.00	1.43
	Shielded*		0.96

*For a lattice of 1 cm radius natural U rods in D_2O at 8 cm pitch.

The second version contains s and p wave energy dependent unresolved resonance parameters from 64.5 eV to 24.8 keV which were converted to smooth cross sections by the ETOM code using a procedure essentially equivalent to Eq. (72), but averaging over the distribution of fission widths as well and allowing for a flux depression factor [15]. These conversions were made at infinite dilution.

For both U-235 and U-238 the scattering cross section in the groups at resonance energies was replaced by $\sigma_p(=4\pi R^2)$. This is consistent with the resonance treatment described and also for comparison with Monte Carlo calculations [3] in which the resonance integrals are derived from the resonance reaction rates using the background value of σ_s for all nuclides.

The resonances of the constituents of the stainless steel cladding were all converted to smooth cross sections and for these nuclides, for which no shielded capture integral calculations are made, the group average resonance σ_s was included in the cross section library.

As regards the moderator nuclides it should be mentioned that σ_s for Deuterium in ENDF/B-II is 3.35 barns below about 10 keV compared with the value of 3.182 barns in the previous version. The higher value is consistent with the neutron age values in D_2O [3].

In ENDF/B-III the resolved resonance data for U-235 have been somewhat extended, and for U-238 the unresolved resonance parameters modified so as to reduce the resonance integrals slightly.

Comparisons Between the HAMMER Lattice Analysis and Monte Carlo Calculations

Three principal areas have been selected for the comparison of parameters obtained by the HAMMER lattice analysis code and Monte Carlo calculations. The statistical estimates were obtained with programs utilising the same basic nuclear data and treating the exact lattice geometry.

The first aspect dealt with relates to the calculation of resonance integrals. The values based on the Nordheim treatment, which is made use of in the HAMMER code in the manner described previously, are overestimates resulting from an individual resonance treatment. It is assumed that the flux recovers completely between resonances so that the absorption integral can be obtained from the collision density calculated by numerical integration of its integral equation and taking its asymptotic form above the resonance to be that which would apply in the absence of absorption. This assumption is particularly severe for U-235 with its small resonance spacing. In addition the flux depression due to one resonance at the peak of a resonance of another sequence, or another nuclide, is completely ignored, an effect which is again quite significant for U-235. As regards the U-238 resonance wing corrections these are evaluated in the absence of flux depression and temperature dependence of the cross sections. At any one neutron energy these effects are quite small, but the integrals are extended to infinity and to thermal cut-off. For reasons of convenience most of the wing contribution is subtracted from the calculated resonance integral as a 1/v term and added again as a smooth absorption cross section which, as shown in Table I has a considerable capture integral. Its magnitude is a measure of that part of the resonance integral which is unshielded in the Nordheim treatment.

The Monte Carlo calculations have been reported previously [13], [17] and were made for ENDF/B-I data. In the code the resonance cross sections at each neutron energy are calculated exactly from the ENDF resonance para-

meters and the smooth point cross section data as appropriate. At the energies where the resonances are unresolved an individual resonance ladder is generated for each neutron history and for each of the relevant resonance sequences. The comparisons with the Nordheim calculations are made after reducing the Monte Carlo values by all the smooth capture integrals in the MUFT groups which are treated as unshielded cross sections in the HAMMER program. Consequently corresponding quantities are compared, which are in fact the total capture integrals of the U-238 resolved resonances and the unresolved s wave resonance sequence but excluding the 1/v effect of the

TABLE III. COMPARISON OF HAMMER LATTICE ANALYSIS AND MONTE-CARLO CALCULATIONS FOR BNL SLIGHTLY ENRICHED URANIUM METAL – H_2O HEXAGONAL LATTICES

Rod Diameter (inch) Volume Ratio	0.25 1.5		0.60 1.5		0.25 3.0		0.60 3.0	
	HAMMER	M.C.	HAMMER	M.C.	HAMMER	M.C.	HAMMER	M.C.
F - 28*	.0488	.0416 (± .0010)	.0519	.0448 (± .0013)	.0340	.0300 (± .0010)	.0372	.0333 (± .0014)
n - 28		.0040 (± .0009)		.0049 (± .0008)		.0052 (± .0009)		.0058 (± .0013)
F - 25	.0426	.0412 (± .0006)	.0523	.0516 (± .0008)	.0255	.0252 (± .0005)	.0309	.0310 (± .0009)
C - 28	.282	.253 (± .004)	.249	.221 (± .003)	.186	.167 (± .004)	.149	.142 (± .004)
C - 25	.0201	.0193 (± .0004)	.0240	.0235 (± .0004)	.0121	.0119 (± .0003)	.0142	.0142 (± .0005)
R_1	.690	.686 (± .007)	.687	.670 (± .007)	.709	.700 (± .007)	.705	.682 (± .009)
R_2	.879	.885 (± .002)	.875	.877 (± .002)	.918	.925 (± .002)	.920	.922 (± .002)
Age (cm^2)	34.6	33.5 ± 0.7	29.3	28.5 ±0.5	30.5	29.9 ± 0.7	27.4	26.6 ±0.8

* All reaction rates refer to one fission neutron injected into the unit cell of the lattice

F epithermal fission rate

n (n,2n) reaction rate

C epithermal capture rate

R_1 slowing down rate past 0.82 Mev

R_2 slowing down rate past 5.5 Kev

Age from fission to 0.625 ev

resonances themselves in a 1/E flux. The unresolved p wave sequences were treated as unshielded in the original ENDF/B-I calculations [3]. It was found that the resolved resonance integrals of U-238 agreed in the first two versions of ENDF/B and small differences in the unresolved s wave contributions could be allowed for. Monte Carlo calculations [17] were therefore only made for lattices which had not been studied before. The statistical uncertainties of the effective resonance integral were of the order of 1 to 2 percent for U-238 and three to four times larger for the resolved resonances of U-235 (below 65 eV). Table VIII compares the values of the absorption integrals for both isotopes as made with the Nordheim and Monte Carlo procedures for the lattices referred to in Tables IV to VII. The former values were multiplied by appropriate factors to reduce them to the latter in the results reported for all the lattices in these tables (IV to VII). It should however be borne in mind that the correction factors for the resonance integrals are uncertain by the statistical errors of the Monte Carlo values, which affect both the resonance escape probability and the contribution to k_{eff} due to resonance fission in U-235.

The second area treated by a Monte Carlo code was the thermal spectrum [18] in a periodic lattice of infinite extent. Comparisons with THERMOS were made for the thermal parameters including $\langle r^2 \rangle$, the mean square distance between the point of injection of the thermal neutron into the lattice and its subsequent absorption. The Monte Carlo program made use of the same scattering kernels Eq. (17) and other thermal nuclear data as the spectrum calculations based on integral transport theory which have been described before. The use of a thirty group thermal cross section set for the Monte Carlo calculations instead of point cross section values is a limitation which was necessitated by the difficulties arising from the use of the scattering function $S(\alpha,\beta,T)$ itself, Eq. (16), for the random choice of the scattering angle and neutron energy after scattering. However a six term Legendre expansion of the scattering function, obtained with the FLANGE code [7], was used in the stochastic calculations. The lattices studied [18] were heavy water moderated systems in which thermal diffusion area considerably exceeds that at epithermal energies. The results showed good agreement of all thermal parameters with the values obtained by THERMOS, except in the case of L^2 which was a few percent lower in the Monte Carlo runs. The effect on k_{eff} amounted to an increase of 0.5 percent for natural Uranium rods of 1.5 cm radius in D_2O near the maximum of the curve of buckling against pitch. At larger rod sizes and

TABLE IV. WÜRENLINGEN NATURAL U – D_2O SQUARE LATTICES (1.0-cm FUEL ROD RADIUS, 0.025 cm AIR GAP, 0.075 cm ALUMINIUM CLADDING)

Pitch (cm) (10^6 B^2 cm^{-2})	D_2O %	Group 1 Σ_a	$\nu\Sigma_f$	Σ_R	D (k_1)	Group 2 Σ_a	$\nu\Sigma_f$	Σ_R	D (k_2)	Group 3 Σ_a	$\nu\Sigma_f$	Σ_R	D (k_3)	Group 4 Σ_a	$\nu\Sigma_f$	D (k_4)	Total k_{eff}
8.0 (780)	99.70	.00382	.00802	.0874 (.0651)	1.94	.00097	.00011	.0349 (.0028)	1.16	.00324	.00092	.0166 (.0402)	1.20	.00903	.01159 (.8692)	.827	.9773
9.0 (835)	99.62	.00369	.00768	.0874 (.0624)	1.94	.00080	.00009	.0354 (.0023)	1.16	.00257	.00073	.0172 (.0320)	1.21	.00724	.00927 (.8826)	.818	.9793
10.0 (840)	99.74	.00360	.00746	.0873 (.0607)	1.95	.00069	.00008	.0356 (.0020)	1.17	.00208	.00059	.0174 (.0263)	1.22	.00590	.00755 (8914)	.817	.9804
12.0 (757)	99.74	.00350	.00720	.0872 (.0588)	1.96	.00054	.00006	.0360 (.0016)	1.18	.00145	.00041	.0180 (.0187)	1.23	.00414	.00527 (.9054)	.810	.9845
14.0 (647)	99.74	.00345	.00707	.0872 (.0579)	1.96	.00046	.00006	.0363 (.0014)	1.18	.00107	.00030	.0184 (.0140)	1.23	.00306	.00387 (.9139)	.806	.9872
16.0 (506)	99.49	.00342	.00700	.0871 (.0575)	1.97	.00041	.00005	.0369 (.0013)	1.18	.00083	.00023	.0189 (.0108)	1.23	.00239	.00295 (.9193)	.794	.9889

Comparison with ENDF/B-1

Pitch (cm)	8.0	10.0	12.0	14.0	16.0
k_{eff} (ENDF/B-I)	.9814	.9840	.9873	.9893	.9888
k_{eff} (ENDF/B-II)	.9773	.9804	.9845	.9872	.9889

TABLE V. BNL 1.03% ENRICHED U-H_2O HEXAGONAL LATTICES (0.25 in. FUEL ROD DIAMETER, 0.002 in. AIR GAP, 0.031 in. ALUMINIUM CLADDING)

Volume Ratio (10^6 B^2 cm^{-2})	Group 1				Group 2				Group 3				Group 4			Total
	Σ_a	$\nu\Sigma_f$	Σ_R	D	Σ_a	$\nu\Sigma_f$	Σ_R	D	Σ_a	$\nu\Sigma_f$	Σ_R	D	Σ_a	$\nu\Sigma_f$	D	k_{eff}
	(k_1)				(k_2)				(k_3)				(k_4)			
1.5	.00768	.01756	.0870	1.85	.00462	.00071	.0712	.997	.03002	.00932	.0600	.757	.09504	.12574	.303	
(1214)		(.1363)				(.0085)				(.0875)				(.7416)		.9739
2.0	.00682	.01556	.0898	1.87	.00404	.00062	.0815	1.00	.02747	.00816	.0714	.731	.08800	.11384	.270	
(1995)		(.1166)				(.0065)				(.0697)				(.7846)		.9774
3.0	.00562	.01272	.0933	1.90	.00322	.00049	.0955	1.02	.02299	.00655	.0875	.698	.07617	.09409	.233	
(2515)		(.0923)				(.0044)				(.0509)				(.8332)		.9808
4.0	.00483	.01085	.0953	1.93	.00266	.00040	.1046	1.03	.01951	.00547	.0983	.679	.06730	.07939	.213	
(2207)		(.0782)				(.0034)				(.0409)				(.8615)		.9840

TABLE VI. HARWELL 0.928% ENRICHED U-H_2O SQUARE LATTICES (1.20 in. DIAMETER, 0.0075 in. AIR GAP, 0.020 in. ALUMINIUM CLADDING)

Volume ratio (10^6 B^2 cm^{-2})	Group 1				Group 2				Group 3				Group 4			Total
	Σ_a	$\nu\Sigma_f$	Σ_R	D	Σ_a	$\nu\Sigma_f$	Σ_R	D	Σ_a	$\nu\Sigma_f$	Σ_R	D	Σ_a	$\nu\Sigma_f$	D	k_{eff}
	(k_1)				(k_2)				(k_3)				(k_4)			
0.590	.01309	.03001	.0929	1.65	.00772	.00117	.0500	.905	.03391	.01261	.0406	.687	.11902	.15831	.333	
(-1020)		(.2164)				(.0189)				(.1381)				(.5926)		.9660
0.896	.01158	.02671	.0956	1.68	.00661	.00100	.0635	.919	.03010	.01112	.0561	.672	.10560	.13689	.281	
(1110)		(.1842)				(.0127)				(.1036)				(.6756)		.9761
1.402	.00985	.02274	.0980	1.72	.00545	.00082	.0785	.938	.02546	.00932	.0741	.656	.08656	.10669	.235	
(2410)		(.1528)				(.0086)				(.0760)				(.7400)		.9774
1.931	.00866	.01996	.0992	1.76	.00468	.00069	.0887	.954	.02205	.00798	.0869	.645	.07233	.08418	.210	
(2190)		(.1344)				(.0066)				(.0615)				(.7749)		.9774

TABLE VII. UO_2 – H_2O SQUARE LATTICES

Volume Ratio 10^6 B^2 cm^{-2})	Group 1 Σ_a	$\nu\Sigma_f$	Σ_R	D	Group 2 Σ_a	$\nu\Sigma_f$	Σ_R	D	Group 3 Σ_a	$\nu\Sigma_f$	Σ_R	D	Group 4 Σ_a	$\nu\Sigma_f$	D	Total k_{eff}
		(k_1)				(k_2)				(k_3)				(k_4)		
B & W*																
0.959	.00555	.01219	.0776	1.82	.00368	.00176	.0662	.977	.03366	.02290	.0531	.691	.15372	.25613	.314	.9746
(7900)		(.0941)				(.0192)				(.1793				(.6820)		
B & W*																
1.139	.00519	.01136	.0798	1.82	.00340	.00163	.0722	.981	.03159	.02127	.0600	.683	.14688	.24314	.294	.9797
(8824)		(.0846)				(.0162)				(.1572)				(.7217)		
WINF**	.00584	.01287	.0793	1.81	.00370	.00142	.0676	.963	.03222	.01868	.0551	.692	.13611	.22060	.309	.9866
1.001		(.0997)				(.0157)				(.1527)				(.7185)		
(6600)																

* Babcock & Wilcox 4.02% enriched UO_2 lattices (0.444 inch diameter rod, 0.016 inch stainless steel cladding)

** Winfrith 3.003% enriched UO_2 lattices (1.0119 cm diameter rod, 0.0136 cm air gap, 0.0267 cm stainless steel cladding)

TABLE VIII. RESONANCE INTEGRALS* FOR THE LATTICES OF TABLES IV TO VII, AS OBTAINED BY THE HAMMER CODE (R) AND AFTER CORRECTION IN ACCORDANCE WITH THE MONTE-CARLO ESTIMATES R'

Lattice	Rod Diameter	Pitch (cm) or Volume Ratio	U-235 R	U-235 R'	U-238 R	U-238 R'
Würenlingen U-metal	2.0 cm	8.0 cm	216.8	188.8	10.63	9.66
		9.0 cm	217.1	189.1	10.69	9.69
		10.0 - 16.0 cm	217.3	189.3	10.71	9.71
BNL U-metal	0.25"	1.5	226.6	207.8	14.00	13.22
		2.0	230.0	213.0	14.75	14.25
		3.0	233.5	221.3	15.65	15.38
		4.0	235.2	227.0	16.14	15.85
Harwell U-metal	1.20"	0.590	185.5	146.6	8.35	7.39
		0.896	191.5	158.0	8.74	7.86
		1.402	195.2	173.8	8.99	8.45
		1.931	196.7	186.8	9.09	8.94
B & W UO_2	0.444"	0.571	212.2	200.1	17.44	16.50
		0.595	214.4	202.2	17.90	16.93
Winfrith UO_2	1.012 cm	1.00	218.5	206.0	17.16	16.23

* Resolved resonances, and unresolved s and p wave sequences; not including 1/v capture integral in 1/E flux for U-238.

larger values of the pitch the increase of k_{eff} arising from the Monte Carlo estimates of L^2 was even smaller.

A Monte Carlo program based on the MOCA [19] code was also written to treat epithermal effects in accordance with ENDF/B-II specifications of the cross sections. Neutrons were injected into the periodic lattice at

random energies corresponding to the simple fission spectrum $[4E/(\pi\theta^3)]^{1/2}\exp(-E/\theta)$ for neutrons from thermal fissions. The scattering was treated taking the full anisotropy as given in File 4 of the data into account. The procedure adopted to select the random cosine μ_c of the scattering angle in centre of mass coordinates from the Legendre moments $f_\ell(E)$ of the angular distribution $p(\mu_c,E)$, was the same as that used at thermal energies. In fact μ_c is the solution of

$$r = \int_{-1}^{\mu_c} p(\mu'_c,E)d\mu'_c = \frac{1}{2}(\mu_c+1) + \frac{1}{2}\sum_{\ell=1}^{L} f_\ell(E)\,[P_{\ell+1}(\mu_c) - P_{\ell-1}(\mu_c)] \quad (88)$$

where r is a random number selected in the range (0,1) from a uniform random number generator in (0,1) and $f_\ell(E) = \int^1 p(\mu_c,E)\,P_\ell(\mu_c)d\mu_c$ are the tabulated moments truncated at ℓ=L. The choice of μ_c and a random azimuth angle lead to the neutron energy after scattering and its new direction. For inelastic scattering the angular distribution is isotropic in the centre of mass system and from the Q values of the levels excited, as specified with the cross section data the energy of the scattered neutron is found from

$$E' = E[(A^2+1) + 2A\sqrt{1 - \frac{|Q|(A+1)}{AE}}\,\mu_c]/(A+1)^2 - |Q|A/(A+1) \quad (89)$$

where μ_c is chosen from a uniform distribution in (-1, 1). When the evaporation model is appropriate E' is selected from the distribution $E'/[\Theta(E)]^2\exp[-E'/\Theta(E)]$, $0<E'<E-U$ where $\Theta(E)$ and U are given in the nuclear data.

The output of the code was prepared in accordance with that of the HAMMER code, reaction rates being given in three broad epithermal groups with lower energies at 0.82 MeV, 5.5 keV and 0.625 eV, as well as for the entire epithermal range of energies. In Table III comparisons are shown for some BNL slightly enriched lattices of Uranium metal rods in H_2O. For the rods of 0.25" diameter the enrichment was 1.03 percent, and the Aluminium cladding thickness 0.033" including a 0.002" air gap. For the rods of 0.60" diameters the corresponding data were 1.30 percent, 0.033" and 0.005", respectively. The Table lists fission and capture rates in U-238 and U-235 per fission neutron injected into the unit cell for the entire epithermal energy range. Probable errors on the fifty percent level are also shown. The values listed for the

corresponding runs with the HAMMER analysis code also refer to ENDF/B-II data. Here the U-238 fission rate includes the effect of the n-2n cross section at high energies which is given separately in the Monte Carlo calculations although statistical uncertainties are quite considerable. The resonance absorption rates are included in the Monte Carlo values only approximately; group cross sections, based on the effective resonance integrals, replace the actual point values of the resonance cross sections in each group of the MUFT structure. Finally removal rates past the low energies of the first two broad groups as well as the age from fission to thermal energies are given.

The results show reasonable agreement between the two methods of calculation allowing for the approximate nature of the resonance treatment in the Monte Carlo code. There is no significant difference in the age. The values of k_{eff} obtained with the HAMMER code therefore account adequately for the leakage in light water moderated systems. The production rates at high energies may be used to estimate the accuracy of the integral transport theory calculations where scattering anisotropy is significant. In the first lattice for example the lower production rate in U-238 (by fission and the n-2n reaction) as obtained by the Monte Carlo values suggests a slightly softer spectrum. but the statistical accuracy is not sufficient to draw precise conclusions.

In general the comparisons with Monte Carlo calculations did not reveal any considerable inaccuracies in the lattice analysis code except in the case of resonance capture where appropriate corrections were made (Table VIII).

Lattices Studied With the HAMMER Code

In Tables IV to VII detailed results are given of a number of heavy and light water moderated lattices analysed with the HAMMER code using corrected resonance integrals and ENDF/B-II data. The four groups refer to the energies of the fast fission of U-238 above 0.82 MeV, the intermediate region between 0.82 MeV and 5.5 keV, the energies of the U-238 resolved resonances down to 0.625 eV, and the thermal group. The cross sections listed in each group are the ones obtained after unit cell averaging in the absence of leakage for each of the 54 epithermal and 29 thermal groups, and subsequent group collapse after the leakage flux in the homogenised system has been calculated. The values of

k_{eff}, Eq. (86), refer to the experimental bucklings listed. All the calculations are made in accordance with the methods discussed in this paper and the resonance integrals were corrected to those obtained by the Monte Carlo calculations as given in Table VIII.

Table IV gives the results of a set of Würenlingen Natural Uranium D_2O moderated lattices [1]. They are well moderated systems with about 90 percent thermal fissions. In Table V BNL light water moderated slightly enriched U-metal lattices with thin rods are analysed [2]. The Harwell lattices [2] of Table VI have a considerably larger rod diameter and go down to extremely low values of the water to metal volume ratio, systems in which there are about 40 percent of epithermal fissions. Finally some 3 to 4 percent enriched UO_2 lattices [2] are given in Table VII with 25 to 30 percent epithermal fissions.

The values of k_{eff} for all the metal lattices described were about 2 percent less than unity. For the systems of Table IV and V, which had previously been treated with ENDF/B-I data [3], this is consistent with the previous results and the details given regarding the capture cross section of U-238. At epithermal energies the capture integral is rather larger for the ENDF/B-II compilation. There are uncertainties in the final results because of the statistical errors of the Monte Carlo calculations which were used to obtain the corrected effective resonance integrals. However even allowing for errors of about 1 percent in the U-238 and 3 percent in the U-235 resonance integrals the general characteristics of the results obtained remain unaltered. The greatest deviation from unity is for the tightest lattices with very high contributions to k_{eff} from epithermal fissions.

Similar trends were found for the UO_2 water moderated lattices. The k_{eff} values were low by about the same percentage as in the case of the U metal/H_2O lattices.

Conclusion

A number of clean U-metal and UO_2 fueled thermal reactor lattices have been analysed with the HAMMER code to investigate the adequacy of the methods of analysis and nuclear data used. The principal area of difficulty is still that of the resonance absorption both as regards the calculational procedure and from the point of view of the nuclear data.

Regarding the data the low limit of the U-238 resonance regions at 5 eV is close to the resonance which accounts for a very large part of the resonance capture; a lower limit is desirable for certain codes, such as the ones described, which calculate effective resonance integrals. The artificial separation of resonance and smooth absorption made in the HAMMER analysis leads to certain problems which are absent if Monte Carlo calculations are used instead. On the other hand statistical uncertainties now appear.

At the energies of the fission spectrum and in the thermal region no serious discrepancies between the HAMMER analysis and the Monte Carlo calculations were found.

The multiplication factor of the U metal H_2O and D_2O moderated clean lattices was found to about 2 percent less than unity for the ENDF/B-II data. Similar results were obtained for UO_2 light water moderated lattices. Much of the discrepancy appears to be due to the U-238 resonance capture calculations and data.

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DISCUSSION

R.A. BONALUMI: Your main conclusion is that your k_{eff} values calculated on the basis of the HAMMER code system are consistently below unity; this is also the case for oxide lattices, contrary to the table in the paper. I remember using HAMMER a few years ago, in conjunction with oxide-fuelled light-water lattices, and the problem there was that k_{eff} was generally above unity. A popular explanation for this (but one that I never looked into in detail) was that the ^{238}U resonance absorption cross-section was somehow shielded twice in the HAMMER scheme. Do you have any comment on this point? The Canadians seem to be using HAMMER successfully now, even for D_2O lattices.

W. ROTHENSTEIN: I do not think that the ^{238}U resonance absorption is shielded twice. The methods adopted in the version of HAMMER used by us are fully described in the paper.

W.B. LEWIS: On the question of the use of HAMMER in Canada for heavy-water lattices, I can confirm that this has been achieved satisfactorily. However, I wish to state emphatically that this should not be taken as a general approval, as many adjustments were necessary.

W. ROTHENSTEIN: I should like to comment that the results presented here for ENDF/B-II data have not been subjected to any adjustments. They are consistently low in k_{eff} for metal- and oxide-fuelled systems. It should be noted that Table VII of the full paper was corrected to eliminate an

error in our thermal capture cross-sections of the constituents of stainless steel.

G. CASINI: Do you intend to extend your ENDF/B data checking to homogeneous, hydrogen-containing ^{235}U and ^{233}U systems, where the absence of heterogeneity effects provides a basis for direct comparisons of number of the nuclear data of interest in light-water systems? I would also be interested in knowing whether you intend to extend your analysis to plutonium-bearing water systems, both homogeneous and heterogeneous.

W. ROTHENSTEIN: I think such calculations on the homogeneous systems referred to by Dr. Casini would certainly be very useful. We do intend to study plutonium-fuelled systems.

Section X

ACTIVATION ANALYSIS: GENERAL

Chairman

B. GRINBERG (France)

TENDANCES MODERNES DE L'ANALYSE PAR ACTIVATION

Ph. ALBERT
CNRS, Laboratoire d'analyse par activation Pierre Sue,
Centre d'études nucléaires de Saclay, France

Abstract–Résumé

MODERN TRENDS IN ACTIVATION ANALYSIS.

The author reports on two important meetings held at Saclay in October 1972: the Fourth International Conference on Modern Trends in Activation Analysis and the International Colloquium of the CNRS on the Activation Analysis of Microquantities of Elements; he gives a brief survey of the topics discussed and the trends resulting from the meetings.

TENDANCES MODERNES DE L'ANALYSE PAR ACTIVATION.

L'auteur rend compte de deux importantes réunions qui ont eu lieu à Saclay en octobre 1972: la Quatrième Conférence internationale sur les tendances modernes de l'analyse par activation et le Colloque international du CNRS sur l'analyse par activation de micro-quantités d'éléments; il passe rapidement en revue les thèmes discutés et les tendances qui s'en sont dégagées.

En octobre 1972 se sont tenues à Saclay deux réunions internationales sur l'analyse par activation. L'une était la Quatrième Conférence internationale sur les tendances modernes de l'analyse par activation, et l'autre un Colloque international du CNRS sur l'analyse par activation de micro-quantités d'éléments. Ces deux réunions étaient organisées conjointement par le CEA et le CNRS, sous les présidences des Directeurs du laboratoire d'analyse par activation Pierre Sue.

Ces deux réunions ont rassemblé 380 participants. Trente-cinq pays étaient représentés et nous avons eu le plaisir de réunir, à côté de délégations très importantes de différents grands laboratoires des Etats-Unis d'Amérique et de tous les pays, une délégation particulièrement importante de l'URSS puisqu'il y avait 14 participants appartenant à quelques-uns des plus grands laboratoires spécialisés en analyse par activation.

Le nombre des scientifiques invités au Colloque international du CNRS était de l'ordre de 70 à 100 suivant les sessions. Il est évident que bien des scientifiques étaient, selon les journées, intéressés plus ou moins par le Colloque ou par les sessions de la Conférence.

Si ce qui caractérisait, d'une façon générale, les thèmes de la Conférence était la méthodologie en matière d'analyse par activation, la principale particularité du Colloque était de s'intéresser essentiellement à l'analyse des traces d'impuretés dans des matériaux de toutes natures, depuis les minéraux jusqu'au milieu biologique en passant par les métaux très purs, les semi-conducteurs et même des échantillons lunaires comparés à des échantillons terrestres. Ce qui caractérisait également les travaux du Colloque était que les études discutées concernaient les difficultés rencontrées et les solutions choisies pour résoudre des problèmes concrets dans le domaine du dosage de micro-quantités d'éléments.

Je ferai une mention particulière de quelques conférences et communications. Au cours de la première session commune au Colloque et à la Conférence, M. Meinke, Chef de la Division de chimie analytique du National Bureau of Standards, nous a bien montré les objectifs à atteindre pour l'analyse par activation:

a) Développer davantage la méthode sous tous ses aspects sur des échantillons réels pour la solution de problèmes scientifiques et techniques.

b) Améliorer avant tout la précision des dosages (à ce point de vue le Dr Brunninx, des laboratoires Philips, nous a donné un bel exemple de réussite).

c) Grâce à son exactitude et en tenant compte de la précision alors obtenue et des prix des analyses, rechercher les applications spécifiques de cette méthode sous ses différentes formes.

Dès la Conférence du Dr Amsel, nous sommes entrés dans le vif du sujet avec les magnifiques exemples d'applications en physique et chimie que nous a décrits ce spécialiste des méthodes des réactions nucléaires ou de rétrodiffusion. Il s'agit de développements nouveaux pour l'analyse de techniques nucléaires qui consistent à mesurer les rayonnements émis pendant l'irradiation des échantillons par des particules chargées (p, d, α, ^{3}He). Par exemple: dosage de l'oxygène-18 par détection quantitative des α émis par les réactions $^{18}O(p,\alpha)^{15}N$. Il est ainsi possible de doser dans des couches minces des quantités de 10^{-12} g d'oxygène par cm^2 de cible avec une résolution en épaisseur de 150 à 1000 Å.

Dans le domaine des études sur la radiochimie et les analyses de métaux, de très grands progrès ont été faits depuis 4 ans, et ceci aussi bien en chimie qu'en détection, par l'amélioration et la généralisation de l'emploi de bons détecteurs à cristaux Ge-Li; c'est une des importantes conclusions de ces journées. Un autre aspect des sessions de ces journées, c'est la preuve éclatante du développement de l'emploi des activations par photons γ et particules chargées pour doser les traces d'éléments légers dans les métaux purs.

Ces méthodes ont maintenant établi qu'il est possible de préparer des métaux à moins de 1 et même 0,5 ppm de carbone et d'oxygène. Des exemples concrets ont montré les possibilités très intéressantes des particules de 11 MeV comme alternative à l'activation neutronique; cette activation protonique connaîtra certainement un très grand développement dans les années à venir.

Il faut noter particulièrement le très grand succès des discussions conduites par des rapporteurs sur les applications de l'analyse par activation aux Sciences de la Terre et à l'étude de l'environnement. Nous devons ces succès aux Professeurs Morrison et Winchester que je tiens à remercier pour le travail très important qu'ils ont réalisé. Ce succès s'est concrétisé par la réunion en session parallèle spéciale d'une table ronde sur les échantillons lunaires.

Au cours d'une session, le point a été fait de l'évolution des techniques analytiques utilisées pour les échantillons de semi-conducteurs. Il ne semble pas que de grands progrès aient été réalisés dans l'analyse des éléments légers. Les résultats déjà acquis il y a quelques années ont été confirmés et la précision des dosages a été améliorée. De nombreuses études des phénomènes d'interférences nucléaires possibles ont été faites pour délimiter aussi exactement que possible les risques d'erreurs dans l'analyse de certains matériaux (en particulier le silicium). Par contre,

en activation neutronique, nous remarquerons que des résultats considérables ont été obtenus, en particulier par le laboratoire du Professeur Hoste, et il semble bien que l'activation neutronique et la spectrométrie γ sur cristaux de Ge-Li après de judicieuses séparations chimiques doivent s'imposer dans un proche avenir pour ces analyses. Il s'agira dans tous les cas d'étalonner les méthodes de spectrométrie de masse ou infrarouge et, parmi les plus faibles teneurs en certains éléments, d'assurer les dosages qu'aucune autre méthode ne peut réaliser avec certitude.

Les sessions sur l'analyse par activation en biologie ont remporté un très grand succès je crois, et nous devons en remercier l'organisateur, M. Comar. Dans ce domaine en particulier, les mêmes équipes utilisent l'analyse par activation et les traceurs radioactifs pour des études de métabolisme. Il faut noter une très nette tendance au développement de l'utilisation de cyclotrons compacts, aussi bien pour l'activation par toutes les particules que pour la production d'isotopes de très grand intérêt pour des études en traceurs.

DISCUSSION

R.L. JOLY: Since in your conclusions you go somewhat beyond the bounds of activation analysis proper and mention analysis by observation of fluorescent radiations, might I ask your opinion on another method of physical analysis, namely, the observation of neutron resonances by transmission of resonance neutrons by the sample under study?

Ph. ALBERT: I have no experience of work along these lines but, on the basis of general principles, I would say that this method is likely to be further developed only if it can provide solutions to analytical problems which cannot be suitably resolved by other methods. Perhaps Mr. Engelmann has some comment.

Ch. ENGELMANN: No paper on analytical applications of this procedure was presented at the Saclay International Conference on Modern Trends in Activation Analysis (2-6 October 1972). It seems to me that this is a technique which is certainly elegant in principle but for which the equipment requirements are very extensive and difficult to reconcile with the expected yields. Moreover, it is by no means certain that the method would be suitable for trace analysis.

A.T.G. FERGUSON: Analysis by observations of resonances of neutrons in the eV-keV energy range is in fact a method with a great potential for analytical sensitivity. It has been done at Harwell by Moxon for the determination of impurities in uranium samples used in nuclear physics experiments. The samples in question were comparatively large. In more general use, costs would be competitive if the method were carried out on a routine basis. The accelerators used for time-of-flight experiments with neutrons in this energy range can generally run many experiments simultaneously, with resulting reduction of costs.

Although some effort has been made to find applications for this technique outside the nuclear field, in many cases it has to compete with other methods.

A.H.W. ATEN: In connection with Professor Albert's remarks in his oral presentation concerning the use of californium neutron sources for

activation analysis, I should like to point out that this method is very powerful where it is a matter of determining the main components of a solution, the induced activity of which components represents the principal contribution to the activity observed. The precision attainable is comparable with that reached in the calibration of neutron sources in a manganese bath. At the Institut voor Kernphysich Onderzoek, Amsterdam, this method has been studied by Dr. Louwrier, who used alpha-beryllium neutron sources; however the method will be easier to apply and more powerful when californium sources can be used.

Ph. ALBERT: I fully agree with your views on the prospects for the future use of californium sources. We hope to compare their possibilities with those of other neutron sources by trying to delimit the areas of their specific or preferred application.

N.M. SPYROU: Further to Dr. Aten's comment, I believe that Pierce at Harwell is carrying out such activation measurements using californium-252 as his source.

I should also like to ask a general question of Professor Albert: do you foresee the use of activation techniques in large-scale, routine analytical programs, for example in regional hospital centres?

Ph. ALBERT: Only a dialogue between physicians and specialists in activation analysis and their methods can provide guidance concerning experiments to be carried out in an effort to develop routine analytical programs based on activation analysis methods. A second and even more important prerequisite will be the possibility of establishing radiation facilities on the spot or near to the user (hospital). Californium-252 sources and compact cylotrons represent new and decisive developments in this connection. Two novel experiments have now been made possible in France as a result of the establishment of activation analysis admission and service laboratories. These are the P. Sue Laboratory at Saclay (neutron activation in reactors) and the Orleans Cyclotron Laboratory, which is located close to a regional hospital centre. It should also be mentioned that the long-established Frédéric-Joliot Hospital Service at Orsay is now equipped with a compact cyclotron.

N.M. SPYROU: If concentration on short-lived isotopes is necessary to bring about greater use, should we not pay more attention to the technique of cyclic activation (i.e. pulsing the source or pulsing the sample)?

Ph. ALBERT: This type of analysis probably has more chance of development in conjunction with compact cyclotrons.

B. KUCHOWICZ: It would seem important to mention here the relation between activation analysis and mass spectroscopy. In general, we are accustomed to dealing with terrestrial samples of a constant isotopic composition in the case of multi-isotope chemical elements. Now, when we are embarking upon the analysis of extra-terrestrial samples, we should take into account the variability of isotopic composition (known to be a small effect where terrestrial samples are concerned). It might be desirable to combine, say, mass spectroscopy results with activation analysis, in order to avoid false abundance determinations. We cannot overlook the fact that isotopic abundances are certainly not universal constants.

Ph. ALBERT: I fully agree with you. It should be pointed out that activation analysis is, like mass spectrometry, also an isotopic analysis.

NUCLEAR DATA FOR ACTIVATION ANALYSIS Requirements and present state of compilations

V. KRIVÁŇ
Max-Planck-Institut für Metallforschung,
Laboratorium für Reinststoffe,
Schwäbisch-Gmünd,
Federal Republic of Germany

Abstract

NUCLEAR DATA FOR ACTIVATION ANALYSIS: REQUIREMENTS AND PRESENT STATE OF COMPILATIONS.

A survey of the need of nuclear data in solving individual activation-analytical problems is given. Criteria such as sensitivity, accuracy, feasibility of non-destructive analysis and simultaneous determination of several elements permitting the choice of an optimum activation technique and experimental parameters are discussed and related to the relevant nuclear data. The importance of nuclear data in the practical application of activation analysis for both the absolute and the comparative method as well as for the identification of radionuclides and in analytical γ-ray spectrometry is also discussed. – A critical picture is presented of the existing compilations and handbooks with regard to completeness, accuracy and presentation of the nuclear data contained therein. – The activation with thermal, epithermal and fast neutrons, photons and charged particles is considered in detail. In addition, prompt-radiation activation analysis and analysis based on the elastic scattering of heavy charged particles are included.

1. INTRODUCTION

The object of activation-analytical techniques in trace analysis – where the centre of gravity of its application lies – is to determine trace concentrations of elements in a sample (matrix), i.e. concentrations in the ppm- and the ppb-range and below. The absolute amounts of the elements to be determined here are in the μg, ng, and upper pg-range. Not only the minute absolute amounts of the elements to be determined but also the million and billionfold excesses of matrix elements become the central problems of activation analysis.

Activation analysis assumes one of the most important positions among the different existing determination methods. It is, however, not a universal method, and therefore in each special case only a critical comparison of all possible methods enables an optimum choice to be made. But activation analysis itself represents a wide variety of methods. Nowadays activation analysis can be defined as "the revelation of chemical composition through the modification of nuclear behaviour" [1]. Then this description includes, in addition to the conventional activation analysis with thermal, resonance and fast neutrons, with photons and with charged particles, also such rather innovatory methods as prompt-radiation activation analysis and analysis based on the elastic scattering of heavy charged particles. Table I gives a survey of activation analytical techniques.

To give an impression of the increasing spread of activation analysis, the growth-rate of the literature of activation analysis is shown in Fig. 1.

TABLE I. SURVEY OF ACTIVATION ANALYTICAL TECHNIQUES

PROJECTILE	MAIN SOURCE	MAIN NUCLEAR REACTIONS
	CONVENTIONAL ACTIVATION ANALYSIS	
Thermal and Resonance Neutrons	Reactor	(n,γ)
Fast Neutrons	14-MeV Neutron Generators Cyclotrons Reactors	$(n,2n)$ (n,p) (n,α)
Bremsstrahlung	Betatrons	(γ,n) (γ,p)
Charged Particles p, d, ^{3}He, α	Cyclotrons	Various
	PROMPT-RADIATION ACTIVATION ANALYSIS	
Thermal Neutrons	Reactor	(n,γ)
Charged Particles p,α	Cyclotrons Linear Accelerators	(p,α) (α,p)
	ANALYSIS BY ELASTIC SCATTERING	
Charged Particles p,α	Linear Accelerators Van de Graaff Cyclotrons	(p,p) (α,α)

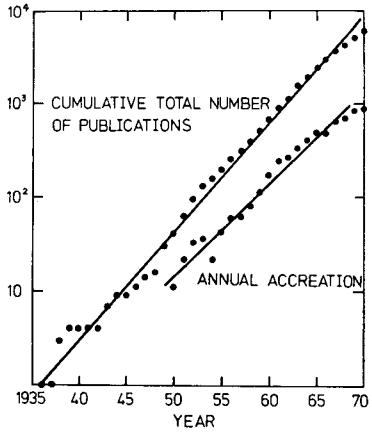

FIG.1. Growth rate of literature of activation analysis [From LUTZ, G.J., BORENI, R.J., MADDOCK, R.S., MEINKE, W.W., Activation Analysis: A Bibliography, NBS Technical Note 467, August 1972].

According to the latest estimates, the total number of publications had already reached 10 000 at the end of 1972. In addition, it should be pointed out that the number of research and application projects in which activation analysis has been used exceeds the number of publications many times.

The ability to solve the various specific problems in activation-analytical practice depends on a good knowledge of the corresponding nuclear data. These are needed, first of all, for answering the fundamental question of whether at least one of the various activation-analytical methods offers itself as a suitable method for solving the given analytical problem. Furthermore the data are necessary for optimization of the experimental conditions for the selected method so as to obtain high sensitivity and accuracy, or to simplify the problem of activity measurements and to avoid chemical separations. More accurate nuclear data are required for the utilization of absolute activation analysis. Last but not least, in all cases the corresponding nuclear data are required for the identification of the radionuclides and for analytical γ-ray spectroscopy.

Several compilations and handbooks have already been published which can be used to obtain most of the required data. A critical judgement of the existing special nuclear data sources for activation analysis shows the need for improving them mainly with respect to completeness, accuracy and presentation. An up-to-date compilation of the most important nuclear data for all kinds of activation analysis, which would enable a rapid orientation and choice of the optimum activation method, and, in the case of a given method, also optimization of experimental conditions in any particular situation, is not yet available.

The present paper gives a survey of the nuclear data needed in the various steps of the individual types of activation analysis and of the present state of nuclear-data compilations for these purposes.

2. NUCLEAR DATA REQUIREMENTS FOR ACTIVATION ANALYSIS

2.1. Nuclear data in preliminary considerations and choice of the optimum activation method

2.1.1. General considerations

When solving a problem by means of activation analysis, it is indeed possible to proceed by simply testing experimentally what can be achieved in the analytical characterization of the given material by an activation. In most cases, however, it is useful, or even absolutely necessary, to consider the problem carefully in advance and to make precise demands. An accurate strategy of determination is also frequently required because of the minute amounts of sample available. Thus, e.g. in high-purity materials, it is usually the case that with an increasingly pure matrix the sample material becomes more precious, and thus for the analysis of very low concentrations only very little sample material is, in general, available. For this reason, very often a non-destructive analysis is required.

In such cases, the corresponding analyses have to be carried out by a rather pinpointed approach. The questions to be answered are then the following ones: Is activation analysis an optimum or, at least, a suitable method for solving the given analytical problem? Which method of activation analysis is most suitable in the given case? Which are the optimum experimental conditions? To answer these questions, in many cases valuable hints can be obtained from the literature concerning already existing results of experimental activation analysis, but, generally, a good knowledge of the corresponding nuclear and other physical data for the matrix and for the trace elements of interest is required.

In the following, the most important criteria are discussed, according to which the applicability of individual methods can be judged and a choice of the optimum method can be made. The relationship between these criteria and the relevant nuclear data is indicated. It is to be noted that only those criteria will be considered here which are directly related to nuclear data, as opposed to e.g. the duration and cost of the analysis, etc.

2.1.2. Sensitivity

When considering the sensitivity, it is of primary importance to evaluate parameters affecting the minimum detectable amount of the elements in question. For estimating the limits of detection the nuclear properties of the initial and final nuclides of the elements to be determined as well as those of the matrix elements have to be taken into account. The starting point of the corresponding calculations is the radioactivation equation. All efforts are directed towards generating a maximum activity of the indicator radionuclide from a given trace amount of an element by an optimum choice of the individual parameters of the radioactivation equation.

In a simple case, i.e. if the energy and the angular distribution of the projectiles and the energy dependency of the reaction cross-sections do not have to be considered, the minimum detectable amount of an element can be calculated according to

$$LD = \frac{IMe^{0.693t'/T}}{\sigma \Phi N_0 f_a \tau \epsilon Y S (1 - e^{-0.693t/T})} \qquad (1)$$

where

I is the minimum detectable counting rate (cps),
M the mass number of the activated isotope,
σ the reaction cross-section at the given projectile energy,
Φ projectile flux,
N_0 Avogadro's number,
f_a isotopic abundance in the natural element,
τ detection efficiency,
ϵ branching ratio of the decay,
Y yield in casual radiochemical separation,
S factor for self-absorption losses,
t the irradiation time,
t' time between the end of the irradiation and the middle of the counting period,
T half-life of the indicator radionuclide.

Equation (1) can only be applied to estimate limits of detection when discrete values can be assigned to the projectile flux Φ and to the reaction cross-section σ, as, e.g. in activation with thermal and 14-MeV neutrons.

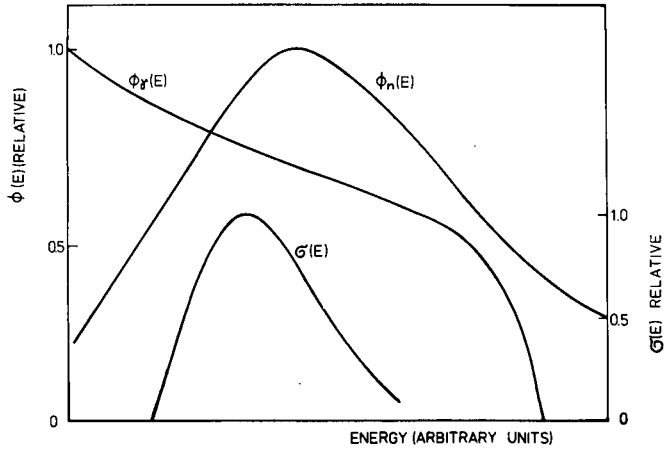

FIG.2. Typical shapes of energy spectra of neutrons [$\phi_n(E)$] produced by bombarding thick Be-targets with deuterons, of energy spectra of bremsstrahlung radiation [$\phi_\gamma(E)$] by bombarding a target with high-energy electrons, and of an excitation function [$\sigma(E)$].

These assumptions are not valid in some other types of activation analysis. Neutrons produced by deuteron bombardment of thick Be-targets and bremsstrahlung radiation from high-energy electrons striking a target have a characteristic energy and angular distribution dependent on the energy of the incident radiation. Figure 2 shows the shape of energy spectra of neutrons and photons produced in the ways mentioned. Thus, the reaction rate per target nucleus P, i.e. the total product of cross-section and projectile flux for a nuclear reaction induced by such radiations must be calculated by integrating the product of these two quantities in an energy

increment over the energy range of interest, by considering the angular distribution

$$P = \int_0^{E_m} \int_0^{\psi_1} \Phi(E,\psi)\sigma(E)\ 2\pi\sin\psi\ dE\ d\psi \qquad (2)$$

where E_m is the maximum projectile energy, ψ_1 the angle given by the size of the sample and its distance from the projectile source, $\Phi(E,\psi)$ is the projectile flux for the given energy E and angle ψ (relative to the deuteron and electron beam directions, respectively), and $\sigma(E)$ the cross-section at the projectile energy E. This integral can then be used instead of the product $\Phi \cdot \sigma$ in Eq. (1).

To estimate the sensitivity in charged-particle activation analysis, when thick samples are used, the average reaction cross-section is required, which can be defined [2,3] as

$$\bar{\sigma} = \frac{\int_0^R \sigma(x)\,dx}{\int_0^R dx} = \frac{\int_0^{E_0} \sigma(E)\left[-\frac{dx}{dE}\right] dE}{\int_0^{E_0}\left[-\frac{dx}{dE}\right] dE} \qquad (3)$$

where $\sigma(x)$ is the value of the cross-section at depth x, R is the total range of the projectile particles in the target, $\sigma(E)$ is the cross-section at particle energy E, $(-dx/dE)$ is the reciprocal of the stopping power, E is the incident projectile energy. An approximation $(-dx/dE) \sim E$ leads to a very simple expression for $\bar{\sigma}$.

The average cross-section can be also used in activations with fast reactor neutrons by (n, threshold) reactions. It is given by

$$\bar{\sigma} = \frac{\int_0^{\infty} \Phi(E)\ \sigma(E)\ dE}{\int_0^{\infty} \Phi(E)\ dE} \qquad (4)$$

where $\Phi(E)$ is the fission neutron flux of energy E and $\sigma(E)$ is the cross-section for neutrons of energy E.

The average cross-section obtained in this way can then be used in Eq. (1) to calculate the limits of detection. In the case of charged-particle activation, the range of the projectiles in the sample must also be taken into account.

Thus, the fundamental nuclear data necessary for such calculations are: the cross-sections, the average cross-sections or the excitation functions, respectively, the stopping powers and the total ranges in the case of charged-particle activation analysis, the energy and angular dis-

tribution of projectiles (if relevant), energies and absolute intensities of the radiation emitted in the decay of the indicator nuclide (affecting the overall counting efficiency), and the half-life of the indicator nuclide influencing the saturation factor and the decay losses.

2.1.3. Accuracy

A number of different systematic errors can cause the results obtained by activation analysis to differ from the real values by as much as several orders of magnitude. For this reason the possible systematic errors should be carefully investigated in each case. The extent and nature of the systematic errors in activation analysis depend upon the constituents of the sample, the nature and energy of the bombarding radiation, and the type of counting system used. In general, the factors described in the following sections may affect the accuracy.

2.1.3.1. Reaction interferences

Interfering nuclear reactions are defined as reactions which modify the linear relationship between the amount of the element to be determined and the amount of the indicator radionuclide produced. Interfering nuclear reactions can increase or decrease the amount of the indicator radionuclide with respect to the element sought.

(a) Primary Interference Reactions. These are nuclear reactions induced by the primary bombarding radiation on elements other than the element to be determined. They yield the same indicator radionuclide as does the desired reaction. Reactions inducing a radionuclide of a neighbouring element which decays with a short half-life to the indicator radionuclide also belong to the primary interference reactions.

Examples of primary interference reactions are given in Table II. Figure 3 illustrates how the proportion of interfering reactions depends on experimental conditions. Interferences can often be avoided by the choice of the projectile energy. In other cases, corrections must be applied. For this purpose, the following nuclear data must be known: thresholds, cross-sections and/or excitation functions for both the desired and the interfering nuclear reactions, and possibly decay constants.

(b) Secondary Interference Reactions. These nuclear reactions, significant in non-reactor irradiations, take place between secondary particles and elements other than the ones to be determined. Such reactions may produce the indicator nuclide and thus may introduce errors. The secondary particles may be formed by nuclear reactions induced by a primary radiation in the sample or its immediate environment. For example, when bombarding an organic matrix with fast neutrons, considerable fluxes of knock on protons can be produced by collision with hydrogen atoms. Relatively high neutron fluxes can be formed by secondary reactions in activation analysis with photons via (γ, n)-reactions and with deuterons via (d, n)-reactions, stripping and breakup processes. Examples for such secondary interference reactions are given in Table III. For example, the secondary interference reactions in the determination of nitrogen via $^{14}N(n, 2n)^{13}N$ reaction can induce an activity corresponding to some hundred ppm of nitrogen [4, 5].

TABLE II. EXAMPLES OF PRIMARY INTERFERING REACTIONS

Projectiles	Reaction used for determination	Threshold	Principal interfering reactions	Threshold
Fission neutrons	$^{23}Na(n,\gamma)^{24}Na$	0.	$^{24}Mg(n,p)^{24}Na$	4.9
			$^{27}Al(n,\alpha)^{24}Na$	3.2
Fast neutrons	$^{28}Si(n,p)^{28}Al$	4.0	$^{31}P(n,\alpha)^{28}Al$	2.0
Photons	$^{12}C(\gamma,n)^{11}C$	18.7	$^{14}N(\gamma,t)^{11}C$	22.7
			$^{16}O(\gamma,\alpha n)^{11}C$	25.9
^3He-particles	$^{16}O(^3He,p)^{18}F$	0.	$^{19}F(^3He,\alpha)^{18}F$	0.
	+		$^{23}Na(^3He,2\alpha)^{18}F$	0.4
	$^{16}O(^3He,n)^{18}Ne$	0.	$^{27}Al(^3He,3\alpha p)^{18}F$	13.5
	$\xrightarrow{\beta^+}$ ^{18}F		$^{27}Al(^3He,3\alpha)^{18}F$	11.5
α-particles	$^{19}F(\alpha,\alpha n)^{18}F$	12.6	$^{16}O(\alpha,2n)^{18}Ne$	29.7
			$\xrightarrow{\beta^+}$ ^{18}F	

To answer the question whether secondary interfering reactions can occur or not and under what conditions they can be avoided or what corrections must be applied, corresponding nuclear data for both the reaction inducing secondary radiation and the secondary interfering reaction itself, have to be known. The nuclear data include: cross-sections and/or excitation functions, and the energy and angular distribution of the secondary particles.

(c) Second-Order Reactions. These are nuclear reactions taking place with the transformation products of the sample constituents. Two types of second-order reactions may occur:

(c1) the amount of the indicator nuclide can be markedly reduced by its activation induced by the original bombarding radiation, if the cross-section is large, i.e. between 10^3 to 10^4 barns. Therefore, these interferences are of importance in thermal-neutron activation analysis using long irradiation times. For example, in the determination of tantalum via the $^{181}Ta(n,\gamma)^{182}Ta$

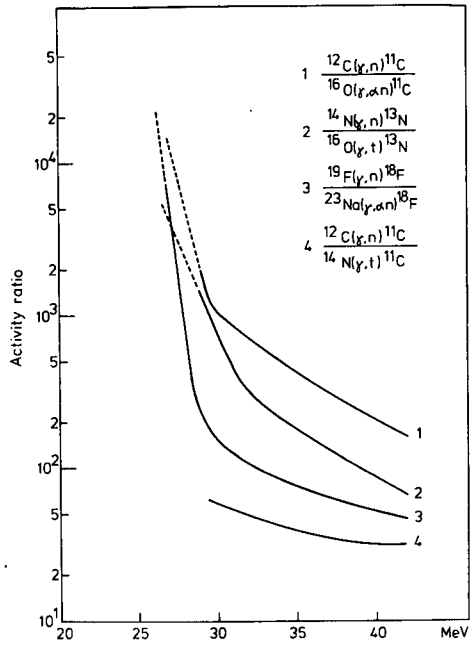

FIG.3. Energy dependence of the contribution of the interfering reactions to the production of the indicator nuclide in determination of carbon (curves 1 and 4), nitrogen (curve 2). and fluorine (curve 3) by photon activation analysis [5].

TABLE III. EXAMPLES OF SECONDARY INTERFERING REACTIONS

Primary radiation	Reaction used for determination	Reaction inducing secondary radiation	Secondary interfering reactions
Fast neutrons	$^{14}N(n,2n)^{13}N$	p-knock on	$^{13}C(p,n)^{13}N$ $^{16}O(p,\alpha)^{13}N$
Photons	$^{25}Mg(\gamma,p)^{24}Na$	(γ,n)	$^{23}Na(n,\gamma)^{24}Na$ $^{27}Al(n,\alpha)^{24}Na$
Deuterons	$^{12}C(d,n)^{13}N$	(d,n) + stripping and break-up	$^{14}N(n,2n)^{13}N$

reaction (σ = 19 b, T = 111 d) one must take into account that the $^{182}Ta(n,\gamma)^{183}Ta$ reaction (σ = 17 000 b, T = 5.5 d) occurs. To judge the importance of this interference, the following nuclear constants have to be known: the cross-sections of both the desired and the second-order reactions and the half-lives of the indicator nuclide and the nuclide produced by the second-order reaction.

(c2) The amount of the nuclide to be activated (and, consequently, also the amount of the indicator nuclide) can be enhanced by the decay of a short-lived radionuclide produced from a neighbouring element. This kind of second-order interfering reaction can be demonstrated by the example of the determination of phosphorus in silicon [6]. When phosphorus is determined via the $^{31}P(n,\gamma)^{32}P$ reaction, the matrix element is also activated forming ^{31}Si which decays to ^{31}P and thereby increases the number of the target nuclei:

$$^{30}Si\,(n,\gamma)^{31}Si \xrightarrow[2.6h]{\beta^-} {}^{31}P(n,\gamma)^{32}P$$

The nuclear data required are again the cross-sections for both nuclear reactions and the decay constants.

2.1.3.2. Self-shielding

In activations with any type of particle, self-shielding effects occur which result in an attenuation of the original radiation flux within the sample itself. This effect is of particular importance in thermal and resonance neutron activation in cases where the total absorption cross-sections are very large for the matrix elements. Systematic errors caused by this effect can occur also in the activation analysis with fast neutrons and photons.

In any particular case, it is necessary to estimate the significance of the self-shielding effect in order to be able to judge whether the flux attenuation is serious or can be neglected.

In thermal-neutron activation it is in general sufficient to utilize the simple attenuation formula

$$\frac{\Phi}{\Phi_0} = e^{-N\sigma x} \qquad (5)$$

where Φ_0 is the incident flux, Φ is the transmitted flux at depth x (in cm) inside the sample, N is the number of atoms of a given element per cm^3 of sample and σ is the neutron absorption cross-section. Several other calculation procedures have also been suggested [7-11]. In some of them the resonance neutron self-shielding is also treated.

In activation analysis with 14 MeV neutrons, an additional important effect is neutron scattering. Therefore, the total removal cross-section instead of the total cross-section has been introduced [12] as a more accurate measure of the attenuation.

Attenuation of bremsstrahlung is usually an unimportant source of error in photon activation analysis, unless the samples are large, or very ac-

curate results are required. For estimating the attenuation, a simple expression has been suggested [13,14], for which the bremsstrahlung absorption coefficients at the energy of the peak cross-section and the densities for each element in the sample must be known.

Unfortunately, the procedures for estimating the significance of the attenuation by self-shielding generally can not be used for obtaining an actual correction factor. One of the most serious reasons is the low accuracy with which many absorption cross-sections have been determined. This would increase the uncertainties in the correction factor considerably. Very often, the sample composition is not sufficiently well known prior to the analysis to permit an accurate statement concerning the elements present. Therefore, if preliminary estimates indicate that a significant flux attenuation will exist, either steps should be taken to avoid errors due to self-shielding (e.g. use of small samples, suitable dilution, preparing the standard in a matrix of the same cross-section and the same shape and volume) or a correction should be applied on the basis of the experimental measurement of the self-shielding effect (e.g. by using a series of samples of different weight and by extrapolating the specific activity curve to zero sample weight).

It can easily be recognized that the self-shielding effects are most severe in activation analysis with charged particles. In this case, however, the self-shielding problems are different from those in neutron and photon activation. Whereas self-shielding problems are caused by flux attenuation in neutron and photon activation, in charged-particle activation they arise because of both the rapid energy decrease of the incident particles penetrating into the matrix and the variation of the activation cross-section with projectile energy and, therefore, with the depth of their penetration. For this reason, in charged-particle activation the self-shielding must be considered in any case. Some practical aspects of this problem are discussed in section 3.2.

2.1.4. Non-destructive activation analysis

Activation analysis offers the special advantage that determination can be carried out non-destructively. Chemical separations are generally time-consuming and an additional source of error. When solving a given problem using activation analysis, it should therefore first be investigated whether it is possible to work non-destructively. In principle, this is possible only when the activity from the matrix elements is low enough during the measurement. If higher activity is present, the main limitation is caused by the Compton continuum in the pulse-height spectra in cases where the energies of interfering gamma rays are higher than the gamma-ray energies of the indicator radionuclides. Then the peaks of interest are superimposed on the Compton level corresponding to the gamma rays of the nuclides produced from the matrix elements. The limits of detection for non-destructive activation analysis will in general be higher than those for radiochemical activation analysis, or a non-destructive determination is impossible, at all. The possibility of overlapping of two peaks must in some cases also be taken into account.

The basic requirements for non-destructive activation analysis are met when 1) the activation cross-sections of the reactions with the matrix nuclides are substantially smaller than those of the desired reactions, or

2) when the half-lives of the radionuclides resulting from the matrix are very long or very short compared with the half-lives of the radionuclides of interest: in the first case, only little activity results from the matrix, in the second case, the activity resulting from the matrix elements can be allowed to decay. For each case there is an optimum ratio between irradiation time and the time of waiting before the measurement is carried out.

In many cases non-destructive analysis is only possible under the optimum irradiation conditions. Thus, e.g. activation with resonance neutrons is more favourable than that with thermal neutrons or with a fission spectrum in all those cases when

$$\frac{J_m}{\sigma_x} << \frac{J_x}{\sigma_x} \tag{6}$$

where J_m and J_x are the resonance integrals and σ_m and σ_x are the thermal neutron cross-sections for the matrix element or the element to be determined.

Activation analysis with cyclotron-produced fast neutrons (bombarding a thick beryllium target with deuterons) is a very powerful method [15]. A great advantage of this method is the possibility of varying the position of the maximum of the neutron spectrum by varying the deuteron energy. The deuteron energy can be selected in such a way that the maximum of the neutron spectrum is located at the same energy as the maximum of the excitation function for the desired reaction. In this way, an optimum deuteron energy may be selected which ensures maximum sensitivity for the desired reaction with a minimum interference yield. Figure 4 demonstrates this by the example of the determination of aluminium traces in niobium.

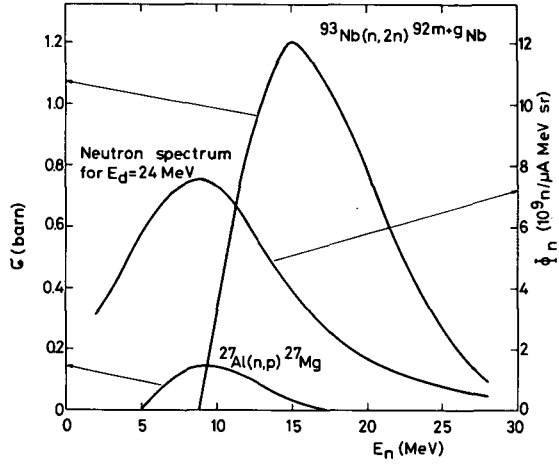

FIG. 4. Choice of optimum neutron spectrum in the determination of aluminium in niobium by fast-neutron activation analysis using a cyclotron [15].

Similarly, the optimum bombarding energy can be chosen also in activation analysis with charged particles and bremsstrahlung.

Knowledge of the following nuclear data is essential for preliminary calculations giving an answer to the question of whether a non-destructive determination is possible or not as well as for its optimization and for calculating the limits of detection (according to the kind of activation analysis): activation cross-sections, resonance integrals, excitation functions and average cross-sections, decay schemes, half-lives.

2.1.5. Ability to determine several elements simultaneously

In many cases it is very desirable that several or – if possible – all elements of interest should be determined simultaneously. This requirement may become one of the most important criteria and is predominantly of economical character either with respect to time and price of analysis or with respect to costliness of the sample. Preliminary calculations of sensitivity and accuracy must be made for each element to be determined in order to answer the question if a simultaneous determination is possible and if so under what conditions.

2.2. Carrying out the activation analysis

The importance attached to the knowledge of the nuclear data for the performance of activation analysis chiefly depends on whether the absolute or the comparative method is used.

2.2.1. Absolute activation analysis

This method is based on activating the samples, measuring the activities with a calibrated gamma-ray spectrometer, and calculating the unknown amount of the elements to be determined by using Eq. (1). Absolute activation analysis would have some essential advantages over the more generally used comparative method. The greatest advantage is to be seen in the fact that a considerable portion of experimental work – namely, the frequently very time-consuming preparation of standards, their irradiation and measurement as well as the evaluation of the errors due to standard preparation and inhomogeneity of the projectile flux between standard and sample – could be avoided. Thus, the absolute assay technique would certainly be the essentially simpler alternative in many cases. Unfortunately, this direct method can only very rarely be applied in practice and then mostly only as semiquantitative method. In the absolute method many factors must be considered which contribute to the overall accuracy and precision.

To give an impression of what kind of errors may occur in absolute activation analysis and what signficance the errors introduced by nuclear data may have thereby, generalized data of error analysis obtained by Cali et al. [16] for thermal neutron activation analysis are shown in Table IV. A similar error distribution has been obtained by Girardi et al. [17].

Table IV shows clearly that uncertainties in nuclear data constitute the main obstacle to the applicability of the absolute method. The errors introduced by nuclear data may exceed the errors caused by other sources by a factor of up to one order of magnitude, and so they are decisive for

TABLE IV. SURVEY OF SOURCES OF ERRORS AND THEIR EXTENT IN ABSOLUTE NEUTRON ACTIVATION ANALYSIS [2]

Source	Estimated Error (%)
1. Chemical manipulations	
a. Sample weight	± 1
b. Yield determination	± 2
2. Irradiation	
a. Self shielding (correction < 50 %)	± 4
b. Flux depression	± 2
c. Thermal enhancement	± 2
d. Absolute value of thermal flux	± 5
e. Value of cadmium ratio	± 2
f. Irradiation time (< 1 min)	± 3
g. Inhomogeneity of neutron flux	± 1
3. Counting	
a. Detector calibration	± 3
b. Counting rate (< 10^3 cps)	± 4
c. Geometrical factors	± 1
4. Interfering and competing nuclear reactions	variable
5. Nuclear data	
a. Half-life	± 2-10
b. Decay schemes	± 2-50
c. Thermal activation cross sections	± 5-30

the accuracy of the absolute activation analysis. The random erros in this method generally do not exceed those obtained by the comparative method As will be shown in the discussion of the accuracy of the existing nuclear data, the size of errors in other kinds of activation as compared with (n, γ) activation by thermal neutrons are in general even larger since the thermal neutron cross-sections for (n, γ) reactions are, on the average, more precisely known than the cross-sections or excitation functions for other nuclear reactions. In charged-particle activation analysis, in addition to the exact knowledge of excitation functions, also the stopping powers and the ranges are required for the absolute method.

Of course, the successful application of the absolute method does not depend only on a knowledge of nuclear data, though this is generally the most critical point. It can be stated with certainty that absolute activation analysis could be used to an essentially greater extent, if more accurate nuclear data, especially cross-sections and/or excitation functions, and decay data, were available.

2.2.2. Comparative activation analysis

This method utilizes comparative standards containing known amounts of the trace elements to be determined. These standards are processed (irradiated and measured) in the same way as the samples. The method requires relative measurements only so that accurate values of reaction cross-sections (excitation functions) and the branching ratio of the decay are not necessary. These nuclear data must only be known for preliminary considerations and for the choice of optimum irradiation conditions as well as for the post-irradiation assay.

The only data often needed with high accuracy are the half-lives used in corrections for post-irradiation decay.

When there are no great differences in the composition and amount of the sample and the standard, the attenuation of the projectile flux is an unimportant source of error in neutron and photon activation. This is rarely the case in charged-particle activation, because the monoenergetic incident particles are rapidly slowed down and stopped in solid matrices. Since the cross-section is strongly dependent on the projectile energy and the stopping powers on the target composition, activation analysis with charged particles is generally more involved than that with neutrons and photons.

Two methods have been developed for the mathematical treatment of charged-particle activation analysis: the method of the equivalent thickness [18] and the simpler method of the average cross-section [2, 3]. In the first method, excitation functions, stopping powers, and bombarding energy do not enter into the calculations, but an activation curve must be measured for each nuclear reaction and each sample using very thin foils. With the second method, the unknown concentration C of the element to be determined can be obtained from

$$C = C_s \frac{AI_s R_s}{A_s I R} \tag{7}$$

where C_s is the concentration of the sought element in the standard, A and A_s are the activities, I and I_s are the beam intensities, and R and R_s are the particle ranges for sample and standard, respectively. The validity of this equation is based on the assumption that the properties of the sample and standard do not differ essentially with respect to the average cross-section. Therefore, the sample and the standard should have similar densities and atomic numbers, if really accurate results are to be obtained. From Eq. (7) it is evident that the determination or calculation of the average cross-section is not necessary in the comparative method of charged-particle activation analysis. What must be known are the particle ranges in sample and standard, whereby the accuracy of these data can play an important role.

2.2.3. Evaluation and interpretation of experimental results

Today, the basic measuring technique of activation analysis is undoubtedly gamma-spectrometry using semiconductor detectors or NaI(Tl)

crystals. Therefore, the final part of activation analysis usually consists of the evaluation and the interpretation of the gamma-spectra. These provide: 1) the qualitative identification of the radionuclides by means of the energy lines contained in the gamma spectrum, and 2) quantitative evaluation of the spectra from peak intensities corrected for background, the Compton background, and, possibly, the interfering reactions.

In most cases the qualitative identification of the radionuclides is accomplished only by means of γ-energies and their intensities. Sometimes, however, the half-lives of the spectrometrically separated γ-energies must also be evaluated. For the purpose of analytical γ-ray spectroscopy, special tables, which are simple to handle and which enable rapid, accurate, and precise evaluation and interpretation of experimental results, are required. This can be achieved by arranging the nuclides according to the proper parameters, thus, e.g. according to γ-energies, half-lives, and atomic numbers. The completeness of the gamma-energies for each nuclide can be of great significance. In addition to these, still some supplementary data should be properly included in such a table to facilitate the identification of unknown γ-spectra, e.g. production reactions, saturation activities, etc. Naturally, such a compilation of energies should fully meet the demands made on the accuracy of measurements using germanium-lithium detectors.

The method based on the determination of the half-life may be applied to the identification of radionuclides with an arbitrary type of radiation emitted. This method is applicable in cases where the radionuclides of interest are sufficiently short-lived to obtain significant reduction in radioactivity during acceptable periods of counting. The identification is made by comparing the measured half-life with tabulated values. Therefore, the accuracy of the compiled half-lives can possibly be of importance.

3. PRESENT STATE IN NUCLEAR DATA COMPILATIONS

In section 2 the requirements for nuclear data and the demands on their accuracy for activation analysis have been discussed in detail. In Table V the nuclear data needed for the most important preliminary calculations and the performance of activation analysis are summarized. It is also indicated if accurate data are required or if approximate values are sufficient. Naturally, in such a presentation one can only generalize and is not able to draw an exact borderline between requirements for approximate and for accurate data.

3.1. Survey of existing compilations

The existing compilations of nuclear data for activation analysis, their usefulness, presentation, and completeness are surveyed below. In this consideration also compilations are included which indeed do not contain any direct nuclear data but parameters closely related to them, e.g. tabulation of sensitivities, reaction interferences, etc. Obviously, old compilations are incomplete as seen from today's standpoint, or they contain many inaccurate data. For this reason, compilations edited in the forties and fifties have not been considered. Furthermore, with few exeptions only compilations have been taken into account which contain

TABLE V. SURVEY OF NUCLEAR DATA NEEDED FOR INDIVIDUAL PURPOSES IN ACTIVATION ANALYSIS AND THE DEMANDS ON THEIR ACCURACY: ● ACCURATE DATA REQUIRED, O LESS ACCURATE DATA SUFFICIENT

Used for \ Data	σ_{act} (E) of desired reactions	σ_{act} (E) of interfering reactions	Abs. cross-sections	Stopping powers, total ranges	Absorption coefficients	Isotopic abundance	Half-lives	Decay data	Q-values, thresholds
Estim. of sensitivity	O			O		O	O	O	O
Estim. of reaction interferences		O		O		O	O	O	O
Estim. of self-shielding effect			O	O	O				
Correction for reaction interferences		●		●		●	●	●	
Correction for self-shielding			●	●	●				
Choice of optimum exp. conditions	O	O	O	O	O	O	O	O	O
Performance according to absolute method	●	●	●	●	●	●	●	●	●
Performance according to comperative method	O	O	O	O	O	O	●	O	O
Radionuclide identification and γ-ray spectrometry						O	●	●	O

data on more than 20 elements. Bibliographies giving only a listing of literature but no data are generally not taken into account.

Table VI contains a survey of existing handbooks and data compilations giving the first of the authors (but both if there are only two), year of publication, bibliographical reference, and the most important data contained which are needed for activation analysis with reactor neutrons, fast neutrons, photons and charged particles as well as data needed for nuclide identification and gamma-ray spectrometry. If the given data are fully contained in the compilation, this is indicated by a full circle. In other cases, i.e. if the

TABLE VI. SURVEY OF EXISTING NUCLEAR DATA COMPILATIONS USABLE FOR ACTIVATION ANALYSIS

Author(s) of the compilation, year	References	Data contained																													
		Reactor neutron activation								Fast neutron activation						Photon activation				Charged particles activation				General							
		Thermal act. cross sections	Thermal abs. cross sections	Act. resonance integrals	Abs. resonance integrals	Cross sections for (n,threshold)	Exp. or calc. sensitivities/ standard spectra		Data for interfering reactions	Half-lives, main decay data	14-MeV act. cross sections	14-MeV exp. or calc. sensitivities/standard spectra	14-MeV data for interfering reactions	Excitation functions for (n,threshold)/systematics	Q-values/thresholds	Half-lives, main decay data	Excitation functions/ systematics	Exp. or calc. sensitivities/ standard spectra	Data for interfering reactions	Q-values/thresholds	Half-lives, main decay data	Excitation functions/systematics	Exp. or calc. sensitivities/ standard spectra	Data for interfering reactions	Q-values/thresholds	Half-lives	β^--Energies and branching ratios		Additional decay data	Exp. or calc. β^--ray spectra	
Koch, 1960	19	●	●	●	●	●	●		○		●			○	●		○			●		○			●	○				●	
Yule et al., 1964	20						●			○																					
Howerton, 1964	21														●		○														
Goryachev, 1964	22																○				●										
Heath, 1964	23																														●
Yule, 1965	24						●			○																					
Girardi et al., 1965	25	●					●			●																					
Listien, Paulsen, 1963/66	26													●	●																
Goldberg et al., 1964/66	27-30	●	●	●	●						●			●																	
Drake, 1966	31			●	●																										
Jessen et al., 1966	32													●	●																
Kenna, Conrad, 1966	33										●	●			●	○															
Baumgärtner, 1967	34	●	●	●		●				○																					
Mathur, Oldham, 1967	4												●																		
Pearlstein, 1967	35										●																				

Routti, 1967	36	●				●	●		○					●														
Baker et al., 1967/68	37, 38																●											
Lederer et al., 1968	39																								●	●	●	
Seelmann-Eggebert et al., 1968	40	●	●																						●	○	○	
Cuypers, Cuypers, 1968	41										●				○													
Kochevanov, Kuznecov, 1968	42																●		●	○								
Lange, Münzel, 1968	43																				●							
Aliev et al., 1969	44	●	●	●	●	●			●	●			●		●										●	○	○	
Adams, Dams, 1969	45								●																			
Op de Beeck, 1969/70	46, 47							●																				
K.A.P. Laboratory, 1970	48	●	●																						●	○	○	
Large, Bullock, 1970	49																								●		○	
Pannetier, Chevillon, 1970	50																								●	●	○	
Meixner, 1970	51																								●	●		
Selinov, 1970	52																									○	○	
Adams, Dams, 1970	53																											●
Lutz, 1971	14																●			○								
Slunéčko, Kosta, 1971	54														○		●			●								
Dams, Adams, 1971	55						●		●																			
Padgen et al., 1971	56-58	●					●		●																			
Vogg, 1971	59						●		●																			
Meixner, 1971	60																								●	●	○	●
Wakat, 1971	61																								●	●		
Maslov, Lubnickij, 1971	62	●		●		●	●		○	●	○				○										●	○		
DeBruin, Korthoven, 1972	63								●																			
Galatanu, Grecescu, 1972	64																			●								
Kriváň, Münzel, 1972	65, 66												●															
Van der Linden et al., 1972/73	67, 68	●		●					○																			
Keller et al., 1973	69													●					●					●				
Lange et al., 1973	70																				●			●				

data are not presented in a form directly usable or if an important component is missing for completeness, this is indicated by an empty circle. It should also be noted that the compilation given in Table V does not pretend to be complete. This may be particularly true for cases where the data sources refer only to a relatively narrow field of activation analysis.

All the existing compilations cover only a smaller or greater part of the entire field of activation analysis. The most universal is the "Activation Analysis Handbook" by Koch [19], which, however, was published as long ago as 1960. Taking into account the state of development of activation analysis and the knowledge of nuclear data at that time, the author has still to be congratulated for the conception of this handbook. It contains detailed data for neutron activation, including a listing of possible interfering reactions and self-shielding effects. In addition, some valuable information for charged-particle and photon activation is given. The information for each element and various activation techniques is summarized on facing pages and so is readily accessible.

Now, however, many additional nuclear reactions are utilized for activation analysis, and many new and especially more accurate data are available.

In the field of neutron activation analysis there exist several special compilations. The most universal in its application is the "Handbook of Nuclear Data for Neutron Activation Analysis" by Aliev et al. [44], published in 1969, which can be utilized for practically all neutron sources. In addition to activation data for thermal, resonance, fission, and 14-MeV neutrons as well as excitation functions for neutron threshold reactions it contains decay data of radionuclides produced by neutron activation and self-shielding data. Unfortunately, the tables are based on data which were obtained up to 1965, so that in several instances data are either missing, or, in part, old and less accurate values are presented. Reaction interferences and corresponding data have not been treated.

The "Handbook of Neutron Activation Analysis" by Maslov and Lubnicki [62], published in 1971, is indeed the most recent handbook in the field of activation analysis; it contains, however, only activation data for thermal, resonance and 14-MeV neutrons. Unfortunately, the data are arranged according to the atomic number of the generated nuclide without direct indication of the nuclear reaction or at least its type. This makes the use of the compilation a little cumbersome. It is somewhat surprising that in such a special handbook, in the column "Intensity" it is only indicated whether the γ-intensity is "strong", "medium" or "weak" without giving any quantitative information. Alos in this handbook the possible interfering reactions with corresponding data are not given.

For activation analysis using reactor neutrons there exists a successful and very practical presentation of data in the "Table of Neutron Activation Constants" by Baumgärtner [34]. It enables its user to carry out very easily and rapidly preliminary calculations for sensitivities, primary interfering reactions, for the non-destructive activation analysis, and for the optimization of experimental conditions. Unfortunately, the data making up this table have to be replaced by newer and more accurate ones in many cases or have to be supplemented by data now available.

All cross-section data essential for neutron activation are compiled in the well-known BNL 325 series [27-30]. But also in this case a new edition would be highly desirable.

Practically no detailed handbook or data compilation exists for activation analysis with photons and charged particles.

The most extensive and still very useful compilation of cross-sections of photonuclear reactions has been published by Goryachev in 1964 [22]. For many purposes the catalogue of gamma spectra for photon activation prepared by Baker, Hunter and Wood [37, 38] can be of great practical importance. However, the applicability of empirical spectra measured under defined experimental conditions is rather limited. Some good extracts of the most important data for significant photonuclear reactions can be found in reviews and special contributions, e.g. by Lutz [14, 71] and Slunečko and Kosta [51]. In this situation the most important key to nuclear data on photon activation is still the Photonuclear Data Index [72, 73]. It is more than a bibliography since it supplies quantitative information on the content of the papers. Information is given on the type of measurement, the range of excitation energies covered, source type and energies, detector type and angular ranges.

An essential improvement in nuclear data compilations for charged-particle activation is made by the comprehensive compilation of excitation functions by Lange et al. [70] which is now becoming available. In addition to graphical presentations of excitation functions for projectiles from proton up to ^{40}Ar, it includes a tabulation of projectile energy ranges, maximum cross-sections and their energy positions, FWHM of the excitation functions, Q-values and thresholds ordered according to target and product nuclides. The stopping powers and ranges can be obtained from an extensive table prepared by Williamson et al. [74].

The situation is most favourable in tabular compilations for nuclide identification and gamma-ray spectrometry. Two recently published tabular compilations are to be mentioned especially in this connexion, namely the "Catalogue of γ-rays Emitted by Radionuclides" by Wakat [61] and "Gammaenergien" by Meixner [60]. In some cases it is very useful for the evaluation of gamma spectra if, in addition to catalogues of gamma-rays prepared according to nuclear aspects, also a catalogue of experimentally measured standard spectra (e.g. [23, 37, 38, 41, 53, 59]) is available. For example, such standard spectra can help in identifying the escape peaks in measured spectra. Normally, no information is given in tabulations of gamma-ray energies concerning the photon escape peaks.

3.2. Quality of the nuclear data

In section 2 the significance of the accuracy and precision of the existing nuclear data has already been pointed out on several occasions. When high demands are made in this respect, this frequently becomes a serious problem. In most cases there are considerable discrepancies between the values of the same nuclear constants found by different authors, and it is difficult to choose the most accurate value.

In Table VII the estimated scattering of the experimental values of nuclear data obtained by different authors is shown, i.e. the typical scatter as well as the maximum scatter of experimental data which can occur in individual cases. The scatter of experimental thermal neutron activation cross-sections, activation resonance integrals and 14-MeV neutron activation cross-sections is shown in more detail in Figs 5-7 respectively. It is defined as (range/average $\times$ 100).

TABLE VII. SCATTERING OF EXPERIMENTAL NUCLEAR DATA*

Nuclear data	Typical scattering (%)	Maximum scattering (%)
Half-lives	1 - 10	≈ 60
Decay-scheme values	2 - 15	≈ 80
Thermal neutron act. and abs. cross sections	5 - 40	> 100
Neutron act. and abs. resonance integrals	10 - 50	> 100
(n, threshold) 14 MeV neutron cross sections	30 - 80	> 100
(n, threshold)-reaction excitation functions	20 - 60	> 100
Charged-particle-induced reaction excitation functions	5 - 50	> 100

x Defined as (range/average x 100)

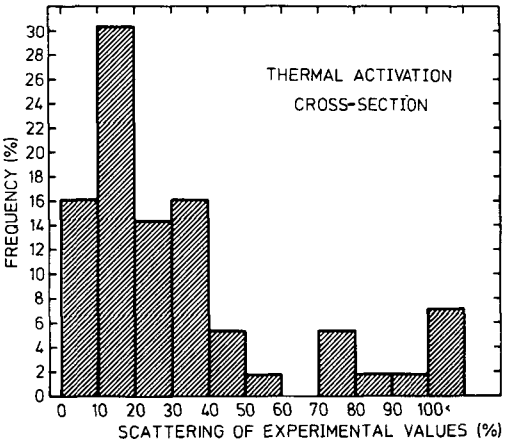

FIG. 5. Scattering of experimental thermal activation cross-sections obtained by different authors.

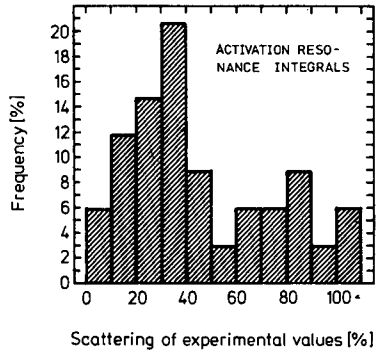

FIG. 6. Scattering of experimental activation resonance integrals obtained by different authors.

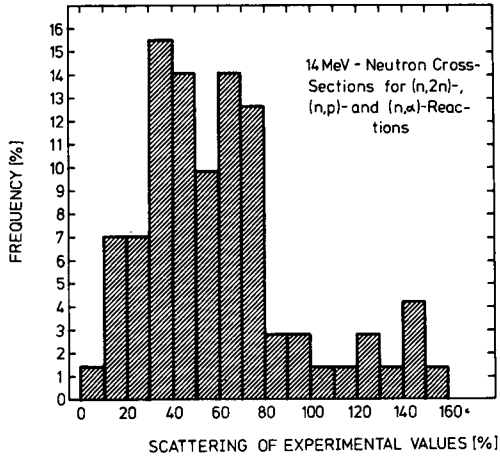

FIG. 7. Scattering of 14-MeV neutron activation cross-sections for (n, 2n)-, (n, p)- and (n, α)-reactions obtained by different authors.

The scattering of the data is in general surprisingly large. Naturally, these deviations are not to be regarded directly as the actual error. By means of a critical analysis (consideration of experimental conditions, primarily those referring to the measuring technique), hints can be obtained as to which values may be more correct and which are less correct. However, there are no entirely objective criteria for such an analysis, and thus a choice of the recommended value from all existing data will always be more or less erroneous because of subjective selections. As a rule, the more recent data, which have been obtained using modern measuring techniques, above all semiconductor detectors, are more correct than older data. In general, they show smaller scatter than the older ones. The selection of recommended values based on careful analysis of the available data can be considered one of the most important tasks for the compilators and evaluators. In this direction much has already been accomplished in the area of decay data by the Nuclear Data Group of Oak Ridge (Nuclear Data Sheets), Ajzenberg-Selove et al. [75-79] and Endt and van der Leun [80].

TABLE VIII. EXAMPLES OF SCATTERING OF NUCLEAR DATA IN DIFFERENT COMPILATIONS

Nuclear data	Koch, 1960	Gillespie, Hill, 1961	BNL 325, 1966	Kenna, Conrad, 1966	Baumgärtner, 1967	Seelmann-Eggebert et al., 1968	Aliev et al. 1970	Maslov, Lubnickij, 1971	Meixner, 1972
Half-life of ^{20}F (sec)	10.7	-	-	10.7	11.0	11.2	11.36	11.36	11.56
Half-life of ^{69}Zn (min)	52	-	-	-	59	55	58.5	58.5	55
Thermal act. cross-section of $^{202}Hg(n,\gamma)^{203}Hg$ (b)	3.8	-	4.5	-	4.0	5.0	3.8	5.04	-
Thermal act. cross-section of $^{94}Zr(n,\gamma)^{95}Zr$ (b)	0.08	-	0.075	-	0.1	0.075	0.36	0.08	-
Thermal act. resonance integral of $^{186}W(n,\gamma)^{187}W$ (b)	355	-	-	-	484	-	355	562	-
Thermal act. integral of $^{51}V(n,\gamma)^{52}V$ (b)	2.2	-	-	-	0.46	-	2.2	0.46	-
14-MeV neutr. cross-section of $^{90}Zr(n,2n)^{89m}Zr$ (mb)	79.8	80	-	770	-	-	150	170	-
14-MeV neutr. cross-section of $^{31}P(n,\alpha)^{28}Al$ (mb)	146	150	-	150	-	-	118	100	-

The difficulty of selecting the most accurate value from the many existing data is reflected in handbooks and compilations, even in those published recently. For some nuclear constants very different values may be found in different compilations. Several examples are listed in Table VIII. It is surprising that this situation also holds true for data for which recommended values are given in Nuclear Data Sheets.

The state of half-lives, if compared with other nuclear data, can be termed satisfactory: in general, the values given in different tabular compilations agree quite well. But in several cases the values for the half-lives of nuclides important in activation analysis given in different tables deviate by 5 to 10%. The situation is best for half-lives in the range of hours and days, where the data given in compilations scatter between 1 and 3%.

In most cases the errors stated for decay data are also within the range of 1 to 5%. However, for some nuclides the uncertainty is substantially greater, up to several ten per cent, e.g. for ^{65}Ni (gamma abundance), ^{42}K (gamma-to-beta ratio), ^{203}Hg (internal conversion coefficient ratio), ^{86}Rb (beta-branching ratio), etc.

Conditions are more critical with the agreement of individual compilations in reaction cross-sections for neutron activation, where values for reactions of considerable interest in activation analysis can be found, which differ by a factor up to 2, e.g. for ^{202}Hg(n, γ)^{203}Hg or ^{65}Cu(n, γ)^{66}Cu. There even exist cases where the suggested values differ by a factor of about 10, for instance for the 14-MeV neutron cross-section of the ^{90}Zr(n, 2n)^{89m}Zr reaction which can be interesting for activation analysis. One always has to keep in mind that even the best known thermal activation cross-sections have a possible error of approximately 10%, only few have smaller ones. Normally, a substantially larger error has to be expected for resonance integrals and 14-MeV neutrons cross-sections as well as for excitation functions of reactions using fast neutrons, photons and charged particles.

3.3. The significance of systematics

An additional serious problem may arise with respect to completeness of existing nuclear data. Above all, this concerns the excitation functions, where many gaps in experimental data are to be found. For most nuclides only parts of the excitation functions are known, or the excitation functions have not been measured, at all. This is particularly true for (n, threshold) reactions. Therefore, substantial emphasis is laid on the development of special systematics.

The already existing systematics of excitation functions for charged-particle induced reactions [43] and for fast neutron induced reactions [65, 66] are based on the dependence of the characteristic magnitudes (maximum cross-section, the position of the maximum cross-section, FWHM, and the asymmetry-factor at FWHM) of an excitation function on the atomic number of the target nuclide. Using these systematics, an unknown excitation function can be constructed. In the case of (n, threshold) reactions, the systematics were checked by comparing the activities calculated using the constructed excitation functions and the experimentally determined activities [65, 66]. The deviations obtained for nuclides, which are close to the stability curve, are in general smaller than the scattering of experimental data obtained by different authors.

The systematics of excitation functions for (n, threshold) reactions have been applied in calculations to obtain trends for sensitivities of fast neutron activation analysis using a cyclotron [15] – to give only one example of the application of such systematics. Certainly, these systematics are very useful, above all in those cases where no or only incomplete, experimental excitation functions are available. But they may also be useful when the experimental data are very contradictory.

Systematic trends of excitation function parameters for photon induced reactions [14, 81, 82] and of cross-sections for 14-MeV neutron induced reactions [83-85] have also been described.

For many radionuclides decay data are also missing. In activation analysis this makes itself especially felt in branching ratios where absolute values are frequently missing. For instance, the most recent catalogue of gamma energies by Meixner [60] gives gamma abundancies with respect to the listed nuclides as follows: 52% absolute values for all energies, 12% absolute values for only some energies, 24% relative values for all energies, 7% relative values for only some energies, and for approximately 5% of the nuclides neither the absolute nor the relative intensities are known. Unfortunately, the systematics for decay data is substantially more difficult than that for cross-sections and excitation functions.

3.4. Some aspects of preparing future compilations

As has been shown, a number of valuable compilations of nuclear data exist which have either been especially prepared for activation analysis or which can be used largely for its purposes. Nevertheless, the present situation in handbooks and compilations could still be improved. This is, of course, less important for laboratories where an activation method is used for daily routine analysis. The need for improved compilations and handbooks will be felt especially in research establishments where only a small part of analysis is of routine nature and where new materials are continually being analysed, i.e. the kind of trace elements to be determined and of the matrix materials vary frequently. This need will increase even more if the instrumental equipment for several activation analytical methods is available. In such cases the nuclear data have to be looked for in many data sources, frequently also in original papers. An activation analytical handbook containing the most recent recommended nuclear data for the most important types of activation for each element, namely as element to be determined and as matrix, would be very desirable. On the other hand, compiling a new handbook is not unproblematic. Such a special compilation of data is only meaningful if no substantial alteration in the state of the data is to be expected within the next few years.

The excellent energy resolution of semiconductor detectors introduced in the years 1964-1966 led to a break-through in the quality of the data. The first detectors were unfortunately very small, and only the semiconductor detectors manufactured in the last 5-6 years satisfy the highest demands of measurement techniques.

In general, the radionuclides located close to the stability curve are of interest for activation analysis. The nuclear data for the predominant majority of these nuclides have already been measured using the most modern measuring techniques. Certainly, in the years to come development of the experimental techniques will go on yielding improvements in some

points, however, no break-through is to be expected which would enable one to obtain extensively more accurate and more precise nuclear data than in recent years.

Therefore, the time would now seem ripe for beginning the preparation of a new comprehensive compilation for activation analysis. A successful compilation can only result from close collaboration of analysts, compilers, evaluators and experimenters. The analysts would clearly determine the desirable data and their optimum presentation in the compilation. The most demanding part of the preparation of such a compilation would be up to the compilers and evaluators, i.e. selection of the most reliable values from the available experimental data, use of systematics and, if possible, also of nuclear theory for obtaining data in cases where experimental data are missing, and indication of cases where reliable data cannot be obtained by either of the two ways. Then it could be a task for experimenters to try to supply the most important missing data.

The international co-operation, developing successfully in evaluation and compilation of nuclear data [86] could facilitate or even make possible the realization of such a project. The necessary expenditure would pay off for two reasons. One of them is to be seen in the significance and widespread utilization of activation analysis, the other in the fact that most of the data are required also for other nuclear applications.

4. ANALYSIS BY DIRECT OBSERVATION OF NUCLEAR REACTIONS

As has already been mentioned, prompt-radiation activation analysis and analysis based on elastic scattering of heavy charged particles may be viewed as partially belonging to some extent to activation analytical techniques.

4.1. Prompt-radiation activation analysis

This kind of activation analysis is based on the measurement of the prompt particle or photon released from the compound nucleus following absorption of a projectile particle.

Analysis by prompt-gamma radiation following neutron capture in the (n, γ) reaction has been most extensively developed. The excitation energy of the compound nucleus is given off by the emission of prompt-gamma-radiation. The branching ratios and the energies of the transition from the excited state are characteristic for each nuclide. The energy of the neutron capture radiation covers a range from 50 keV to approximately 10 MeV. The spectra of most elements are very complex because of numerous populated levels. The data required for activation analysis by prompt-gamma-ray following neutron capture are isotopic abundance, total thermal-neutron-capture cross-sections, prompt-gamma ray energies, and absolute and relative gamma-ray intensities. The prompt-gamma-ray energies and intensities are relatively well known, and there exist also excellent tabulations by Bartholomew et al. [87-89], by Rasmussen et al. [90], and by Duffey et al. [91, 92].

Activation analysis by observation of prompt-charged particles emitted in nuclear reactions induced by charged particles on light nuclides is being increasingly applied. The most important data for these purposes are:

cross-sections, Q-values, Coulomb barriers, and stopping powers. They have been already discussed in connexion with charged-particle activation analysis.

4.2. Analysis by elastic scattering

Significant new developments have been made in the analytical application of elastic scattering of heavy charged particles, such as protons, α-particles,

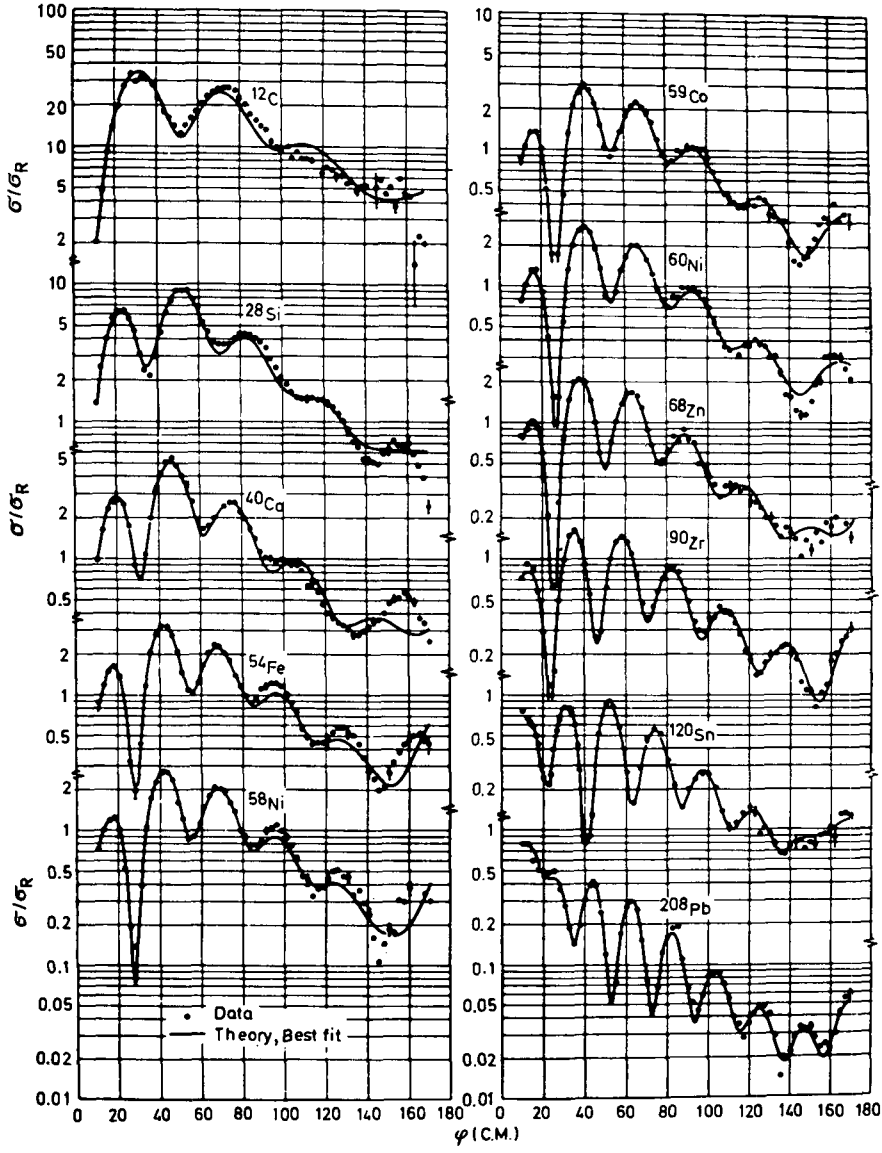

FIG. 8. The ratio of experimental (σ) and Rutherford (σ_R) elastic scattering cross-sections as a function of the scattering angle φ for 40-MeV protons [94].

and heavier ions [93]. The most important data needed for this technique are the scattering cross-sections and stopping powers.

The Rutherford model of elastic scattering can be applied to explain the physical principle of the method. However, in practical applications the Rutherford cross-sections σ_R do not suffice. The knowledge of the dependence of the experimental cross-sections σ or the ratio σ/σ_R on the scattering angle φ is essential for the nuclides of the trace elements to be determined as well as the nuclides of the matrix elements in order to optimize the irradiation conditions and/or to carry out the analysis according to the absolute method. Figure 8 shows examples of such a dependence. Unfortunately, these experimental data are not available for all nuclides and energy ranges coming in question for carrying out this analysis. As yet, no compilations of elastic-scattering data, similar to those existing for conventional activation analysis, have been prepared. For this reason, the existing bibliography [95-98] is of special significance. The same is true for inelastic-scattering cross-sections which must be considered one of the possible interferences. To master interferences of this type the excitation states must also be known. Additional interferences can be caused by prompt radiation of nuclear reactions discussed above.

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DISCUSSION

D. BERÉNYI: I would like to inquire about the availability of tabulations of cross-sections for fast neutrons, i.e. with energies different from 14 MeV.

These would be very useful for planning activation analysis experiments with a neutron source.

V. KRIVÁŇ: Cross-sections of this kind can be found in the BNL 325 series or, for (n, 2n)-, (n, γ)- and (n, d)-reactions, use can be made of the systematics developed by us, which enables one to construct unknown excitation functions.

D. BERÉNYI: In cases where the cross-section data are as scattered as those we have seen in some of the tables shown, the problem cannot be solved simply by computation or by carrying out one further measurement. Thorough analysis must be made of the experimental conditions on which the published measurements are based, in certain cases with control experiments to demonstrate why particular values are so scattered. Only after such an analysis can we plan and perform a new and more accurate experiment and thus hope to settle the question.

V. KRIVÁŇ: I would like to add the following to your comment. In principle, there are four steps which we can take in making "recommended values" for the case of very contradictory experimental data: (1) careful analysis of the experimental conditions under which the relevant studies were carried out; (2) performance of additional measurements; (3) comparison with theoretical calculations; and (4) the use of systematics, where available.

A.H.W. ATEN: I should like to come back to the fact mentioned by the preceding speakers, namely, that new measurements do not eliminate the spread in published data. For this reason it would be useful if two or more good physicists could remeasure standard cross-sections and try to arrive at results which agree. With the use of modern methods this should be possible with an accuracy of a few per cent. The work could be done in modestly equipped laboratories. The measurements in question would presumably be thermal cross-sections, resonance integrals and 14 MeV cross-sections. In future one might also include cross-sections for fission neutrons, which by then would presumably be californium neutrons.

A second point I should like to mention is a recommendation made by several international organizations interested in the subject of neutron measurements. If a certain neutron fluence is used for irradiation and if a gamma-measurement is performed the intensity of the signal is determined by the product of the cross-section and the intensity (percentage abundance) of the gamma-ray. For this reason a cross-section measurement can only be evaluated or used if details are known concerning the decay scheme adopted in the interpretation of the original measurement. As recommended by the international organizations in the field, such details should be mentioned in connection with cross-section measurement.

D.J. HOREN: In connection with Dr. Aten's comment, it seems to me that it may be time for the users of nuclear data to convey their desires for researchers to include the details of their measurements in submitting their papers to the editors of the basic nuclear physics journals. Some of us have not been having much success in inducing them to do so.

N.M. SPYROU: I should like to make the specific request that future compilations indicate not only the energies of internal conversion electrons but also their absolute intensities. In some cases it may be more suitable to measure a radionuclide by electron spectrometry (by measuring internal conversion electrons) rather than by gamma-ray spectrometry; although the latter of course is the usual method.

K.H. LIESER: As we chemists prefer to avoid chemical operations whenever possible, I would like to emphasize that for non-destructive activation analysis a thorough knowledge of all γ-lines is very important. To take an example: in the ppm range, there may be interference by unknown γ-lines of macrocomponents with transition probabilities of 10^{-6}.

M. LEDERER: In reactor irradiations, variability of neutron flux implies a need for internal standards; variability of the neutron spectrum requires that a standard of the same element be used. In view of these problems, how useful are accurate neutron cross-sections? Or, in other words, can accurate analysis be done by absolute methods?

V. KRIVÁŇ: I believe that absolute activation analysis can be used successfully in many situations especially where an accuracy of 10-20% is acceptable, which is very frequently the case. And I must admit that, from the point of view of accuracy, there is less risk in using the comparative method, particularly when the sample and the standard are as similar in composition as possible. For dealing with the variability of neutral fluxes, use is made of monitors, and in this case the accuracy of nuclear data is significant as a matter of principle.

F. DYER: I might comment that at the Oak Ridge National Laboratory we carry out neutron activation analysis almost entirely by absolute methods because of its lower cost compared with the comparative methods.

I should also like to ask how many radionuclides are included in Meixner's Table.

V. KRIVÁŇ: As Mr. Meixner is present, I shall ask him to answer your question.

C. MEIXNER: I think it is about 450.

F. DYER: How is the table being distributed?

C. MEIXNER: You can receive it by writing to me.

D.J. HOREN: I should like to ask Mr. Meixner what effort went into his compilation? Did it involve a detailed analysis of all the data to determine absolute intensities?

C. MEIXNER: The time required for preparing the compilation was about three years. A check was made on the literature since 1967, on the basis of recent references of the Nuclear Data Sheets, and for the literature from the period before 1967 I used information from the Table of Isotopes (LEDERER, C.M., HOLLANDER, J.M., PERLMAN, I., 6th ed., Wiley, New York 1967).

So far it has seemed impossible to me to determine all the absolute intensities of gamma rays because of the lack of experimental information, e.g. if there is only one publication which gives only relative intensities, without further information on conversion coefficients and absolute β-intensities or anything else, it is impossible to calculate absolute intensities.

Speaking for myself I have always tried to calculate absolute intensities but very often this has proved impossible, especially in the case of short-lived fission products.

Section XI

ACTIVATION ANALYSIS: NEUTRONS

Chairman

P. ALBERT (France)

NUCLEAR DATA BASES FOR ACTIVATION ANALYSES

J.M. BEWERS, G.J. PEARSON
Atlantic Oceanographic Laboratory,
Bedford Institute of Oceanography,
Dartmouth, Nova Scotia, Canada

W.F. MERRITT
Biology and Health Physics Division,
Chalk River Nuclear Laboratories,
Atomic Energy of Canada Limited,
Chalk River, Ontario, Canada

Abstract

NUCLEAR DATA BASES FOR ACTIVATION ANALYSES.

This paper surveys several existing nuclear data compilations and discusses their applicability to activation analysis. These compilations generally fall into two categories: those dealing solely with neutron activation data, both for instrumental and radiochemical purposes, and those providing more general data for a wide range of nuclear activation mechanisms. The former type is mainly restricted to a single application while, to date, the latter type is only useful for reference purposes. Two approaches to data reduction in instrumental activation analysis are currently employed. The first method utilizes a library of standard spectra which are unique to the particular irradiation and counting facilities used. In contrast, the second approach uses a library of basic nuclear parameters together with a mono-standard flux determination. Experience with an existing data base of this latter type for the semi-automatic analysis of environmental samples has revealed some deficiencies in contemporary nuclear information. These predominate in the areas of absolute gamma-ray intensities and particle capture cross-sections. Extensive use of this system has revealed other requirements not directly related to the reliability of the available nuclear data. To meet these requirements for quality control purposes, it was possible to generate different hard-copy sub-sets of the data file, notably photon energy and isotope-ordered listings and a compilation of gamma-ray interferences. There is also an obvious need for more technical information regarding reactor characteristics, e.g. neutron energy profiles and flux gradients. A list of parameters for charged-particle, photon, slow-neutron and fast-neutron activation analysis is discussed in relation to the establishment of an international master data base.

Introduction

Since the introduction of high resolution (Ge(Li)) γ-ray detectors it has been feasible to carry out instrumental, or non-destructive, neutron activation analysis on a wide variety of samples. Generally the methods employed can be divided into two categories. The first, regularly used when the content of the material to be analyzed is known qualitatively, compares the spectrum from the unknown with a catalogue of spectral data generated by the irradiation of known quantities of the elements involved. The second method has advantages when the composition of the samples is unknown, varying, or when a large number of elements are to be determined. It utilizes a library of nuclear data and a neutron flux monitor for the solution of spectra from the irradiated samples. This paper will confine itself primarily to the second method and will discuss the compilations of nuclear data and the techniques required for its use.

Compilations

Since the advent of neutron activation analysis there has been a continual need for reliable nuclear data. Initially much of this material was extracted from primary sources such as journals and secondary sources such as the Nuclear Data Sheets (1). The first important compilation of data directly useful for activation analysis was Heath's "Gamma Ray Spectrum Catalogue" (2) which appeared in 1957. In the early 1960's many such sources were published. Noteworthy were Koch's "Activation Analysis Handbook" (3) and "Tables of Gamma Rays from the Decay of Radionuclides" (4) by Hawkings, Edwards and McLeod. Koch provided a thorough compilation of nuclear reactions (with cross sections and experimental sensitivities) applicable to activation analysis of each element together with a comprehensive list of interfering reactions. Hawkings et al. were the first to provide both ascending energy and ascending half-life subsets of radionuclide γ-rays as a further aid to spectral peak identification. With these and other compilations (5,6) it was possible to carry out multi-element analysis, using sodium iodide detectors and post-irradiation chemistry, without continual reference to primary data sources.

When Ge(Li) detectors came into general use, the inherent selectivity of the counting procedures was markedly increased. Many energy levels and decay schemes were subsequently re-examined to produce better γ-ray energy and intensity values; and in 1968 and 1969 there appeared several compilations containing improved data. Undoubtedly the most versatile of these publications was the sixth edition of "Table of Isotopes" (7) by Lederer et al. which included basic nuclear level schemes, abbreviated data compilations for engineering and analytical applications and a list of precisely determined γ-ray calibration energies. At about this time it was realized that the continuing improvement in resolution of Ge(Li) detectors, combined with computerized spectral reduction, might obviate post-irradiation chemical separations. To assist analysts moving in this direction, Adams and Dams (8) published in 1969 a compilation of precisely determined energies of the main γ-rays for radionuclides produced by thermal neutron capture. Subsequently these authors published an additional list of γ-rays from fission product nuclides (9). Also in 1969 Heath (10) produced a new catalogue of nuclear data for both stable and radioactive nuclides, ordered by element only. It is significant that the author adopted numerical as opposed to spectral or analogue data presentation.

A comprehensive catalogue of thermal and fast neutron activated nuclides and their associated gamma radiations was prepared by the Nuclear Data Centre of Washington State University in 1969 and was revised in 1970 (11). It is perhaps still the most complete compilation for thermal neutron activation purposes. In 1970, Balagna and Helmick (12) at Los Alamos irradiated some 70 elements with thermal neutrons and analyzed their induced activities using a high resolution Ge(Li) spectrometer. Their atlas presents plots of the spectra and tables of the γ-rays ordered both by energy and by element. A more general catalogue of γ-rays was published by Wakat in 1971 (13), although the author had discussed its format and content some three years earlier (14). This compilation contains an energy-ordered list of γ-rays for radionuclides produced by all reaction modes.

However, since thermal neutron cross sections, atomic numbers and parent isotope abundances were not included, it is useful only for manual identification and inspection procedures.

Also in 1971, Pagden, Pearson and Bewers (15) of the Bedford Institute of Oceanography, Canada, published a printed version of their computer library for instrumental activation analysis. It is similar in content to that of the Washington State University group but more easily understood because of its self-explanatory headings and comparatively open format. Much common material exists in the previously described data compilations but the important distinction between that of Pagden et al. and the others is that the former is a working data base for instrumental thermal neutron activation analysis. While some of the compilations, in particular the Washington State University file and Heath's "Table of the Isotopes" contain sufficient material to be adapted to these purposes, the principal applications are for visual reference and spectral inspection. None of the catalogues, as published, is easily adapted to assessing the degree of radiochemical separation required for conventional activation analysis, and this aspect will be dealt with later.

Instrumental Activation Analysis

As previously mentioned there are two basic approaches to instrumental or non-destructive activation analysis. In the first category, used when the general composition of the sample is known, the γ-ray spectra of the irradiated sample can be solved by choosing component-element standard spectra from a library and iteratively transforming them into a least-squares regression fit to the unknown (16, 17, 18, 19, 20, 21). Within the same category, another method is to calculate peak areas and energies and relate these data to a library of similar parameters obtained from irradiating standards (22, 23, 24). These methods require that the data for the libraries be obtained under identical conditions to those pertaining to the sample and therefore they are only applicable to the irradiation and counting facilities for which they were designed. The libraries must of course contain examples of all sample constituent elements in order to account completely for all possible interferences.

A library of basic nuclear data, rather than a library of experimentally determined parameters is used in the second major category of instrumental procedures for analyzing spectra. This approach is preferable when the general composition of the sample is unknown or when such large numbers of elements are to be determined that time and costs prohibit the preparation and analysis of standards. Many environmental samples fall into this class. Although calibrated γ-ray standards are used to measure the efficiency of the counting system and standards may be used to characterize a particular irradiation facility initially, the aim of the method is to provide a reliable analytical procedure using a single standard such as a cobalt flux monitor. A unique characteristic of this approach is the entirely numerical handling of spectra. It is therefore necessary that γ-ray spectra be reduced to peak areas and energies. Methods in this second category are based upon the use of the equation for post-irradiation photon emission rate of a radioisotope formed by thermal neutron capture:

$$S_\gamma = \sigma_{th} \phi I_\gamma \frac{W k}{M} N (1 - e^{-0.693 t_{irr}/T_{\frac{1}{2}}}) e^{-0.693 t_{dec}/T_{\frac{1}{2}}}$$

where
- S_γ is the emission rate of a gamma ray of energy Eγ from a product radioisotope R
- σ_{th} is the thermal neutron capture cross section
- ϕ is the thermal neutron flux
- I_γ is the absolute intensity of photons of energy Eγ from the product radioisotope R in γ's/decay
- W is the mass of the target element in the irradiated sample
- k is the fractional abundance of the target isotope
- N is Avogadro's number
- t_{irr} is the irradiation period
- $T_{\frac{1}{2}}$ is the half-life of the product radioisotope R
- t_{dec} is the post-irradiation decay time
- M is the atomic weight of the target element

The parameters k, N, M, I_γ, σ_{th} and $T_{\frac{1}{2}}$ are constants of a chemical or nuclear nature, while t_{irr}, t_{dec} and ϕ describe experimental conditions. If ϕ and S_γ can be determined, it is possible to deduce W, the mass of the target element in the sample. ϕ is measured by using a single standard such as a cobalt flux monitor. The evaluation of S_γ is based upon the determination of the peak areas and energies of the γ-ray spectrum.

The accuracy with which W is determined will depend upon the accuracy with which all the other parameters are known, but in general, the limiting factors will be the uncertainties in cross section, flux and S_γ. For most samples the values in the library should be adequate but if high accuracy is required it may be necessary to measure σ_{th} and ϕ initially with standards. For subsequent irradiations, determination of ϕ with a flux monitor is sufficient, provided that the neutron spectrum of the irradiation facility has not changed. The accuracy with which S_γ can be determined is largely a function of the complexity of the γ-ray spectra. In addition, it is essential that all possible products of thermal neutron bombardment are considered as possible constituents of the post-irradiation sample mixture, and the library of nuclear data must include information on significant (n, f), (n, P) etc. reaction products.

In 1967, Gunnink, Levy and Niday (25, 26), developed an instrumental activation technique of this type. Their basic library consisted of nuclear data including half-lives, γ-ray energies, γ-ray intensities and parent-daughter relationships. Their procedure could be subdivided into three stages: the conversion of spectral data to photopeak energies and areas, the identification of constituent nuclides and the conversion of peak intensities to activities based on counting geometry and detector efficiency.

Constituent elements are then calculated using thermal neutron capture cross sections, neutron fluxes, isotopic abundances and irradiation times.

In 1970, the Bedford Institute group reported on a system of instrumental activation analysis (27) using the single standard approach. The procedure was designed and tested for the analysis by neutron activation of a wide range of oceanographic samples for which it was not possible to make prior assumptions regarding composition.

The system consisted of a set of computer routine together with a disc-based library of nuclear data and detector efficiency calibrations. A subset of the library was subsequently published (15) and has already been discussed. The complete analytical procedure involves the selection for each spectrum of an overall set of possible constituent radionuclides which is systematically reduced to a "best set" by various tests based on the catalogued nuclear data. First the photopeak areas and energies of the sample spectrum are determined by the method of Routti (28). The initial nuclide set is then assembled by comparing these photopeak energies with all γ-ray lines stored in the data base. Nuclides having lines which match spectral lines to within a specified energy limit based on detector resolution are then retained in the set. Subsequent computer routines then test each nuclide of the set for suitability of half-life, presence or absence of most intense line, presence or absence of second and third strongest lines and the probability of production during the sample irradiation. These tests are carried out sequentially and employ the catalogue parameters of half-life, γ-ray energy, γ-ray relative intensity, target stable isotope abundance and thermal neutron capture cross section. Input data for the tests include duration of irradiation, decay time of the sample prior to counting and pulse height analyzer live-time. At the completion of each test the user has the option of obtaining print outs of surviving and rejected nuclides. Manual intervention is possible before the application of any of the routines but is normally only exercised after the probability of production test.

The surviving "best set" of possible constituent nuclides is then arranged into an activity matrix and equated to the spectral peak areas in an equation of the form

$$I_i = \sum_{j=1,m} \varepsilon_i A_j a_{ij} \qquad \begin{matrix} i = 1, n \\ j = 1, m \end{matrix}$$

where
- I_i is the peak sum of the i th photopeak of energy E_i
- ε_i is the detector efficiency at the energy E_i
- A_j is the relative activity of the j th nuclide
- a_{ij} is the relative photon intensity of the i th line of the j th nuclide
- n is the number of spectral lines
- m is the number of nuclides in the "best set"

TABLE I. INTENSITIES OF ^{182}Ta GAMMA RAYS (γ's/100 DECAYS)

Gamma Ray Energy (keV)	Original catalogue 1967*	Matrix predictions 1969*	Lederer et al. (7) 1968	Heath (10) 1970	Filby et al. (11) 1970	Wakat (13) 1971	Pagden et al. (15) 1971
152	13.6	6.5	7	57	7.1	7	7.3
222	13.6	8.5	8	97	7.98	8	8.0
229	6.8	3.9	3	3.6	3.8	3.4	4.0
1122	37	37	-	35.6	37	37	35
1222	31	31	-	27.9	28.9	28.9	27.8

* Relative intensities normalised to 37 @ 1122 keV.

The solution is weighted by the spectral line intensities and the resulting matrix equation is solved by the method of least squares. Since it is implicit that the matrix of the a_{ij} terms must be non-singular, a computer routine excludes non-soluble nuclides, while ensuring that the maximum number of solutions are obtained. The resulting A_j's are a set of numbers proportional to the activities of the j nuclides. A final routine uses catalogued values of thermal neutron cross sections and ratios of relative to absolute γ-ray intensities, together with input figures for cobalt flux monitor and sample mass to calculate final values of element concentration in the sample. In cases where a spectrum is found to contain products of reactions other than (n, γ)'s (e.g. fission), no calculations other than isotope activity are carried out.

A set of test spectra from irradiated samples of Cr, Ru, Tb, Hf, and Ta, counted both singly and in combination achieved acceptable results but pointed out discrepancies in intensity values for Ta and Hf. Table I shows the data for ^{182}Ta used in the original catalogue (1967, unpublished) of the Bedford Institute group, the predicted values from the matrix analysis and the values presented in more recent compilations. It is immediately evident that intensity values predicted by the matrix analysis are in good agreement with the most recent compilations. The large differences noticeable in Heath's compilation (10) are probably due to typographical errors.

Because of the least-squares method used for the solution of spectra, the system is quite sensitive to errors in the catalogued data, and an error in the intensity data for one nuclide may affect the calculations of activity for many others. Therefore reliable intensity values are necessary to carry out effective comparative analyses. In order to extend the use of the system to absolute analyses, accurate cross-section data are also required.

The foregoing discussion of a particular analysis system clearly indicates the advantage of a computer-based data file over a printed reference catalogue. Subsets and rearrangements of the base file are easily obtained and can provide compilations ordered by energy, half-life, nuclide, etc., through new programming. The subsets can be made available as either printed compilations or further computer files for analytical purposes. Appropriate programming and estimates of the probable sample matrix can be used with the data base to predict optimum irradiation and counting procedures for the analysis of particular samples.

A particularly useful mathematical treatment of the existing data file (29) has provided a catalogue of possible spectral interferences encountered in thermal neutron activation analysis. This computes the degree of interference of the five most intense γ-rays of each catalogued radionuclide with all other lines in the catalogue subject to an "interference width" of $\pm$ 5 keV from the reference energy. It is divided into three subsets corresponding to reactor irradiation times of 10 minutes, 1.5 hours and 12 hours and values of relative peak height at the end of the irradiation are given for equal element mass.

This compilation can be used very profitably by laboratories which do not have sophisticated computer techniques to predict possible constituents

of multiplets in the sample spectrum. Also, predictions of detectability of one element in the presence of another can be made, optimum lines for detecting a particular element can be selected and the most advantageous use of radiochemical separations for unambiguous analysis can be determined.

The interference listing has also shown that the method of assigning radionuclides with well-known decay schemes to specific spectral peaks is not practical for the analysis of samples containing more than a few elements. Even the highest resolution Ge(Li) detectors do not resolve this problem since significant interferences with centroid differences of less than 1 keV are common, contrary to the remarks of Verheijke (30). The matrix method, which by its nature accounts for all interferences, is the most reliable approach to the analysis of complex samples.

Concluding Remarks

The advancement of automated activation analysis is dependent upon refinement in the areas of available nuclear data and information regarding service irradiation facilities. Irradiation positions which are well characterized for total flux and neutron energy distribution are essential for reliable analysis. It is relatively simple to obtain these conditions in low flux positions near the reactor shield or in the thermal column. It will not, however, be possible to use high flux core positions unless detailed information regarding the neutron spectrum is available.

With respect to nuclear data, the limiting factors for reliable automated analysis at the time of the most recent data compilations (10, 11, 15) are the accuracy of absolute γ-ray intensity and neutron capture cross-section values. An example of this type of problem is given in Table II which shows a chronological survey of the γ-ray intensity data for ^{57}Co as given by commercial suppliers and in the most recent compilations. Although ^{57}Co has been used in standard calibration sources for a number of years, the ratio of the intensity of the 122 and 137 keV lines varies from 7.4 to 10.2. Similar discrepancies will probably be apparent in other more complex and less thoroughly studied decay schemes. Reliable cross-section data is also needed if absolute, rather than comparative analyses are to be performed using this technique.

Further nuclear research directed toward the correction of these deficiencies would be welcome.

As better information becomes available the need for an international nuclear data bank for all purposes can be foreseen. The basis of such a compilation would naturally be the nuclear level structure. The base should include all nuclear parameters such as name of element, atomic number, mass number, decay mode, level lifetimes, γ-ray energies, γ-ray absolute intensities, half-life, branching ratios, particle decay energies, level energies and spins, transition multipolarity, isomeric transition strengths, Q values, cascade characterizations (doubles), particle capture cross-section profiles, etc. Chemical and atomic parameters such as isotopic

TABLE II. INTENSITIES OF ^{57}Co GAMMA RAYS (γ's/100 DECAYS)

Source		122 keV intensity	136 keV intensity	Ratio of 122 keV/136 keV intensities
Amersham	1962	88	10	8.8
IAEA	1966	85.3	8.4	10.2
Amersham	1966	88.8	8.8	10.1
Lederer et al.(7)	1967	87	11	7.9
Heath (10)	1969	85	11	7.7
Filby (11)	1970	85.3	8.54	10.0
Wakat (13)	1971	87	11	7.9
Amersham	1971	84.8	11.4	7.4
Pagden et al.(15)	1971	85	11	7.7

abundances, atomic mass, electronic structure and chemical periodicity should all be included for analytical purposes.

The large quantities of data available and the complex nature of the nuclear processes involved would require a detailed systems analysis to define the methods of data storage and retrieval. Continual updating of the file is essential and conventions regarding experimental uncertainties and file acceptance must be included as part of the systems approach. The end result would be a comprehensive data base which could be accessed to produce sub-files for any desired application including evaluation of contemporary data deficiencies. These sub-files would be made available either as printed outputs or on magnetic tape in order to be compatible with computer-based systems of analysis.

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DISCUSSION

D.J. HOREN: The comparison in Table II of the paper might be meaningless without a knowledge of the uncertainties assigned by the authors. Actually, there may very well be good agreement with these uncertainties.

As regards your desire for an international computerized data base, I too would like to see this. Maybe if you all write the NSF they might give us some funds to pursue this matter faster!

W.B. LEWIS: I would like to refer to another aspect of the question raised by Dr. Horen on your table of the ratio of intensities tabulated for the ^{57}Co γ-rays. These values derive from values given by measurers. We need some means of weeding the garden, i.e. cutting out results that the experimenter now knows to be false and may wish to withdraw. We need a means of communication to bring this into effect, say, by writing to the compilers and asking them to approach measurers with a request to exclude such information. Otherwise we may continue averaging in data that no one would really sponsor.

J.J. SCHMIDT: As an example to illustrate the comment of Dr. Lewis, I would like to mention that in the so-called EXFOR system, which is a system for international exchange (by means of computers) of experimental neutron nuclear data between the four main neutron data centres, each computer being in constant contact with the experimenter whose data are being entered into the EXFOR file, in order to verify whether the data are still valid, whether the experimenter wants to make changes, etc. Only with the explicit approval of the author are the data and associated physics information characterizing the experiment then finally entered into the file and through the four-centre network made available to users requesting these data.

M. LEDERER: With reference to Dr. Lewis' remark, I recall a comment by Miss K. Way some years ago that "the past goes away". Given the rate at which new and better measurements appear, one can hope that many incorrect measurements will be rejected by the evaluator on the light of newer data (especially in the matter of gamma-ray spectra).

Miss K. WAY: We used to write to experimenters whose values seemed discrepant with those of others to ask them to review their results. Often they found reason to correct or withdraw their data. This was duly noted on the Data Sheets.

RESONANCE INTEGRALS APPLIED TO THE MULTIPLE-COMPARATOR METHOD IN REACTOR NEUTRON ACTIVATION ANALYSIS

R. VAN DER LINDEN*, F. DE CORTE**, J. HOSTE
Institute for Nuclear Sciences, Ghent University,
Belgium

Abstract

RESONANCE INTEGRALS APPLIED TO THE MULTIPLE-COMPARATOR METHOD IN REACTOR NEUTRON ACTIVATION ANALYSIS.

Classical reactor neutron activation analysis requires preparation, irradiation and counting of a standard for each element to be determined. This method is time-consuming, especially for routine multi-element analysis, and may be a cause of errors, mainly due to flux gradients in the irradiation capsule. Moreover, unexpected elements, for which no standards are co-irradiated, cannot be determined.

These disadvantages can be avoided by applying a multiple comparator method. In this technique, the specific activities of the standards are derived from, at least, two comparator isotopes irradiated together with the sample.

If the ratios of the specific activities of the isotopes investigated to the specific activities of the comparator isotopes (k-values) are experimentally determined in a well-defined standard reactor site, they can be converted for each new irradiation position. As the isotopic activity induced by reactor neutron irradiation is an additive function for thermal and epithermal activation, the conversion of the k-values should take into account the thermal and the epithermal activation parameters, not only of the reactor neutron spectrum, but also of the isotopes. As far as the former is concerned, it can be proved mathematically that the knowledge of the ratios of thermal to epithermal neutron fluxes ($\varphi_{th}/\varphi_{epi}$) for the standard and the unknown irradiation position is sufficient for the conversion of the k-values. As to the isotopic parameters, it can be shown that only the ratios of the infinite dilution resonance integrals to the thermal activation cross-sections (I_0/σ_{th}) are needed for the isotopes determined and for the comparator isotopes. Thus it can be stated that the multiple-comparator method requires: a) experimental determination of the k-values under well-defined irradiation and counting conditions. These ratios can be evaluated afterwards, thus offering the possibility of determining unexpected elements; b) knowledge of the flux ratios for the irradiation positions. These ratios can be calculated from the activities of, at least, two irradiated comparator isotopes, using a relative activity measurement method; c) knowledge of accurate I_0/σ_{th}-values. Because of the lack of well-known I_0-values for many isotopes, the I_0/σ_{th}-values were determined for all γ-emitting nuclides useful in reaction neutron activation analysis. The measurements were carried out according to the Høgdahl flux convention. The values were determined in different reactor neutron spectra. In some cases, an experimental study was made to avoid self-shielding. Errors are estimated to be lower than 5%.

A study of the error multiplication, when applying the multiple-comparator method, was performed. The method was analysed stepwise, and the change of the initial error caused by irradiating and measuring the isotopes is described as a function of the mathematical operations. A general expression is derived for the overall error on the final analytical result. This study of error multiplication is especially important in giving a mathematical solution for the choice of comparator isotopes and irradiation conditions for each individual analytical problem.

INTRODUCTION

As reactor neutron activation analysis is founded on the fact that the induced isotopic activities are directly proportional to the weights of the elements, it requires, in its classical form, preparation, irradiation and

* Research associate of the IWONL.

** Research associate of the NFWO.

counting of a standard for each element to be determined. Consequently, when dealing with routine multi-element analyses, a lack of space in the irradiation capsule can become a problem. Moreover, this procedure is very time-consuming, and, thus, to ensure identical irradiation conditions for standards and samples, it necessitates a number of precautions concerning the self-shielding of the elements and the flux gradients in the irradiation can. Finally, with classical activation analysis, it is impossible to analyse for unexpected elements, as no standards will be available.

In the literature, some of these problems have been solved in one way or another. A mixture or several mixture groups of the standards can be irradiated as a solution, followed by chemical separation [1] or Ge(Li) γ-spectrometry [2-4]. The solutions can be replaced by small paper strips [5] or ion exchange beads [6], on which the standards are spotted or adsorbed, respectively. For analyses of biological and geological samples, multi-standard materials can be used, such as dried kale powder [7] or standard rocks [8-10].

Another possibility consists in using a single-comparator method, e.g. a flux monitor irradiated together with the samples. This method has been critically evaluated by Girardi et al. [11]. It was found that the accuracy and precision are similar to those of the classical method. However, as stated earlier [12], the method is restricted to a constant ratio of thermal-to-epithermal reactor neutron flux, ϕ_{th}/ϕ_{epi}. If this condition is not met, the applicability of the procedure can be extended by an appropriate choice of the single comparator, namely with a I_0/σ_{th}-value (I_0 = activation resonance integral at infinite dilution; σ_{th} = thermal activation cross-section) close to the one of the elements. In spite of these adaptations, with the single-comparator method, no exact correction procedure is possible for variations of ϕ_{th}/ϕ_{epi}.

In this work, the single-comparator method is extended to a relative multiple-comparator method, making it possible to correct for the different ratios of thermal-to-epithermal neutron flux in the diverse irradiation positions. In this procedure, knowledge of the I_0/σ_{th}-values for the comparators and the elements under investigation is of fundamental importance. Because of lack of well-known values for the resonance integrals, the I_0/σ_{th}-ratios for 122 (n, γ) reactions were experimentally determined. To check the accuracy and precision of the method, a study of the error change, when applying the multiple-comparator method, was performed.

PRINCIPLE OF THE RELATIVE MULTIPLE-COMPARATOR METHOD

When irradiating an element in a nuclear reactor, the photopeak activity A of a radioisotope formed by the (n, γ)-reaction is given by the relation

$$A = \frac{\vartheta N_A E S D w (\sigma_{th} \phi_{th} + I_0 \phi_{epi})}{M} \qquad (1)$$

where ϑ is the isotopic abundance of the target nuclide, N_A Avogadro's number, E the counting efficiency for the γ-ray measured ($A = E_x$ disintegration rate), S the saturation factor = 1 - exp ($-\lambda t$) (λ being the decay constant and

t the irradiation time), D the decay factor = exp (-λT) (with T the decay time), w the weight of the element, M the atomic weight of the element, σ_{th} the thermal activation cross-section, I_0 the activation resonance integral at infinite dilution, ϕ_{th} the thermal neutron flux, and ϕ_{epi} the epithermal neutron flux.

If one defines the specific photopeak activity A_{sp} as

$$A_{sp} = \frac{A}{w S D} \tag{2}$$

and if one equals

$$\frac{\vartheta N_A E}{M} = C \tag{3}$$

Eq.(1) can be written as

$$A_{sp} = C(\sigma_{th} \phi_{th} + I_0 \phi_{epi}) \tag{4}$$

In classical neutron activation analysis the unknown weight of the element to be determined in the sample is calculated from the equation

$$A_{sp,x} = A_{sp,st} \tag{5}$$

where the subscript "x" refers to the sample and "st" to the standard.

Using a comparator method one can introduce a k-factor for each standard defined as,

$$k = \frac{A_{sp,st}}{A_{sp,comp}} \tag{6}$$

where the subscript "comp" refers to the comparator.

When determining these k-factors in a "reference" reactor position (subscript "ref"), they can be written as

$$k_{ref} = \left(\frac{A_{sp,st}}{A_{sp,comp}}\right)_{ref} = \frac{C_{st}}{C_{comp}} \frac{\left[\sigma_{th,st}\left(\frac{\phi_{th}}{\phi_{epi}}\right)_{ref} + I_{0,st}\right]}{\left[\sigma_{th,comp}\left(\frac{\phi_{th}}{\phi_{epi}}\right)_{ref} + I_{0,comp}\right]} \tag{7}$$

It should be mentioned that a "reference" reactor position refers to a known ratio of thermal-to-epithermal reactor neutron flux.

When analysing the sample by irradiation, together with the comparator, in an arbitrary reactor position (subscript "anal"), these k-factors are

$$k_{anal} = \frac{C_{st}}{C_{comp}} \frac{\left[\sigma_{th,st}\left(\frac{\phi_{th}}{\phi_{epi}}\right)_{anal} + I_{0,st}\right]}{\left[\sigma_{th,comp}\left(\frac{\phi_{th}}{\phi_{epi}}\right)_{anal} + I_{0,comp}\right]} \tag{8}$$

From Eqs (7) and (8) it follows that k_{anal} can be computed from k_{ref} by the relation

$$k_{anal} = k_{ref} \frac{\left[\left(\frac{\phi_{th}}{\phi_{epi}}\right)_{anal} + \left(\frac{I_0}{\sigma_{th}}\right)_{st}\right]\left[\left(\frac{\phi_{th}}{\phi_{epi}}\right)_{ref} + \left(\frac{I_0}{\sigma_{th}}\right)_{comp}\right]}{\left[\left(\frac{\phi_{th}}{\phi_{epi}}\right)_{anal} + \left(\frac{I_0}{\sigma_{th}}\right)_{comp}\right]\left[\left(\frac{\phi_{th}}{\phi_{epi}}\right)_{ref} + \left(\frac{I_0}{\sigma_{th}}\right)_{st}\right]} \tag{9}$$

Equation (9) shows that, concerning the reactor parameters, knowledge of the ratios of thermal-to-epithermal neutron fluxes for the "reference" and the "analysis" irradiation position is sufficient for the conversion of the k-values. For the "reference" reactor position, ϕ_{th}/ϕ_{epi} can be readily determined from the cadmium ratio, R_{Cd}, of a radioisotope with a known I_0/σ_{th}-value, according to the relations

$$R_{Cd} = \frac{A_{sp}}{(A_{sp})_{Cd}} \tag{10}$$

and

$$\frac{\phi_{th}}{\phi_{epi}} = (R_{Cd} - 1)\frac{I_0}{\sigma_{th}} \tag{11}$$

where $(A_{sp})_{Cd}$ is the specific photopeak activity induced by irradiation under a cadmium cover.

Since this technique has the disadvantage of causing large flux depressions in the neighbourhood of the cadmium foil, it cannot be used for the determination of the flux ratio in the "analysis" irradiation position. It is, however, possible to evaluate $(\phi_{th}/\phi_{epi})_{anal}$ when irradiating, at least, two comparators. For the activity ratio of two comparators, denoted hereafter as 1 and 2, in the "reference" reactor position, one can write

$$\left(\frac{A_{sp,\ comp\ 1}}{A_{sp,\ comp\ 2}}\right)_{ref} = \frac{C_{comp\ 1}\ \sigma_{th,\ comp\ 1}}{C_{comp\ 2}\ \sigma_{th,\ comp\ 2}} \frac{\left[\left(\frac{\phi_{th}}{\phi_{epi}}\right)_{ref} + \left(\frac{I_0}{\sigma_{th}}\right)_{comp\ 1}\right]}{\left[\left(\frac{\phi_{th}}{\phi_{epi}}\right)_{ref} + \left(\frac{I_0}{\sigma_{th}}\right)_{comp\ 2}\right]} \tag{12}$$

An analogous relation holds for the "analysis" reactor position

$$\left(\frac{A_{sp,\ comp\ 1}}{A_{sp,\ comp\ 2}}\right) = \frac{C_{comp\ 1}\ \sigma_{th,\ comp\ 1}}{C_{comp\ 2}\ \sigma_{th,\ comp\ 2}} \frac{\left[\left(\frac{\phi_{th}}{\phi_{epi}}\right)_{anal} + \left(\frac{I_0}{\sigma_{th}}\right)_{comp\ 1}\right]}{\left[\left(\frac{\phi_{th}}{\phi_{epi}}\right)_{anal} + \left(\frac{I_0}{\sigma_{th}}\right)_{comp\ 2}\right]} \tag{13}$$

By combination of Eqs (12) and (13) an R-value can be defined as:

$$R = \frac{\left(\frac{A_{sp,\ comp\ 1}}{A_{sp,\ comp\ 2}}\right)_{ref}}{\left(\frac{A_{sp,\ comp\ 1}}{A_{sp,\ comp\ 2}}\right)_{anal}} = \frac{\left[\left(\frac{\phi_{th}}{\phi_{epi}}\right)_{ref} + \left(\frac{I_0}{\sigma_{th}}\right)_{comp\ 1}\right]\left[\left(\frac{\phi_{th}}{\phi_{epi}}\right)_{anal} + \left(\frac{I_0}{\sigma_{th}}\right)_{comp\ 2}\right]}{\left[\left(\frac{\phi_{th}}{\phi_{epi}}\right)_{ref} + \left(\frac{I_0}{\sigma_{th}}\right)_{comp\ 2}\right]\left[\left(\frac{\phi_{th}}{\phi_{epi}}\right)_{anal} + \left(\frac{I_0}{\sigma_{th}}\right)_{comp\ 1}\right]} \qquad (14)$$

Finally, $(\phi_{th}/\phi_{epi})_{anal}$ can be computed from the experimentally determined R-value by the expression:

$$\left(\frac{\phi_{th}}{\phi_{epi}}\right)_{anal} = \frac{R\left[\left(\frac{\phi_{th}}{\phi_{epi}}\right)_{ref}\left(\frac{I_0}{\sigma_{th}}\right)_{comp\ 1} + \left(\frac{I_0}{\sigma_{th}}\right)_{comp\ 1}\left(\frac{I_0}{\sigma_{th}}\right)_{comp\ 2}\right] - \left[\left(\frac{\phi_{th}}{\phi_{epi}}\right)_{ref}\left(\frac{I_0}{\sigma_{th}}\right)_{comp\ 2} + \left(\frac{I_0}{\sigma_{th}}\right)_{comp\ 1}\left(\frac{I_0}{\sigma_{th}}\right)_{comp\ 2}\right]}{\left[\left(\frac{\phi_{th}}{\phi_{epi}}\right)_{ref} + \left(\frac{I_0}{\sigma_{th}}\right)_{comp\ 1}\right] - R\left[\left(\frac{\phi_{th}}{\phi_{epi}}\right)_{ref} + \left(\frac{I_0}{\sigma_{th}}\right)_{comp\ 2}\right]} \qquad (15)$$

The above-described comparator method is called a relative one, since the flux ratio $(\phi_{th}/\phi_{epi})_{anal}$ is calculated from relative activity measurements, i.e. without knowledge of the counting efficiency. It is obvious, however, that the counting conditions [E-factor from Eq.(1)] should be kept constant during the experimental work. This is an essential requirement for the application of the comparator method.

Although theoretically only two comparators are needed, the precision on the evaluation of the flux ratio $(\phi_{th}/\phi_{epi})_{anal}$ can be improved by the use of a multiple-comparator method. If N comparators are irradiated together with the sample and counted, $N(N-1)/2$ values of $(\phi_{th}/\phi_{epi})_{anal}$ can be computed. Another alternative of the relative multiple-comparator technique consists in using a multi-isotopic element as comparator. In this case, it is, of course, required that the photopeak activities of the different radioisotopes, produced by (n, γ)-reaction, can be measured simultaneously on a Ge(Li) detector.

From the above-described comparator method it follows that one of the attractive advantages is the possibility of analysing for unexpected elements. Indeed, the k_{ref}-factors of Eq.(7) can be determined experimentally afterwards, so that, with the aid of Eqs (15), (9) and (6), the specific photopeak activities $A_{sp,st}$ can be computed.

From Eqs (9) and (15) it is apparent that the relative multiple-comparator method can only give reliable results when the ratios of the activation resonance integrals at infinite dilution to the thermal cross-sections are on hand, both for the comparator and for the standard isotopes. Because of the lack of well-known I_0-values for many (n,γ)-reactions, the I_0/σ_{th} ratios were experimentally determined for most of the γ-emitting nuclides, useful in reactor neutron activation analysis. It should be mentioned that the I_0-values, used in this work, are defined according to the approach proposed by Høgdahl [13]:

$$I_0 = \int_{E_{Cd}}^{\infty} \sigma(E)\,\frac{dE}{E} \qquad (16)$$

where E stands for the neutron energy and E_{Cd} is the cadmium cut-off energy. According to the EANDC [14], E_{Cd} is set at 0.55 eV for a cylindrical

cadmium box with a wall thickness of 1 mm. The I_0/σ_{th}-ratios can be determined from the cadmium ratio R_{Cd} in a reactor position with a known ϕ_{th}/ϕ_{epi}-value, according to Eqs (10) and (11).

ERROR MULTIPLICATION BY APPLYING THE RELATIVE MULTIPLE-COMPARATOR METHOD

To summarize the above-described technique, it can be stated that the relative multiple-comparator method proceeds in four well-defined steps:

1. Experimental determination of the k_{ref}-ratios, [Eq.(6)], in a reference reactor position (with known $(\phi_{th}/\phi_{epi})_{ref}$-value). These k_{ref}-values can refer to one or more comparator isotopes.
2. Calculation of $(\phi_{th}/\phi_{epi})_{anal}$ [Eq.(15)].
3. Conversion of the k_{ref}-values to the k_{anal}-values [Eq.(9)].
4. Computation of the specific standard activities in the "analysis" reactor position [Eq.(6)].

Apparently, it is important to know the error on the k_{anal}-values, as a function of the experimentally determined R-values. For the sake of clearness, the calculations are sub-divided into two parts, namely the error on $(\phi_{th}/\phi_{epi})_{anal}$ relative to the error on R [Eq.(15)] and the error on k_{anal} relative to the error on $(\phi_{th}/\phi_{epi})_{anal}$ [Eq. (9)]. Finally, the error on k_{anal} relative to the error on R can be evaluated by combining the two separate parts. To simplify the mathematical equations, the following abbreviations are used:

$$\phi_{th}/\phi_{epi} = f$$

and

$$\frac{I_0}{\sigma_{th}} = D$$

Error on f_{anal} relative to the error on R

The error on f_{anal}, relative to the error on the experimentally determined R-value, can be expressed as

$$Z_f = \left| \frac{\frac{df_{anal}}{f_{anal}}}{\frac{dR}{R}} \right| \qquad (17)$$

Differentiating Eq.(15) gives for this Z_f-value:

$$Z_f = \left| \frac{R(f_{ref} + D_{comp\,1})(f_{ref} + D_{comp\,2})(D_{comp\,2} - D_{comp\,1})}{\left[R D_{comp\,1}(f_{ref} + D_{comp\,2}) - D_{comp\,2}(f_{ref} + D_{comp\,1})\right]\left[(f_{ref} + D_{comp\,1}) - R(f_{ref} + D_{comp\,2})\right]} \right| \qquad (18)$$

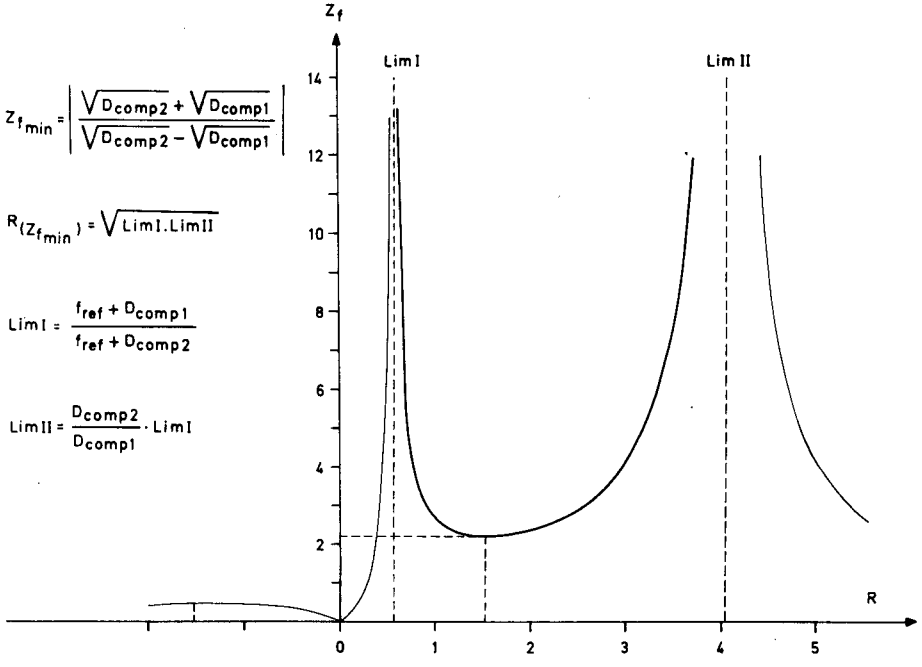

FIG.1. Z_f as a function of R[Eq.(18)].

$D_{comp\ 1} = 3.3$ (^{103}Ru); $D_{comp\ 2} = 23.1$ (^{97}Ru); $f_{ref} = 23.8$.

Equation (18), from which Z_f can be calculated as a function of R, is graphically plotted in Fig. 1. It is worth mentioning that the limits for R are given by

$$\frac{f_{ref} + D_{comp\ 1}}{f_{ref} + D_{comp\ 2}} \quad \text{and} \quad \frac{D_{comp\ 2}}{D_{comp\ 1}} \frac{f_{ref} + D_{comp\ 1}}{f_{ref} + D_{comp\ 2}} \tag{19}$$

the upper and lower limit depending on the value of $D_{comp\ 2}/D_{comp\ 1}$ compared to unity. The limits (19) can be derived from Eq.(15) where f_{anal} should have a positive value.

From Fig. 1 it can be concluded that the minimum Z_f-value corresponds to

$$R_{(Z_{f_{min}})} = \frac{f_{ref} + D_{comp\ 1}}{f_{ref} + D_{comp\ 2}} \sqrt{\frac{D_{comp\ 2}}{D_{comp\ 1}}} \tag{20}$$

this expression being the harmonical average of the limits for physical meaning of R. In this case $Z_{f_{min}}$ becomes

$$Z_{f_{min}} = \left| \frac{\sqrt{D_{comp\ 2}} + \sqrt{D_{comp\ 1}}}{\sqrt{D_{comp\ 2}} - \sqrt{D_{comp\ 1}}} \right| \tag{21}$$

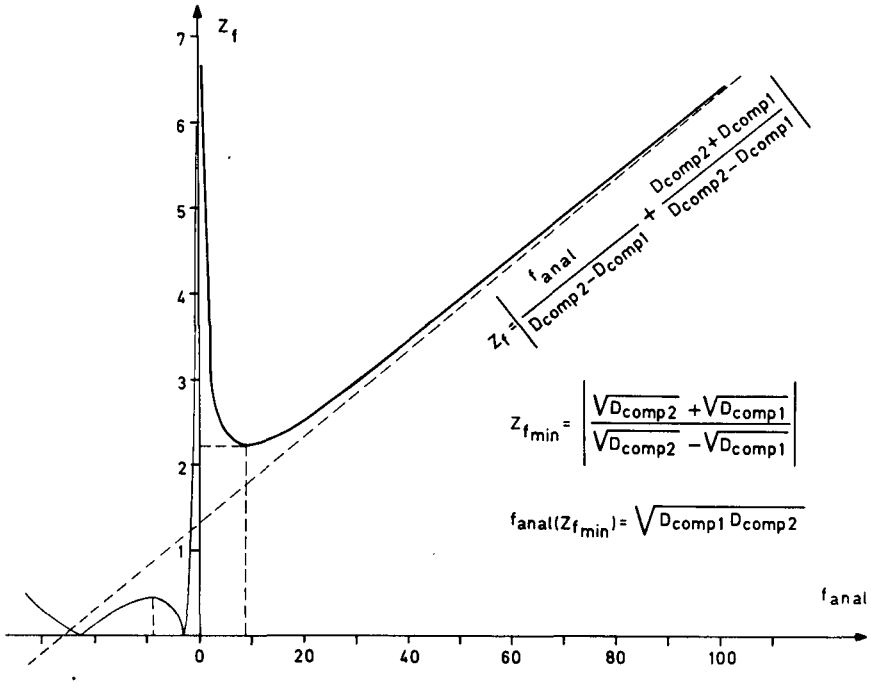

FIG. 2. Z_f as a function of f_{anal} [Eq. (22)].
$D_{comp\ 1} = 3.3$ (^{103}Ru); $D_{comp\ 2} = 23.1$ (^{97}Ru).

Equation (21), independent of f_{ref}, shows that the value of $Z_{f_{min}}$ is always larger than unity, so that Z_f can be considered to be a real error multiplication factor. If the two comparators have the same D-value it is apparent from Eqs (14) and (20) that $R = R_{(Z_{f_{min}})} = 1$, independent of the f_{ref}-value. In this case, however, the values of $Z_{f_{min}}$ and Z_f are infinite.

By combination of Eqs (15) and (18), one can set up the equation in which Z_f is given as a function of the activation parameters f_{anal}, $D_{comp\ 1}$ and $D_{comp\ 2}$:

$$Z_f = \left| \frac{(f_{anal} + D_{comp\ 1})(f_{anal} + D_{comp\ 2})}{f_{anal}(D_{comp\ 2} - D_{comp\ 1})} \right| \qquad (22)$$

From Eq. (22) it is obvious that the error multiplication factor Z_f is not influenced by f_{ref}.
The function corresponding to Eq. (22) is plotted in Fig. 2 as Z_f versus f_{anal}. Again, it should be mentioned that the curve has only a physical meaning for positive f_{anal}-values. Z_f shows a minimum value at

$$f_{anal\,(Z_{f_{min}})} = \sqrt{D_{comp\ 1}\ D_{comp\ 2}} \qquad (23)$$

this expression being the harmonic average of the D-values for the two comparator isotopes. For this value, $Z_{f_{min}}$, can be expressed identically to Eq.(21). From Eqs (21) and (22) it can be concluded that, if $D_{comp\,1} = D_{comp\,2}$, $Z_f = Z_{f_{min}} = \infty$. In case one of the comparators has a very large D-value compared to the other one, $Z_{f_{min}}$ approaches unity. Furthermore, from Eqs (21) and (22) (Fig.2) it appears that, for any value of f_{anal}, the best results are obtained:

1. if $Z_{f_{min}}$ is as low as possible. Equation (21) shows that this is performed by using two comparators with a large spreading of their D-values, i.e. the ratio $D_{comp\,2}/D_{comp\,1}$ should be very small or very large;
2. if the asymptote

$$Z_f = \left| \frac{f_{anal}}{D_{comp\,2} - D_{comp\,1}} + \frac{D_{comp\,2} + D_{comp\,1}}{D_{comp\,2} - D_{comp\,1}} \right|$$

has a small first derivative, i.e. if the value $|1/D_{comp\,2} - D_{comp\,1}|$ is small. Again, this condition required a large spreading of the D-values for both comparators.

So as to give a graphical solution for the most suitable conditions, the following reasoning can be helpful. If an error multiplication factor $Z_{f_{max}}$ is tolerated, the limits for $D_{comp\,2}$ can be calculated as a function of $D_{comp\,1}$ for the flux ratio interval that might be expected, i.e. from $f_{anal\,(min)}$ to $f_{anal\,(max)}$. These relations can be written as

$$D_{comp\,2\,(min)} = \frac{f_{anal}\,[f_{anal} + D_{comp\,1}(Z_{f_{max}} + 1)]}{f_{anal}(Z_{f_{max}} - 1) - D_{comp\,1}} \qquad (24)$$

if $D_{comp\,1} < f_{anal}(Z_{f_{max}} - 1)$ and $D_{comp\,1} < D_{comp\,2}$ and also:

$$D_{comp\,2\,(max)} = \frac{f_{anal}[-f_{anal} + D_{comp\,1}(Z_{f_{max}} - 1)]}{f_{anal}(Z_{f_{max}} + 1) + D_{comp\,1}} \qquad (25)$$

if $D_{comp\,1} > f_{anal}/Z_{f_{max}} - 1)$ and $D_{comp\,1} > D_{comp\,2}$.

From Fig. 3, where the limits of $D_{comp\,2}$ are plotted as a function of $D_{comp\,1}$ for two values of f_{anal} and for a $Z_{f_{max}} = 5$, it is apparent that there is a region (shaded area) where the combination of $D_{comp\,1}$ and $D_{comp\,2}$ may lead to a larger error multiplication factor than the tolerated $Z_{f_{max}}$. Obviously, for large $D_{comp\,1}$-values, the curve corresponding to $f_{anal\,(min)}$ is determining the unfavourable area. The reverse is true for low $D_{comp\,1}$-values.

Error on k_{anal} relative to the error on f_{anal}

The error on k_{anal}, induced from computing this factor from the f_{anal}-value with the aid of Eq.(9), can be expressed relative to the error on f_{anal}:

$$Z_k = \left| \frac{\frac{d\,k_{anal}}{k_{anal}}}{\frac{d\,f_{anal}}{f_{anal}}} \right| \tag{26}$$

Differentiating Eq.(9) gives:

$$Z_k = \left| \frac{f_{anal}(D_{st} - D_{comp})}{(f_{anal} + D_{comp})(f_{anal} + D_{st})} \right| \tag{27}$$

Figure 4, where Z_k is plotted as a function of f_{anal}, shows that the curve has a maximum value at:

$$f_{anal}(Z_{k_{max}}) = \sqrt{D_{st}\ D_{comp}} \tag{28}$$

this expression being the harmonical average of the D-values for the standard and the comparator isotope.

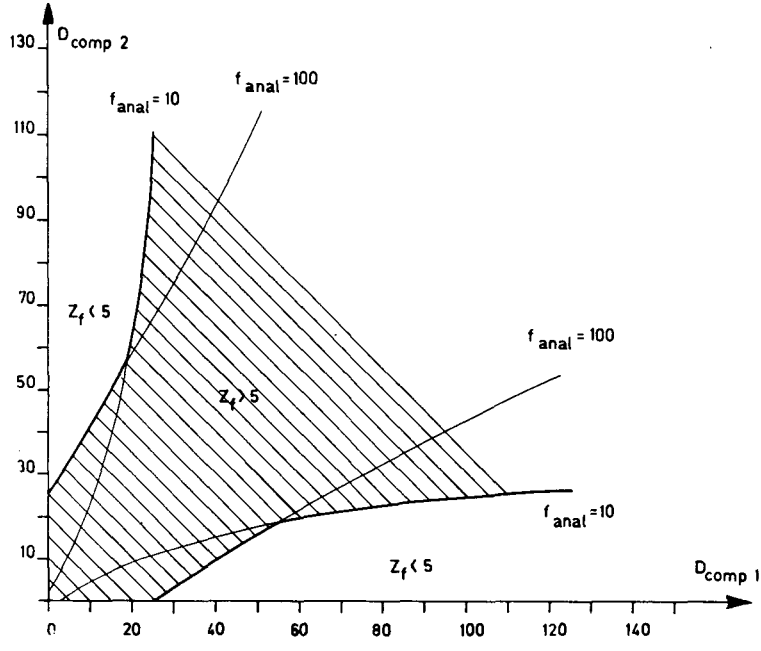

FIG.3. Working conditions for the determination of f_{anal} [Eqs (24) and (25)].

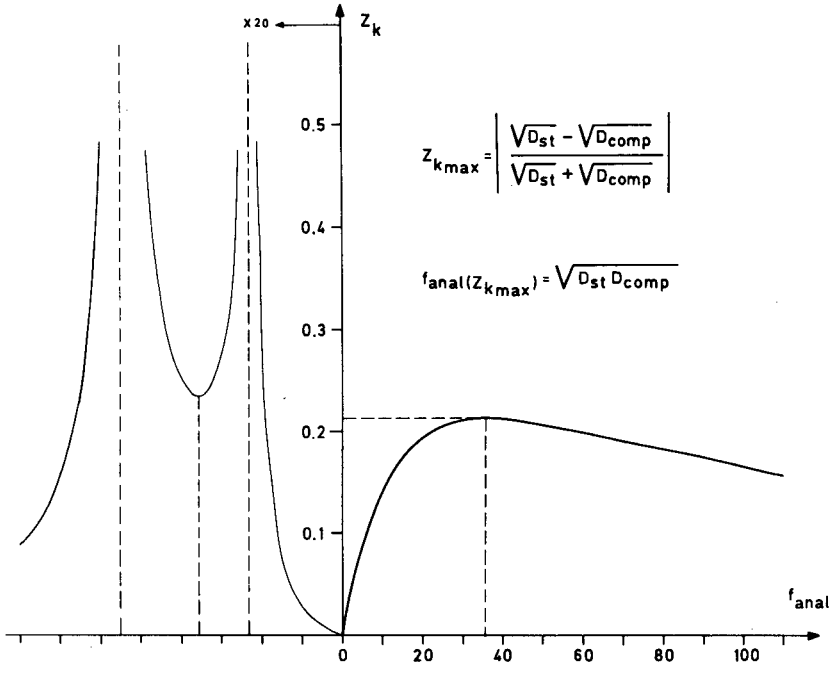

FIG.4. Z_k as a function of f_{anal} [Eq.(27)].
D_{st} = 54.9 (^{124}Sb); D_{comp} = 23.1 (^{97}Ru).

At this point $Z_{k_{max}}$ becomes:

$$Z_{k_{max}} = \left| \frac{\sqrt{D_{st}} - \sqrt{D_{comp}}}{\sqrt{D_{st}} + \sqrt{D_{comp}}} \right| \qquad (29)$$

From Eq.(29) it appears that $Z_{k_{max}}$ is always smaller than unity, so that Z_k can be considered as an error reduction factor. If $D_{st} = D_{comp}$ it is obvious that $Z_k = Z_{k_{max}} = 0$ [Eqs (27) and (29)]. This is the case in classical activation analysis.

From the above-discussed considerations it can be stated that the best results are obtained if the D-values for standard and comparator are close together, and if their harmonical average is much different from f_{anal}.

Error on k_{anal} relative to the error on R

The total error change factor Z can be defined by the combination of Eqs (17) and (26):

$$Z = Z_f Z_k \qquad (30)$$

and thus by multiplying Eqs (18) and (27) and substituting f_{anal} by its value from Eq. (15), one can write

$$Z = \left| \frac{R(f_{ref} + D_{comp\,1})(f_{ref} + D_{comp\,2})(D_{comp\,2} - D_{comp\,1})(D_{st} - D_{comp})}{\left[R(f_{ref} + D_{comp\,2})(D_{comp\,1} - D_{st}) + (f_{ref} + D_{comp\,1})(D_{st} - D_{comp\,2})\right]\left[R(f_{ref} + D_{comp\,2})(D_{comp\,1} - D_{comp}) + (f_{ref} + D_{comp\,1})(D_{comp} - D_{comp\,2})\right]} \right| \quad (31)$$

The curve Z versus R, corresponding to Eq. (31) shows two optimum values at a positive R-value of:

$$R_{(Z\,opt)} = \frac{f_{ref} + D_{comp\,1}}{f_{ref} + D_{comp\,2}} \sqrt{\frac{(D_{st} - D_{comp\,2})(D_{comp} - D_{comp\,2})}{(D_{st} - D_{comp\,1})(D_{comp} - D_{comp\,1})}} \quad (32)$$

At this point Z reaches a value of:

$$Z_{opt} = \left| \frac{(D_{comp\,2} - D_{comp\,1})(D_{st} - D_{comp})}{\sqrt{(D_{comp} - D_{comp\,2})(D_{comp\,1} - D_{st})} + \sqrt{(D_{st} - D_{comp\,2})(D_{comp\,1} - D_{comp})}} \right| \quad (33)$$

At the limits for R, given by Eq. (19), the Z-values become:

$$Z_{lim\,I} = \left| \frac{D_{st} - D_{comp}}{D_{comp\,2} - D_{comp\,1}} \right| \quad \text{and} \quad Z_{lim\,II} = \frac{D_{comp\,1}\, D_{comp\,2}}{D_{st}\, D_{comp}}\, Z_{lim\,I} \quad (34)$$

From Eqs (31), (32) and (33), it is apparent that the form and the position of the curve are highly dependent on the relative values of the four D-ratios. Figure 5, where Z is plotted as a function of R, gives an idea about some possibilities when dealing with practical circumstances. It should be mentioned that in every case D_{comp} is identical to $D_{comp\,1}$ or $D_{comp\,2}$, i.e. that the k-values are related to one of the comparators used for the determination of f_{anal}. Furthermore, it is apparent that, in all the cases under consideration, the $R_{(Z\,opt)}$-value is positioned beyond the limits for physical meaning of R, so that Z is limited by the two Z_{lim} values [Eq. (34)].

As a conclusion, it can be stated that the error change factor Z is decreasing if there is a large spreading of the D-values for both comparators, and if the D-values of the standards and the comparators, to which they are related to determine the k-factors, are close to each other.

EXPERIMENTAL

Determination of I_0/σ_{th}-values

The irradiations were performed in channels 3, 9 and 16 of the Thetis reactor (Ghent). The ratios of the thermal-to-epithermal neutron fluxes were computed from the cadmium ratio of ^{198}Au, when irradiating a 0.500% Au-Al alloy [Eqs (10) and (11)], using the value $(I_0/\sigma_{th})^{198}Au = 15.7$ [15].

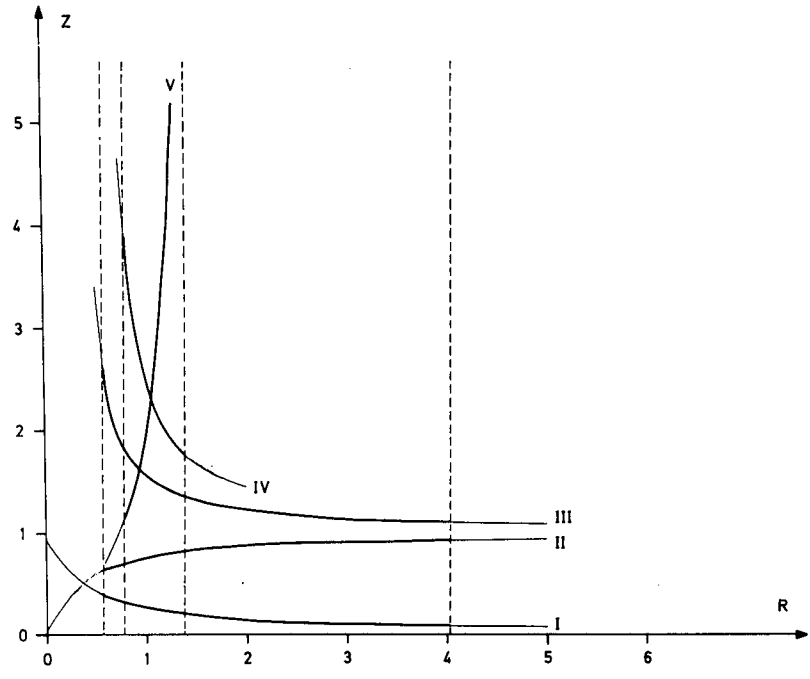

FIG. 5. Z as a function of R [Eq. (31)].

I. $D_{comp\ 1} = 3.3$ (^{103}Ru); $D_{comp\ 2} = D_{comp} = 23.1$ (^{97}Ru); $D_{st} = 15.7$ (^{198}Au); $f_{ref} = 23.8$.

II. $D_{comp\ 1} = D_{comp} = 3.3$ (^{103}Ru); $D_{comp\ 2} = 23.1$ (^{97}Ru); $D_{st} = 15.7$ (^{198}Au); $f_{ref} = 23.8$.

III. $D_{comp\ 1} = D_{comp} = 3.3$ (^{103}Ru); $D_{comp\ 2} = 23.1$ (^{97}Ru); $D_{st} = 54.9$ (^{124}Sb); $f_{ref} = 23.8$.

IV. $D_{comp\ 1} = D_{comp} = 13.0$ (^{105}Ru); $D_{comp\ 2} = 23.1$ (^{97}Ru); $D_{st} = 54.9$ (^{124}Sb); $f_{ref} = 23.8$.

V. $D_{comp\ 1} = D_{comp} = 13.0$ (^{105}Ru); $D_{comp\ 2} = 23.1$ (^{97}Ru); $D_{st} = 2.03$ (^{60}Co); $f_{ref} = 23.8$).

The following results were obtained:

$$(\phi_{th}/\phi_{epi})_3 = 23.8 \pm 0.3$$
$$(\phi_{th}/\phi_{epi})_9 = 18.3 \pm 0.2$$
$$(\phi_{th}/\phi_{epi})_{16} = 144 \pm 1$$

The errors are expressed as standard deviations on the mean (average of 20 experiments).

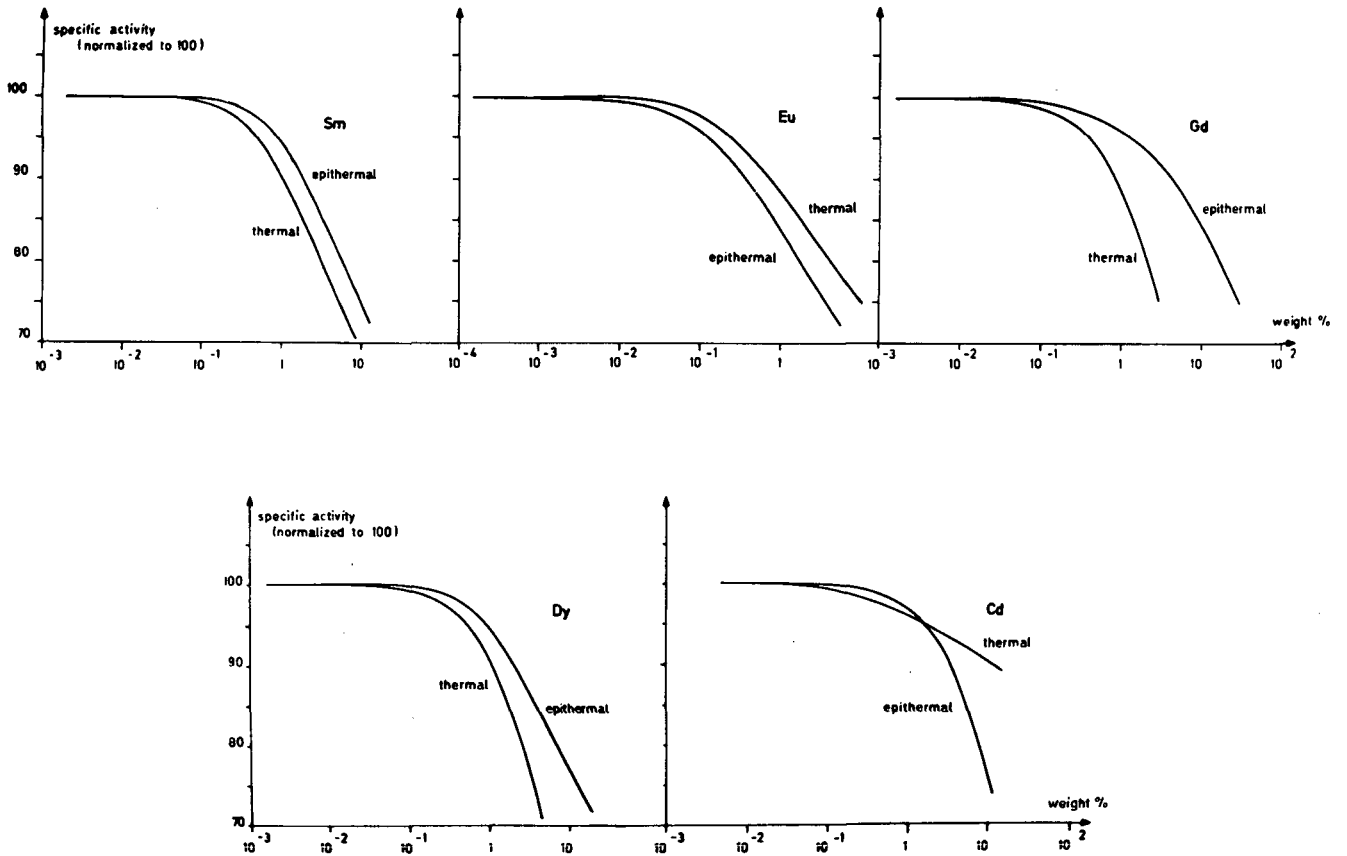

FIG.6. Thermal and epithermal self-shielding curves for Sm, Eu, Gd, Dy and Cd.

The I_0/σ_{th}-values were calculated from the cadmium ratios of the isotopes under investigation [Eqs (10) and (11)]. From Eq.(11) it is apparent that the cadmium ratio should differ significantly from unity to minimize the error on I_0/σ_{th}. As R_{Cd} is dependent on the flux ratio ϕ_{th}/ϕ_{epi}, the experimental conditions can be optimized by irradiation in a reactor channel with a suitable ϕ_{th}/ϕ_{epi}-value. In this work it was possible for almost all the (n,γ)-reactions to perform the irradiations in the three above-cited reactor channels. Normally, about 1 mg of a suitable high-purity compound of the element was irradiated with and without a cadmium cover during an appropriate time. In cases where high absorption cross-sections or absorption resonance integrals were involved, special precautions were taken to avoid self-shielding effects during irradiation. This was achieved either by mechanical mixing of a suitable compound of the element with Al_2O_3, or by spotting 100 μl of a 6N HNO_3 solution of the element on a small Whatmann-41 filter paper, followed by drying under a I.R. lamp. For Sm, Eu, Gd, Dy and Cd, all these elements having a very high absorption cross-section, dilution was performed by co-precipation with calcium oxalate, followed by conversion to the carbonate at 500°C. Even with concentrations of 10^{-3} weight per cents of the elements in the calcium carbonate, the homogeneity of the mixtures was better than 1.5% (standard deviation on the average value). Furthermore, the gravimetric yield was found to be better than 99%. Some typical self-shielding curves, obtained by irradiating mixtures with different concentrations with and without a cadmium cover, are shown in Fig.6. It is apparent that, except for Eu, a concentration as low as 10^{-2} weight per cents is sufficient to avoid thermal and epithermal self-shielding effects. In the case of Eu, the concentration should be lower than 10^{-3} weight per cents.

The activity measurements were carried out on a Ge(Li) diode (resolution = 2.25 keV, efficiency = 7.8%) or a low energy photon detector (resolution = 245 eV for the 5.9 keV Fe X-ray) coupled to a 4000 channel analyser. In these counting conditions, it was possible in most cases to determine simultaneously the I_0/σ_{th}-values for the different isotopes of the same element. When dealing with long-lived radioisotopes some activity measurements were performed on a NaI(Tl) well-type detector (3"×3") coupled to a 400-channel analyser. The identification of the radioisotopes was effected either by determining the photopeak energies [16] or by half-life control [17].

In Table I, the I_0/σ_{th}-values for 122 (n,γ)-reactions are listed. In general, one reported value is the average of six experimental results, namely two for each of the three irradiation channels mentioned. The error is expressed as the standard deviation on the mean.

From Table I it can be seen that Ru can serve as a choice for a multi-isotopic comparator element. The I_0/σ_{th}-values for the three Ru-isotopes are: 3.3 (^{103}Ru); 13.0 (^{105}Ru) and 23.1 (^{97}Ru). In Fig.7 the Z_f-values are plotted versus the flux ratio in the "analysis" reactor position for the three possible comparator combinations. Apparently, the combination ^{103}Ru + ^{97}Ru will give reliable results for most flux ratios encountered in practical work. The other combinations are only giving acceptable results when the flux ratio is not much differing from the harmonical average of the respective I_0/σ_{th}-values. It is worth mentioning that the three radioisotopes of ruthenium can be measured simultaneously on a Ge(Li) detector, by counting the following γ-rays: 215.7 keV for ^{97}Ru, 497.9 keV for ^{103}Ru and 724.3 keV for ^{105}Ru [19].

TABLE I. COMPILATION OF I_0/σ_{th}- VALUES

Produced radioisotope	Half-life	$\frac{I_o}{\sigma_{th}}$
^{20}F	11.2 s	3.9 ± 0.1
^{24}Na	15 h	0.66 ± 0.02
^{27}Mg	9.45 m	1.0 ± 0.1
^{28}Al	2.31 m	1.05 ± 0.02
^{31}Si	2.62 h	6.6 ± 0.3
^{38}Cl	37.3 m	0.49 ± 0.02
^{42}K	12.52 h	0.90 ± 0.04
^{49}Ca	8.8 m	0.82 ± 0.01
^{46}Sc	83.9 d	0.50 ± 0.04
^{51}Ti	5.8 m	31 ± 3
^{52}V	3.76 m	0.60 ± 0.02
^{51}Cr	27.8 d	0.49 ± 0.02
^{56}Mn	2.58 h	1.04 ± 0.06
^{59}Fe	45.1 d	1.4 ± 0.08
^{60}Co	5.2 y	2.03 (18)
^{65}Ni	2.56 h	0.51 ± 0.02
^{64}Cu	12.8 h	1.3 ± 0.1
^{66}Cu	5.1 min	1.2 ± 0.1
^{65}Zn	245 d	2.2 ± 0.1
^{69m}Zn	13.8 h	3.1 ±0.3
^{70}Ga	21.1 m	6.2 ± 1.0
^{72}Ga	14.3 h	2.7 ± 0.1
^{75}Ge	79 m	1.59 ± 0.06
^{77}Ge	11.3 h	12 ± 1
^{76}As	26.3 h	9.5 ± 0.2
^{75}Se	121 d	8.2 ± 0.3
^{81m}Se	56.8 m	6.0 ± 0.2
^{80}Br	17.6 m	18.5 ± 0.5
^{82}Br	35.9 h	15 ± 1
^{86}Rb	18.66 d	7.93 ± 0.43
^{86m}Rb	1 m	19.3 ± 0.5

TABLE I. (continued)

^{88}Rb	17.8 d	0.16 ± 0.01
^{85}Sr	64 d	25.8 ± 3.2
^{85m}Sr	70 m	7.9 ± 0.3
^{87m}Sr	2.84 h	5.70 ± 0.30
^{90m}Y	3.14 h	885 ± 80
^{95}Zr	65 d	7.6 ± 0.4
^{94m}Nb	6.6 m	7.3 ± 2.2
^{99}Mo	66 h	9.22 ± 0.71
^{101}Mo	14.6 m	21.2 ± 1.0
^{97}Ru	2.88 d	23.1 ± 1.0
^{103}Ru	39.8 d	3.3 ± 0.3
^{105}Ru	4.5 h	13.0 ± 0.6
^{104m}Rh	4.4 m	55 ± 2
^{109}Pd	13.46 h	15.5 ± 0.8
^{109m}Pd	4.69 m	11.9 ± 0.2
^{111}Pd	22 m	12.7 ± 1.5
^{111m}Pd	5.5 h	20 ± 1
^{108}Ag	2.42 m	3.91 ± 0.16
^{110}Ag	24.6 s	12.6 ± 0.8
^{110m}Ag	253 d	12.2 ± 0.2
^{111m}Cd	48.6 m	19.7 ± 0.9
^{115}Cd	53.5 h	11.4 ± 0.6
^{114m}In	50.0 d	27.3 ± 1.0
^{116m}In	54 m	13.6 ± 0.1
^{113}Sn	115 d	38.6 ± 1.9
^{117m}Sn	14 d	81 ± 3
^{123m}Sn	40.3 m	5.55 ± 0.13
^{125m}Sn	9.6 m	62 + 3
^{122}Sb	2.74 d	20.9 ± 0.8
^{124}Sb	60.2 d	54.9 ± 2.6
^{127}Te	9.3 h	8.9 ± 0.7
^{129}Te	69 m	7.9 ± 0.3

TABLE I. (continued)

^{131}Te	25 m	1.7 ± 0.2
^{128}I	25.0 m	15.3 ± 0.9
^{134}Cs	2.06 y	12 ± 3
^{134m}Cs	2.9 h	11 ± 2
^{131}Ba	11.7 d	25.1 ± 0.8
^{133m}Ba	38.9 h	5.6 ± 0.3
^{135m}Ba	28.7 h	151 ± 7
^{137m}Ba	2.551 m	68 ± 4
^{139}Ba	83.2 m	0.88 ± 0.04
^{140}La	40.2 h	1.28 ± 0.08
^{141}Ce	32.5 d	0.76 ± 0.04
^{143}Ce	33 h	1.10 ± 0.06
^{142}Pr	19.2 h	1.80 ± 0.15
^{147}Nd	11.06 d	2.03 ± 0.15
^{149}Nd	1.73 h	5.6 ± 0.3
^{151}Nd	12.4 m	15.8 ± 0.9
^{153}Sm	46.8 h	15.1 ± 0.7
^{155}Sm	22.3 m	4.2 ± 0.2
^{152}Eu	14 y	0.67 ± 0.03
^{152m}Eu	9.3 h	0.92 ± 0.05
^{154}Eu	7.8 y	3.81 ± 0.19
^{159}Gd	18.0 h	33.3 ± 1.6
^{160}Tb	72.4 d	16.1 ± 0.8
^{165}Dy	2.35 h	0.30 ± 0.02
^{166}Ho	26.8 h	10.1 ± 1.5
^{171}Er	7.5 h	4.2 ± 0.5
^{170}Tm	129 d	13.7 ± 0.5
^{169}Yb	31 d	7.2 ± 1.7
^{175}Yb	4.19 d	0.58 ± 0.02
^{177}Yb	1.9 h	2.4 ± 0.2
^{176m}Lu	3.69 h	30.4 ± 4.5
^{177}Lu	6.71 d	0.52 ± 0.02
^{175}Hf	70 d	0.78 ± 0.04
^{180m}Hf	5.5 h	14.4 ± 0.5
^{181}Hf	42.4 d	2.58 ± 0.13

TABLE I. (continued)

^{182}Ta	115 d	32.5 ± 3
^{182m}Ta	15.9 m	27.6 ± 1
^{185}W	75 d	12.2 ± 0.5
^{187}W	23.9 h	11.4 ± 1.3
^{186}Re	91 h	12.9 ± 0.7
^{188}Re	16.8 h	4.2 ± 0.4
^{188m}Re	18.6 m	4.4 ± 0.4
^{185}Os	94 d	0.200 ± 0.015
^{190m}Os	9.9 m	50.5 ± 5
^{191}Os	15.5 d	2.44 ± 0.04
^{191m}Os	13 h	2.40 ± 0.05
^{193}Os	31 h	2.25 ± 0.06
^{192}Ir	74.3 d	3.64 ± 0.4
^{194}Ir	17.8 h	12.6 ± 1.0
^{197}Pt	20.0 h	7.0 ± 0.2
^{199}Pt	31 m	13.0 ± 0.8
^{198}Au	2.7 d	15.7 (15)
^{197}Hg	64.1 h	0.41 ± 0.04
^{197m}Hg	23.8 h	0.39 ± 0.02
^{199m}Hg	43 m	102 ± 15
^{203}Hg	46.6 d	0.83 ± 0.03
^{205}Hg	5.5 m	1.98 ± 0.10
^{233}Th	22.2 m	9.8 ± 0.6
^{239}U	23.5 m	97 ± 8

However, it should be taken into account that the 497.9 keV photopeak of ^{103}Ru is disturbed by the 499.3 keV γ-line of the 4.5 h ^{105}Ru-isotope.

Thus to investigate the accuracy and the reproducibility of the flux ratio determinations with the aid of a Ru comparator, the relative multiple-comparator method was applied to evaluate ϕ_{th}/ϕ_{epi} in channels 9 and 11 of the Thetis reactor. Channel 3 [ϕ_{th}/ϕ_{epi} = 23.8 ± 0.3] was considered as the reference irradiation place. The flux ratios of channels 9 and 11 as determined from the cadmium ratio of ^{198}Au, were found to be 18.3 ± 0.2 and 33.2 ± 0.3, respectively.

Next, the ϕ_{th}/ϕ_{epi}-values for the two "analysis" channels were evaluated from the multiple-comparator method using the three possible comparator sets of the Ru-isotopes [Eq.(15)]. The results are summarized in Table II. Each result is the average of 16 experiments.

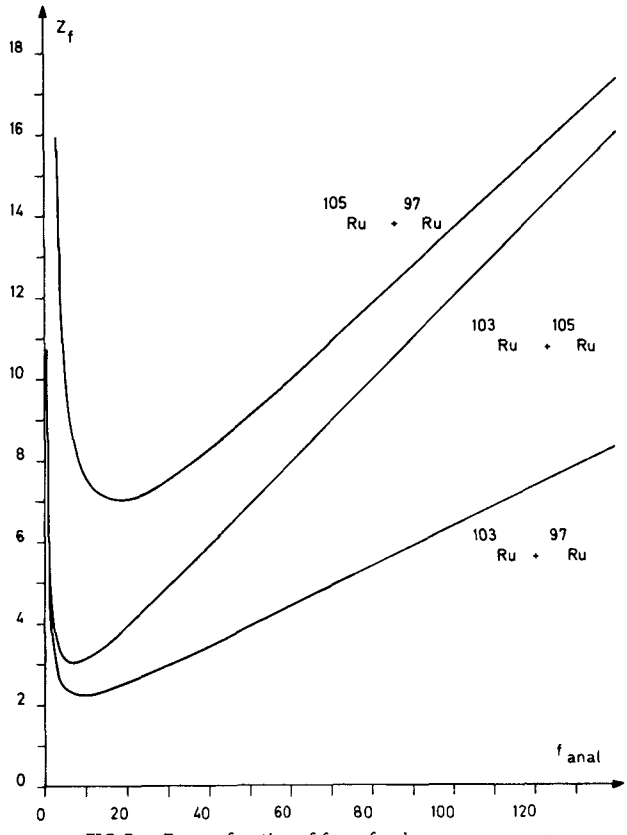

FIG. 7. Z_f as a function of f_{anal} for the comparator sets $^{105}Ru + ^{97}Ru$, $^{103}Ru + ^{105}Ru$ and $^{103}Ru + ^{97}Ru$.

TABLE II. ACCURACY OF THE DETERMINATIONS OF THE FLUX RATIO DETERMINED BY THE RELATIVE MULTIPLE-COMPARATOR METHOD

Comparator Set	ϕ_{th}/ϕ_{epi} in channel number		
	3	9	11
$^{97}Ru + ^{103}Ru$	-	17.5 ± 1.0*	31.5 ± 2.5
$^{103}Ru + ^{105}Ru$	-	19.7 ± 2.4	31.5 ± 4.1
$^{97}Ru + ^{105}Ru$	-	15.8 ± 2.7	34.9 ± 7.0
^{198}Au (R_{Cd})	23.8 ± 0.3	18.3 ± 0.2	33.2 ± 0.3

* Standard deviation on a single determination.

TABLE III. COMPARISON OF THEORETICAL AND EXPERIMENTAL STANDARD DEVIATIONS ON THE FLUX RATIO
(Standard deviations calculated on a single determination)

Channel number	Comparator Set											
	^{97}Ru - ^{103}Ru				^{103}Ru - ^{105}Ru				^{97}Ru - ^{105}Ru			
	s_R %	Z_f	$s_{f_{anal}}$ % theor.	$s_{f_{anal}}$ % exp.	s_R %	Z_f	$s_{f_{anal}}$ % theor.	$s_{f_{anal}}$ % exp	s_R %	Z_f	$s_{f_{anal}}$ % theor.	$s_{f_{anal}}$ % exp.
9	3.0	2.4	7.2	6	2.8	3.9	10.9	12	2.9	7.0	20.3	17
11	2.6	3.5	9.1	8	2.9	5.5	15.9	13	2.5	7.2	18.0	20

From Table II it is apparent that the results are more accurate when using two comparators with a large spreading of their I_0/σ_{th}-values. This is also true with respect to the reproducibility of the results. In Table III, the relative standard deviation on the flux ratios, sf_{anal} %, was calculated with the aid of Eqs (17) and (18), and compared to the experimental relative standard deviations. Apparently, the experimental and theoretical results are in good agreement.

CONCLUSIONS

From the former discussions it can be stated that the determination of the flux ratio in the "analysis" reactor position is the most important source of error in the relative multiple-comparator method. However, the accuracy and the precision can be improved by an appropriate choice of the comparator isotopes. These isotopes should preferably have a large spreading on their I_0/σ_{th}-values, with their harmonical average not too much differing from the expected flux ratio in the "analysis" reactor position. Furthermore, the total error on the final result (the specific activities of the standards), originating from applying the relative multiple-comparator method, can be markedly decreased when converting the k-values by referring the standards to the most suitable comparator. The I_0/σ_{th}-values for a standard and a comparator, to which the standard is related through the k-value, should not differ too largely, and their harmonical average should not approach the flux ratio in the "analysis" reactor position.

When applying the relative multiple-comparator method it is always possible for a known flux ratio in the reference reactor position, and for a given set of comparators and standards, to calculate the error on the final result, arising from the mathematical operations, as a function of the experimentally determined R-value. This supplies information of the precision on the standard activities in the given circumstances. Furthermore, it was found that Ru can serve as a suitable multi-isotopic comparator element. Although the comparator sets ^{97}Ru (I_0/σ_{th} = 23.1) - ^{105}Ru (I_0/σ_{th} = 13.0) and ^{103}Ru (I_0/σ_{th} = 33) - ^{105}Ru (I_0/σ_{th} = 13.0) are far from being an ideal combination, they can give reliable results when the flux ratio of the "analysis" reactor position is not too much different from the harmonical average of both I_0/σ_{th}-values.

Finally, the relative multiple-comparator method can now be applied to a large variety of multi-element analyses, as the I_0/σ_{th}-values for most (n,γ)-reactions, useful in reactor neutron activation analysis, have been determined. A comparison of the resonance integrals of infinite dilution calculated from the I_0/σ_{th} ratios and the most recently reported thermal cross-sections, with the values previously given in the literature, has been published elsewhere [20, 21].

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DISCUSSION

A.H.W. ATEN: Is anything known concerning the energy distribution of the epithermal neutron fluxes in which the resonance integrals have been determined, i.e. to what extent they deviate from a 1/E-spectrum?

F.DE CORTE: Although we did not study in detail the extent of deviations from a 1/E-spectrum we could reasonably assume that these deviations were small. We determined experimentally the I_0/σ_{th} -values for all the (n,γ) reactions under investigation by irradiation in three different channels of the Thetis reactor, obtaining a considerable spread of the thermal to epithermal flux ratios (i.e. ranging from 18.3 to 144). The results were considered as belonging to one population and the standard deviations for most of the values did not exceed 10%; they were even lower than 5% for about half of the cases.

NUCLEAR DATA OF IMPORTANCE IN EPITHERMAL NEUTRON ACTIVATION ANALYSIS

E. STEINNES
Institutt for Atomenergi, Isotope Laboratories,
Kjeller, Norway

Abstract

NUCLEAR DATA OF IMPORTANCE IN EPITHERMAL NEUTRON ACTIVATION ANALYSIS.

The use of a suitable filter material such as cadmium to facilitate selective activation with epithermal neutrons is of interest in many cases in reactor activation analysis. This enables considerable suppression of interfering activities, provided that the ratio resonance activation integral/thermal neutron activation cross-section of the target nuclide giving rise to the interfering activity is low compared to that upon which the analysis is based. This technique is primarily of interest for improving precision and sensitivity in purely instrumental activation analysis, but in some cases may also be applied to minimizing fission-product interference, or, simply, to a reduction of the matrix activity to an acceptable level in cases where major elements in the sample have high activation cross-sections. To permit estimation of the possible advantage of introducing epithermal activation in a specific case, it is necessary to know the values of thermal and epithermal activation cross-sections of the nuclides involved. Moreover, it is useful to know the energies of the main resonances contributing to the activation integral, in order to ascertain that the filter used does not interfere significantly with the resonance activation. In some cases, it may be desirable to calculate the extent of resonance neutron shielding in the sample, in which case the knowledge of resonance parameters such as partial widths for neutron and gamma emission is important. Unfortunately, in many cases the necessary data are not available from the literature, which is particularly the case for target nuclides with low isotopic abundances. In the present paper, the importance of various types of nuclear data in resonance neutron activation analysis and the availability of such data in the literature are discussed.

INTRODUCTION

In most activation analyses carried out with a nuclear reactor, the samples are subjected to irradiation with neutrons of the entire energy spectrum, in which case (n,γ)-reactions induced by thermal neutrons are usually responsible for the greater part of the induced activity. It may often be advantageous, however, to discriminate against thermal neutron activation by means of a suitable filter material such as cadmium. This will always be useful in cases where fast-neutron-induced reactions of the (n,p)-, (n,α)- or $(n,2n)$-types are concerned, but may also be of considerable advantage in the case of (n,γ)-reactions introduced by epithermal neutrons. This will be the case if the nuclide upon which the analysis is based has a high $I/\dot{\sigma}_0$-ratio compared to elements interfering in the analysis in some way.

Although the possibility of epithermal neutron activation analysis was pointed out as early as 1951 [1], the applications reported have been relatively few, considering the rapid growth of this field in general. In the author's laboratory, epithermal activation has been found to be particularly useful in the case of geological material [2 - 6]. During this work the need for reliable data for several categories of nuclear data has become evident.

UTILITY OF EPITHERMAL ACTIVATION ANALYSIS

The useful applications of epithermal activation analysis can be classified as follows:

1. Reduction of interference from elements with one or more stable isotopes with high thermal-neutron cross-sections for (n,γ)-reactions.
   (a) Improvement of precision and sensitivity in purely instrumental activation analysis [2 - 4].
   (b) Reduction of the total activity to a more suitable working level. For example, epi-cadmium activation can be utilized for the depression of 15.0 h ^{24}Na activity in geological samples where palladium is to be determined by means of 13.5 h ^{109}Pd [7].
   (c) Elimination of shielding effects caused by materials with high absorption cross-section in the thermal region but not to the same extent above 0.4 eV, such as e.g. in the determination of Au in cadmium telluride [8].

2. Reduction of interference from other nuclear reactions leading to the same nuclide as that under investigation.
   (a) Fission product interference. The reaction ^{235}U (n,f) has a low I/σ_0 ratio compared to many (n,γ)-reactions. Benefit from this fact has been taken in the determination of Mo in silicate rocks, in which case fission product ^{99}Mo is a serious problem unless epithermal activation is employed [5].
   (b) As will be discussed later, the interference from second-order nuclear reactions may in some cases be minimized if epithermal activation is employed.

The feasibility of epithermal activation using cadmium as a filter material can be expressed by means of an "advantage" factor [9], defined as follows:

$$F_a = \frac{(R_{Cd})_d}{(R_{Cd})_D}$$

where d and D denote, respectively, an interfering nuclide and the nuclide of interest in the analysis. Establishment of the advantage that can be obtained in a certain case is thus possible if the cadmium ratios of the nuclides involved are known at the irradiation site to be used. These values can be determined experimentally or can be calculated from the following equation:

$$R_{Cd} = 1 + \frac{\sigma_0 I^{Au}}{\sigma_0^{Au} I} (R_{Cd}^{Au} - 1)$$

provided that the cadmium ratio of ^{197}Au has been determined in the irradiation site and that the constants σ_0 and I (including the "1/v-tail") are known for the nuclide in question. The formula can be used provided that the nuclide follows the 1/v-law below 0.4 eV and has no appreciable resonances close to the Cd-cutoff. Even for nuclides showing some

deviation from the 1/v-law in the thermal neutron region, a useful estimation may be made using the σ_0-value.

Need for nuclear data

From the above formula it is clear that an estimation of the possible benefit of epithermal activation assumes knowledge of the σ_0- and I-values of the (n,γ)-reactions involved.

Considering the thermal-neutron cross-section, values are now available for most stable nuclides which can be transformed to a radioactive isotope by an (n,γ)-reaction. In a great number of cases, dubious values from the early days of nuclear spectroscopy have now been replaced by presumably more accurate data.

As concerns resonance activation integrals, however, the situation does not seem so satisfactory. When preparing a review paper on epithermal activation analysis of geological materials [10] a few years ago, the author found that out of 138 (n,γ)-reactions of potential interest in this connection, resonance integral values were not available in the literature for 49 of these reactions. Furthermore, for a number of reactions, only a single value or several values in mutual disagreement were available. The lack of data was evident especially for nuclides with low fractional isotopic abundance, in which case measurements involving absorption techniques would necessitate an isotopically highly enriched sample. Even in the case of nuclides for which major resonances had been well identified and resonance parameters had been determined by independent groups, there were no data for the resonance integral.

When we started our work on epithermal activation analysis of geological material in 1968, no resonance integral values were available for nuclides such as ^{26}Mg, ^{46}Ca, ^{50}Ti, ^{85}Rb, ^{84}Sr, ^{130}Ba, ^{152}Gd, and ^{158}Gd, which were all of some importance to us. Recently, there has been some emphasis on resonance integral measurements using activation techniques, and several papers have been published during the last couple of years. This trend was verified by inspection of the most recent issue of CINDA, where references to, at least, one value for the resonance integral could be found for each of the nuclides mentioned. The situation still does not seem to be satisfactory, however, and more work is needed to fill the still empty spaces and to verify values for those reactions that have so far been studied by one or two groups only.

EVALUATION OF ERRORS IN EPITHERMAL ACTIVATION ANALYSIS

There are two important sources of errors that should always be considered in reactor neutron activation analysis, namely, the decrease of induced specific activity due to neutron shielding effects, and the possible influence from interfering nuclear reactions leading to the nuclide upon which the analysis is based. The aspects of these two types of errors that apply specifically to epithermal activation and the nuclear data necessary to estimate the extent of such errors are briefly discussed in the following.

Shielding effects

The most serious shielding effects in epithermal neutron activation analysis are associated with nuclides having large resonances above the cadmium cutoff. They may be classified in two different groups, namely, those due to resonances of the element investigated, and those caused by more abundant elements in the sample possessing resonances that overlap in energy major resonances of the element to be determined. The first group is in most cases insignificant for samples of 100 mg or less if the element is present at a concentration below 0.1%, provided that its distribution in the sample is homogeneous. The second type of error may, however, be serious in many cases, such as, e.g. the determination of gold in silver [11], where the 4.9-eV resonance of ^{197}Au, which is responsible for a major part of the ^{198}Au atoms formed by epithermal activation, overlaps the 5.1 eV resonance of ^{107}Ag. To be able to decide whether errors of this kind may be significant, it is necessary to know the energies of the major resonances for the elements to be determined as well as for the major elements in the sample concerned.

As shown by Hϕgdahl [11] the interference from resonance neutron shielding in activation analysis can in some cases be fairly accurately estimated using the formulas of Chernick and Vernon [12] for the calculation of effective resonance integrals. In this case, besides the resonance energy E_r and the total cross-section σ_t at the maximum of the resonance, the resonance width Γ and the partial widths Γ_γ and Γ_n must also be known for all significant resonances.

Interfering nuclear reactions

Errors from (n,p)- and (n,α)-reactions will be more serious in epithermal activation as compared to irradiation with the whole reactor spectrum, since most reactions of this type do not occur with thermal neutrons. The knowledge of reactor fast-neutron cross-sections for this kind of reaction is, therefore, of importance. Unfortunately, a considerable number of data is still lacking here. An additional difficulty is that values determined in one reactor position need not be valid in another position with different energy distribution of the fast-neutron flux, which is particularly the case for reactions with comparatively low threshold values.

Interference from second-order reactions of the type

$$^{A-1}_{Z-1}X\,(n,\gamma)\;{}^{A}_{Z-1}X \xrightarrow{\beta^-} {}^{A}_{Z}Y\,(n,\gamma)\;{}^{A+1}_{Z}Y$$

may often be significant in the determination of element number Z, but the amount of interference is, in general, not the same in thermal and epithermal activation analysis, the difference being dependent on the I/σ_0 ratios of the nuclides

$$^{A-1}_{Z-1}X,\;{}^{A}_{Z-1}X \text{ and } {}^{A}_{Z}Y$$

The calculation of this interference [13, 14] requires knowledge of the σ_0 and I values of the three nuclides.

A similar source of interference, which has not been extensively discussed in the literature, is that of successive neutron capture which is in some cases likely to be significant when high fluxes are used:

$$ {}^{A-1}_{Z-1}X\,(n,\gamma)\,{}^{A}_{Z-1}X\,(n,\gamma)\,{}^{A+1}_{Z-1}X \xrightarrow{\beta^-} {}^{A+1}_{Z}Y $$

The major difficulty when attempting to calculate interferences of this type is probably the lack of resonance integral data for radioactive nuclides of the type ${}^{A}_{Z-1}X$ (and ${}^{A+1}_{Z-1}X$).

As already mentioned, errors due to ^{235}U fission products may in some cases be appreciably reduced since the ^{235}U (n,f) reaction has a relatively low I/σ_0 value. To know these cases and be able to calculate the interference, it is again important that good values for the resonance activation integrals are available for nuclides which upon (n,γ)-reaction lead to nuclides that are also high-abundance fission products. The chain yields of fission products which are also necessary for calculations of this kind are probably in most cases known with sufficient accuracy.

CHOICE OF FILTER MATERIAL

Cadmium filters, in most cases of thickness 0.5 - 1.0 mm, have so far been used in almost all epithermal activation work reported in the literature. To change the effective cutoff energy, however, other filter materials might well be used, separately or together with cadmium. As shown by Rossitto et al. [15], boron, or a cadmium/boron combination, could be useful if an element such as Mn with a major resonance at a relatively high energy is to be determined. Here again, knowledge of the energies of the main resonances of the elements to be determined is very useful.

If future techniques of neutron spectrometry make it feasible to irradiate samples uniformly with a virtually monoenergetic neutron beam, the selectivity of the analysis could be very much improved. This would, however, necessitate a more exact knowledge of the energies of the resonances involved than is necessary at the present state of the art.

CONCLUSION

The knowledge of thermal and resonance neutron cross-sections for (n,γ)-reactions leading to a radioactive product is of major importance in epithermal activation analysis. In some cases also thermal and resonance cross-sections for (n,γ)-reactions induced in radioactive nuclides as well as reactor fast-neutron cross-sections for (n,p)- and (n,α)-reactions must be known in order to permit calculations of interferences. For calculation of resonance shielding effects, several resonance parameters must be available. Although the situation concerning the availability of some of the abovementioned types of data has improved considerably in recent years, there still appears to be a number of "gaps" that need to be filled.

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DISCUSSION

H. JUNG: Can you give a list, or at least some examples, of isotopes for which you would like to know such parameters as resonance energies and partial widths?

E. STEINNES: Such data are of significance when resonance shielding effects are to be estimated. In our work on epithermal activation analysis we have so far been concerned almost exclusively with trace elements in geological samples, where the major elements do not represent any problem in this respect. I therefore cannot give you such a list offhand.

R. NICKS: The response of the irradiated sample depends greatly on the neutron spectrum. Thus the low-energy resonances may make the major contribution to a soft spectrum, whereas the higher resonances (perhaps unresolved) may predominate in harder spectra. Besides the fact that the latter resonances are probably known with less precision, one should in all cases know the neutron spectrum. This neutron spectrum is not easily available, and its precision might affect the precision of the analysis.

E. STEINNES: I agree. However, in our work on epithermal activation analysis we always use the relative method, employing standards for each element to be determined. In this case the actual shape of the neutron spectrum should not affect the precision of the results appreciably.

NUCLEAR DATA FOR NEUTRON METROLOGY

W.L. ZIJP
Reactor Centrum Nederland,
Petten, N.H.,
The Netherlands

Abstract

NUCLEAR DATA FOR NEUTRON METROLOGY.

For many irradiation experiments, knowledge of the neutron field characteristics, such as the neutron spectrum and the flux density distribution, is needed. For the determination of the fluence and the spectrum of neutrons incident on irradiation samples activation and fission detectors are often applied. This well-known method can yield reliable results when the reaction cross-sections and the decay schemes for the product nuclides are well-known, and absolute activities are determined with good precision. Some topics in this field are discussed, and some recommended data are presented, based on the work performed within the framework of the Euratom Working Group on Reactor Dosimetry. Some questionable points for future investigations or measurements of nuclear data for reactions of interest are also mentioned.

1. INTRODUCTION

In the last few years, the "Euratom" Working Group on Reactor Dosimetry, (abbreviated EWGRD) had several discussions on the topic of nuclear data required for neutron metrology by means of activation and fission detectors. It was stressed many times, and not only inside the Working Group, that experimenters should give full reference of nuclear data used in order to facilitate, if necessary, re-calculation by other authors when new nuclear data become available.

It was decided by the Working Group to promote the establishment of recommendations for σ_0 , $\langle\sigma\rangle$ and $T_{1/2}$ values.

The procedure originally planned comprised three steps or phases:

(a) the preparation of a list of recent data, to be sent around to all co-operating laboratories, to ask whether they had any objections to an adoption of this common set of recent data, and if so, for an explanation of these objections. This step aimed at a short-term harmonization of nuclear data in the European Community;

(b) the preparation of a list of evaluated data. At the second meeting of the IAEA Working Group, held in Vienna in April 1971, it was decided to invite the IAEA Nuclear Data Section to prepare a report on the status of the knowledge of the data relevant to neutron measurement. Just before the third meeting of this IAEA Working Group, held in November 1972 in Seattle, this status report became available [1][1]. This report deals only with high-priority reactions, and we hope that the Nuclear Data Section will soon also consider other reactions of interest;

(c) the preparation of a list of best and consistent data carefully checked by integral experiments. In this respect, one should aim at a world-wide

[1] In this paper, there are two distinct sets of references: one for references in the text and the other one for references in the tables and the figures.

TABLE I. TENTATIVE RECOMMENDED VALUES FOR THERMAL AND INTERMEDIATE NEUTRON REACTIONS (1973)

σ_0 : the 2200 m/s cross-section;
E_{Cd} : the cut-off energy, determining the lower energy limit of I_{tot} ;
I_{tot} : the total resonance integral cross-section for activation (thus including 1/v contribution);
I' : the resonance integral cross-section for activation (thus excluding the 1/v contribution), $I' = I_{tot} - I_{1/v}$;
$I_{1/v}$: the contribution to the resonance integral cross-section, arising from the 1/v part of $\sigma(E)$.

Reaction	$T_{\frac{1}{2}}$	Error	Ref.	σ_0 (in b)	Error (in b)	Ref.	E_{Cd} (in eV)	I_{tot} (in b)	Error (in b)	I' (in b)	Error (in b)	Ref.
$^{23}Na(n,\gamma)^{24}Na$	15.030 h	0.003 h	[1]	0.534	0.005	[2]	0.62			0.075	0.010	[3]
$^{30}Si(n,\gamma)^{31}Si$	2.62 h	0.01 h	[4]	0.108	0.002	[5]	0.55	0.66	0.03			[10]
$^{51}V(n,\gamma)^{52}V$	3.760 min	0.008 min	[1]	4.93	0.06	[5]	0.55			0.48	0.09	[5]
$^{55}Mn(n,\gamma)^{56}Mn$	2.587 h	0.006 h	[1]	13.23	0.10	[6]	0.55	14.06		8.46		[6]
$^{58}Fe(n,\gamma)^{59}Fe$	44.5 d	0.2 d	[1]	1.14	0.02	[5]	0.55	1.7	0.01			[10]
$^{59}Co(n,\gamma)^{60}Co$	5.272 a	0.001 a	[7]	37.34	0.09	[8]	0.55	70	6			[9]
$^{63}Cu(n,\gamma)^{64}Cu$	12.71 h	0.01 h	[1]	4.4	0.2	[5]	0.55			2.5	0.2	[5]
$^{64}Ni(n,\gamma)^{65}Ni$	2.521 h	0.005 h	[4]	1.49	0.03	[5]	0.55			0.44	0.14	[5]
$^{98}Mo(n,\gamma)^{99}Mo$	66.02 h	0.01 h	[1]	0.15	0.2	[13]	0.622			9.9	1.1	[3]
$^{109}Ag(n,\gamma)^{110m}Ag$	253 d		[2]	4.98	0.47	[11]	0.55	81.1	2.2	77.7	2.1	[11]
$^{115}In(n,\gamma)^{116m}In$	54.34 min	0.09	[1]	161	3	[5]	0.55			2710	200	[5]
$^{139}La(n,\gamma)^{140}La$	40.27 h	0.05 h	[12]	8.2	0.8	[13]	0.4	12	1			[14]
$^{151}Eu(n,\gamma)^{152m}Eu$	9.3 h		[2]	3100	400	[13]	0.63	1230	30			[15]
$^{164}Dy(n,\gamma)^{165}Dy$	2.32 h		[16]	2700	200	[13]	0.5	332	10			[17]
$^{175}Lu(n,\gamma)^{176m}Lu$	3.69 h		[16]	13.26		[15]	0.63	405	15			[15]
$^{176}Lu(n,\gamma)^{177}Lu$	6.74 d		[16]	2100	150	[13]	0.5	2400	250			[17]
$^{186}W(n,\gamma)^{187}W$	23.9 h		[16]	38	2	[13]	0.5	441	22			[18]
$^{197}Au(n,\gamma)^{198}Au$	2.6946 h	0.0010 d	[19]	98.8	0.3	[13]		1535	40	1490	40	[20]
$^{238}U(n,\gamma)^{239}U$	23.5 min		[2]	2.73	0.02	[21]		280	12			[22]
$^{235}U(n,f)$ F.P.				580.2	1.8	[23]		270	10			[23]
$^{239}Pu(n,f)$ F.P.				741.6	3.1	[23]		300	10			[23]

recommendation. Both the Euratom and the IAEA Working Group realized that a few years would be needed before such a consistent set of cross-sections could be achieved.

The present report considers some activities performed in collecting recent data and comparing available evaluated data.

A rather extensive list of evaluated integral cross-sections, averaged in a fission neutron spectrum, was published last year by Fabry [2]. The IAEA report and the Fabry report both give an excellent and up-to-date review of the state of art of neutron metrology for reactor radiation measurements.

2. THERMAL AND INTERMEDIATE NEUTRON REACTIONS

In 1971, a sub-group of the Euratom Working Group prepared a tentative recommendation for nuclear data for reactor neutron measurements [3]. An up-dated list with data for thermal and intermediate neutron reactions is shown in Table I.

3. INTEGRAL MEASUREMENTS IN A FISSION NEUTRON SPECTRUM

The Fabry report is a very valuable contribution to our knowledge of the integral cross-sections for a fission neutron spectrum. The report considers experimental cross-section ratios as determined by several authors in 1) more or less pure fission neutron spectra (e. g. realized with a ^{235}U converter in cavity), or in 2) reactor neutron spectra in so far as reasonable experimental and/or theoretical evidence existed that deviations from a fission neutron spectrum were without influence or could be corrected for.

All experimental data have been assigned weights in determining average values for the cross-section ratios. As a reference reaction the ^{235}U (n, f) reaction was chosen, for which the most probable value of the average cross-section lies between about 1200 and 1300 mb. For his evaluation study, Fabry accepted a cross-section for the reference reaction in a pure fission spectrum equal to 1250 ± 70 mb. The absolute error in this figure has not been combined to the uncertainties of the reactions under consideration.

The procedure chosen gave an appreciable weight to the cross-section ratio $\langle \sigma(^{235}U) \rangle^f / \langle \sigma(^{238}U) \rangle^f$; it was assigned a value of 3.81 ± 0.15. The results of Fabry's evaluations are mentioned in Table II.

4. CALCULATIONS BASED ON DIFFERENTIAL CROSS-SECTION DATA

This study comprises also calculated cross-sections derived from evaluated differential data. The energy range considered here is 0 to 20 MeV. Evaluated cross-section data sets have been supplied by the CCDN (Centre de Compilation des Données Nucléaires), which collected and distributed evaluated data from the Nuclear Data Libraries in the United Kingdom, the United States of America, and the Federal Republic of Germany (UKDL, ENDF/B and KEDAK, respectively).

TABLE II. TENTATIVE RECOMMENDED VALUES FOR FAST-NEUTRON THRESHOLD REACTIONS (1973)
All cross-section values refer to a fission neutron spectrum and are relative to the average cross-section value of 1250 ± 70 mb for the reference reaction $^{235}U(n,f)$.

Reaction	Half-life	Ref.	Effective threshold (in MeV)	<σ> (in mb)	Ref.
$^{24}Mg(n,p)^{24}Na$	15.030 ± 0.003 h	[1]		1.53 ± 0.03	[24]
$^{27}Al(n,p)^{27}Mg$	9.46 min	[2]	4.5	4.0 ± 0.4	[24]
$^{27}Al(n,\alpha)^{24}Na$	15.030 ± 0.003 h	[1]	7.1	0.725 ± 0.02	[24]
$^{31}P(n,p)^{31}Si$	2.62 h	[2]	2.4	36 ± 2	[24]
$^{32}S(n,p)^{32}P$	14.29 ± 0.03 d	[12]	2.7	69 ± 2	[24]
$^{46}Ti(n,p)^{46}Sc$	84 d	[2]	3.8	12.3 ± 0.5	[24]
$^{47}Ti(n,p)^{47}Sc$	3.35 d	[2]	2.2	20 ± 2	[24]
$^{48}Ti(n,p)^{48}Sc$	44.1 h	[2]	7.6	0.315 ± 0.02	[24]
$^{55}Mn(n,2n)^{54}Mn$	312.5 ± 0.5 d	[12]	11.5	0.253 ± 0.01	[24]
$^{54}Fe(n,p)^{54}Mn$	312.5 ± 0.5 d	[12]	3.3	82.5 ± 2	[24]
$^{56}Fe(n,p)^{56}Mn$	2.587 ± 0.006 h	[1]	6.1	1.07 ± 0.06	[24]
$^{58}Ni(n,p)^{58}Co$	71.3 ± 0.2 d	[12]	2.8	113 ± 2.5[a]	[24]
$^{58}Ni(n,p)^{58m}Co$	9.15 h	[2]		35.4 ± 1.0	[24]
$^{58}Ni(n,\alpha)^{55}Fe$	2.60 a	[2]		4.95	[25]
$^{59}Co(n,\alpha)^{56}Mn$	2.587 ± 0.006 h	[1]	6.8	0.156 ± 0.06	[24]
$^{63}Cu(n,\alpha)^{60}Co$	5.272 ± 0.001 a	[7]	6.7	0.50 ± 0.05	[24]
$^{63}Cu(n,2n)^{62}Cu$	9.76 min	[2]	12.4	0.124 ± 0.09	[24]
$^{65}Cu(n,2n)^{64}Cu$	12.71 ± 0.01 h	[1]	11.2	0.31	[26]
$^{64}Zn(n,p)^{64}Cu$	12.71 ± 0.01 h	[1]		31 ± 1.5	[24]
$^{92}Mo(n,p)^{92m}Nb$	10.2 d	[2]		7.0 ± 0.4	[24]
$^{93}Nb(n,n')^{93m}Nb$	11.4 ± 0.9 a	[27]		97 ± 35	[25]
$^{93}Nb(n,2n)^{92m}Nb$	10.2 d	[2]	10.2	0.47 ± 0.03	[24]
$^{103}Rh(n,n')^{103m}Rh$	56.116 ± 0.009 min	[28]	0.7	560 ± 7	[29]
$^{115}In(n,n')^{115m}In$	4.5 h	[2]	1.4	188 ± 4	[24]
$^{127}I(n,2n)^{126}I$	12.8 d	[2]	11.5	1.09 ± 0.05	[24]
$^{232}Th(n,f)$ F.P.			1.4	70.2 ± 13.2	[30]
$^{238}U(n,f)$ F.P.			1.4	328 ± 10	[24]
$^{237}Np(n,f)$ F.P.			0.7	1289 ± 87	[30]

[a] Included the formation of ^{58}Co, from ^{58m}Co, produced by the reaction $^{58}Ni(n,p)^{58m}Co$.

Also some recent sets of cross-section data from other sources have been included.

With the aid of some computer programs developed in our centre we obtained the following results for each cross-section set:

(a) a graph of the cross-section data points as a function of the energy;
(b) a list of the group cross-section data points in fixed energy groups;
(c) the cross-section averaged with three representations of the fission neutron spectrum;
(d) the effective threshold energy determined by a least-squares method.

The method of calculating the effective threshold energy (T) and the effective cross-section (S) for a neutron spectrum is based on the principle that the step function should give the same response in the neutron spectrum as the actual cross-section, and that there is a best fit between ideal and actual response curve, using a least-squares adjustment [3].

The following three analytical expressions for the fission neutron spectrum of ^{235}U have been used in our calculations. They have been normalized to an integral value of unity. In the following expressions, E denotes the neutron energy, expressed in MeV.

(a) the formula proposed by Watt [4]

$$\chi_1(E) = 0.48395 \exp(-E), \sinh\sqrt{2E}$$

(b) the formula proposed by Cranberg, Frye et al. [5]

$$\chi_2(E) = 0.45274 \exp(-E/0.965) \sinh\sqrt{2.29E}$$

(c) the formula proposed by Leachman [6]

$$\chi_3(E) = 0.76985 \exp(-0.775E)\sqrt{E}$$

A discussion of the representations of the prompt fission neutron spectrum has recently been published [7].

All cross-section values from the evaluated data files have been interpreted as point values belonging to a smooth $\sigma(E)$-function. Detailed results of our study showing the graphs and all numerical results are available as laboratory report [8].

In Figs 1 and 2, the shape and differences between the fission spectra are shown. The results of our calculations for the fission neutron averaged cross-section values applying the various formulae are presented in Table III.

The average cross-section values calculated with our computer program for the reactions ^{235}U (n, f) and ^{237}Np (n, f) might be slightly too low, because of the application of interval widths equal to 100 keV, which is relative large, especially for the lowest energy group.

The calculated effective threshold values using a Watt fission spectrum are presented in Table II.

The values of T obtained with the various fission neutron spectrum formulae do not show differences larger than 0.1 MeV.

Figure 3 shows two examples of the calculated effective threshold (T) and the effective cross-section (S).

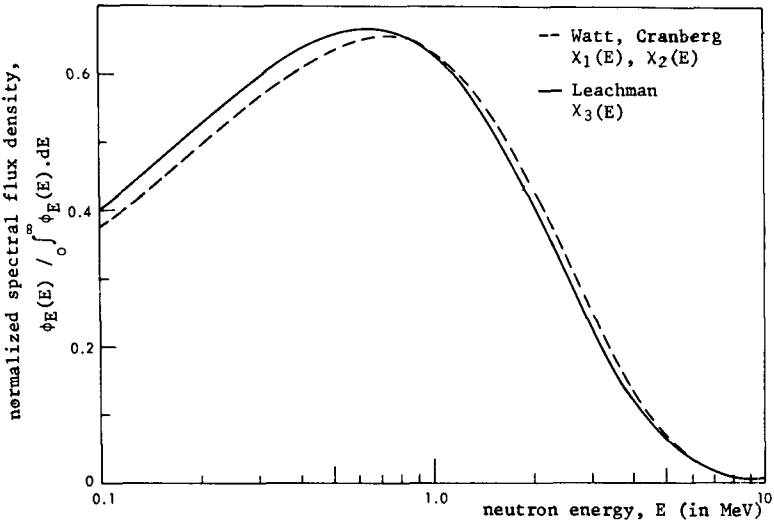

FIG. 1. The fission neutron spectrum according to Watt, Cranberg and Leachman (reported from Ref. [40]).

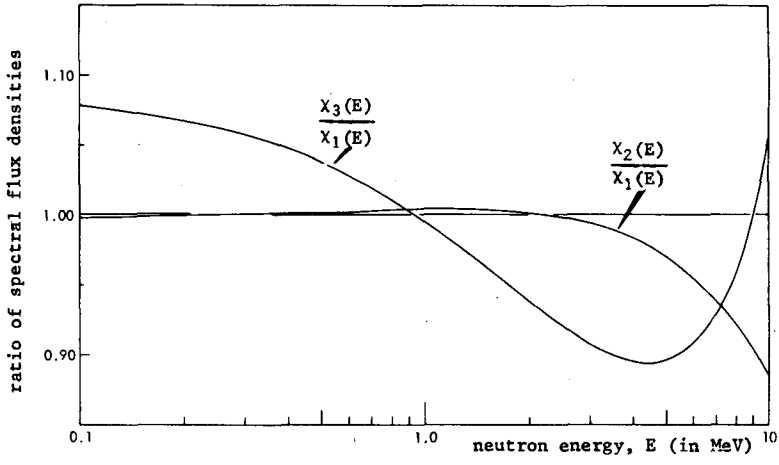

FIG. 2. The ratio of spectral flux densities derived from different spectrum expressions (reproduced from Ref. [40]). $X_1(E)$ = Watt formula; $X_2(E)$ = Cranberg formula; $X_3(E)$ = Leachman formula.

TABLE III. CALCULATIONS OF AVERAGE FISSION NEUTRON CROSS-SECTIONS

All cross-section values are expressed in mb = 0.1 fm^2; $\chi_1(E)$, $\chi_2(E)$, and $\chi_3(E)$ denote the fission neutron spectrum representations by Watt, Cranberg and Leachman. For footnotes see below last part of this table.

Reaction	Code	Library	Reference	Date[c]	Average cross-sections obtained with[a]		
					$\chi_1(E)$	$\chi_2(E)$	$\chi_3(E)$
$^{24}Mg(n,p)$	11202411024	UKDL	DFN 225	6508	1.446	1.329	1.483
$^{27}Al(n,p)$	11302712027		Simons [31]	7005	3.839	3.679	3.678
	31302712027	UKDL	DFN 35E	6402	3.877	3.716	3.718
	41302712027	UKDL	DFN 226	6508	4.114	3.948	3.919
	51302712027	ENDF/B	MAT 1135	7201	4.163		3.991
$^{27}Al(n,\alpha)$	11302711024	UKDL	DFN 226	6508	0.6143	0.5615	0.6378
	21302711024	UKDL	DFN 95	7004	0.6922	0.6332	0.7172
	31302711024	UKDL	DFN 96	7004	0.6769	0.6207	0.7062
	41302711024	UKDL	DFN 35E	6402	0.5905	0.5394	0.6137
	51302711024	ENDF/B	MAT 1015	7006	0.6869	0.6274	0.7136
	61302711024	KEDAK	B 69	6901	0.5905	0.5394	0.6137
	71302711024		Simons [31]	7005	0.6630	0.6061	0.6887
	81302711024		Kanda [32]	7202	0.6922	0.6332	0.7172
	91302711024	ENDF/B	MAT 1135	7201	0.6281	0.5737	0.6528
$^{31}P(n,p)$	1503114031	UKDL	DFN 228	6508	34.45	33.92	32.50
$^{32}S(n,p)$	11603215032	UKDL	DFN 229	6508	60.81	59.61	57.31
	21603215032	UKDL	DFN 97	7004	67.28		63.52
$^{46}Ti(n,p)$	52204621046		Simons [31]	7005	11.28	10.88	10.74
	62204621046	UKDL	DFN 912	7201	11.66	11.22	11.14
$^{47}Ti(n,p)$	52204721047		Simons [31]	7005	17.19	16.96	16.26
$^{48}Ti(n,p)$	52204821048		Simons [31]	7005	0.2365	0.2140	0.2519
$^{54}Fe(n,p)$	12605425054	UKDL	DFN 63	6802	70.35	68.60	66.43
	22605425054	UKDL	DFN 233	6508	96.28	94.40	91.14
	42605425054		Bresesti [33]	7008	76.23	74.35	71.87
	52605425054		Simons [31]	7005	76.28	74.46	72.08
	62605425054		Paulsen [34]	7109	72.50	70.78	68.42
	72605425054		Reed [35]	6609	73.88	72.12	69.65
$^{55}Mn(n,2n)$	12505525055	ENDF/B	MAT 1019	6706	0.1543		0.2036

TABLE III (cont.)

Reaction	Code	Library	Reference	Date [c]	Average cross-sections obtained with [a]		
					$\chi_1(E)$	$\chi_2(E)$	$\chi_3(E)$
$^{56}Fe(n,p)$	12605625056	UKDL	DFN 62	6802	1.027	0.9558	1.029
	22605625056	UKDL	DFN 234	6508	0.9625	0.8985	0.9577
	32605625056	UKDL	DFN 98	7004	1.120	1.046	1.116
	52605625056		Simons [31]	7005	1.084	1.010	1.084
	62605625056		Fabry [36]	7006	1.028	0.9594	1.026
	72605625056		Kanda [32]	7202	1.120	1.046	1.116
$^{58}Ni(n,p)$	12805827058	UKDL	DFN 236	6508	113.6	111.5	107.3
	32805827058	UKDL	DFN 909	7201	110.1	108.0	104.1
	42805827058		Bresesti [33]	7008	102.5	100.6	96.88
	52805827058		Simons [31]	7005	102.2	100.2	96.59
	62805827058		Fabry [36]	7006	111.4	109.2	105.3
	72805827058		Paulsen [34]	7109	106.5	104.5	100.8
	82805827058		Reed [35]	6609	109.8	107.6	103.6
$^{59}Co(n,\alpha)$	12705925056	ENDF/B	MAT 1118	7201	0.1512	0.1389	0.1555
	52705925056		Simons [31]	7005	0.1465	0.1348	0.1504
$^{63}Cu(n,\alpha)$	12906327060	UKDL	DFN 250A	6712	0.3064	0.2822	0.3127
	42906327060	ENDF/B	MAT 1085	6812	0.3009	0.2768	0.3077
	52906327060		Simons [31]	7005	0.3548	0.3268	0.3621
$^{63}Cu(n,2n)$	12906329062		Simons [31]	7005	0.08588	0.07036	0.1198
	22906329062		Kanda [32]	7202	0.07032	0.05749	0.09859
	32906329062	UKDL	DFN 250A	6712	0.07989	0.06543	0.1115
	42906329062	UKDL	DFN 237A	6508	0.08330	0.06805	0.1171
	52906329062	ENDF/B	MAT 1085	6812	0.09014		0.1234
	62906329062	UKDL	DFN 99	7004	0.07032	0.05749	0.09859
$^{65}Cu(n,2n)$	12906529064		Kanda [32]	7202	0.2855	0.2401	0.3678
	22906529064	ENDF/B	MAT 1086	6812	0.2955		
	32906529064	UKDL	DFN 100	7004	0.2856	0.2400	0.3687
	42906529064	UKDL	DFN 251A	6802	0.2995		0.3812
$^{93}Nb(n,2n)$	14109341092	ENDF/B	MAT 1164	6708	0.6854	0.5861	0.8410
$^{103}Rh(n,n')$	14510345103	UKDL	DFN 94A	7004	722.6	720.0	704.2
$^{115}In(n,n')$	14911549115	UKDL	DFN 239	6508	174.3	173.8	167.6
	24911549115		Pauw [37]	7104	205.6	204.8	197.9

TABLE III (cont.)

Reaction	Code	Library	Reference	Date [c]	Average cross-sections obtained with [a]		
					$\chi_1(E)$	$\chi_2(E)$	$\chi_3(E)$
$^{115}In(n,n')$ (cont.)	54911549115		Simons [31]	7005	184.6	183.8	177.3
	64911549115		Fabry [36]	7006	187.6	186.9	180.6
$^{127}I(n,2n)$	15312753126	UKDL	DFN 240	6508	0.6854	0.5861	0.8410
$^{232}Th(n,f)$	19023256140	UKDL	DFN 242	6508	71.58	70.91	68.57
	29023256140	UKDL	DFN 22A	6402	75.67	74.87	72.50
	39023256140	ENDF/B	MAT 1117	6705	71.31	70.64	68.34
	59023256140		Davey [38]	66	69.81	69.13	66.83
$^{235}U(n,f)$ [b]	19223556140	ENDF/B	MAT 1102	7001	1202	1202	1202
	29223556140	KEDAK	B 168	6602	1249	1249	1247
	39223556140	UKDL	DFN 271D	7201	1212	1212	1212
	49223556140	UKDL	DFN 66A	7004	1222	1222	1221
	59223556140		Simons [31]	7005	1208	1208	1209
	69223556140		Fabry [36]	7006	1206	1206	1206
	79223556140		Greene [39]	7201	1202	1202	1202
	89223556140	ENDF/B	MAT 1157	7201			
$^{238}U(n,f)$	19223856140	UKDL	DFN 76D	6709	308.6	306.4	295.6
	29223856140	UKDL	DFN 401D	6802	300.1	298.0	287.6
	39223856140	ENDF/B	MAT 1103	7006	285.7	283.7	273.9
	49223856140	ENDF/B	MAT 1158	7103	300.4	298.3	288.0
	59223856140		Simons [31]	7005	286.5	284.4	274.7
	69223856140		Fabry [36]	7006	300.4	298.5	288.7
	89233856140	UKDL	DFN 272A	7201	284.1	282.1	272.4
$^{237}Np(n,f)$ [b]	19323756140	UKDL	DFN 61	6802	1352	1350	1328
	29323756140	ENDF/B	MAT 1048	6905	1360	1358	1334
	59323756140		Simons [31]	7005	1321	1319	1297

[a] χ_1: formula proposed by Watt.
χ_2: formula proposed by Cranberg, Frye et al.
χ_3: formula proposed by Leachman.

[b] For the reactions $^{235}U(n,f)$ and $^{237}Np(n,f)$, our calculated values for the average cross-section might be slightly too low due to the interval width of 100 keV, which width is relatively large especially for the lowest energy group.

[c] The calendar dates are the issue dates of the reports in which the values have been published, or in the case of unpublished values, the calendar dates are the dates when the files have been released.

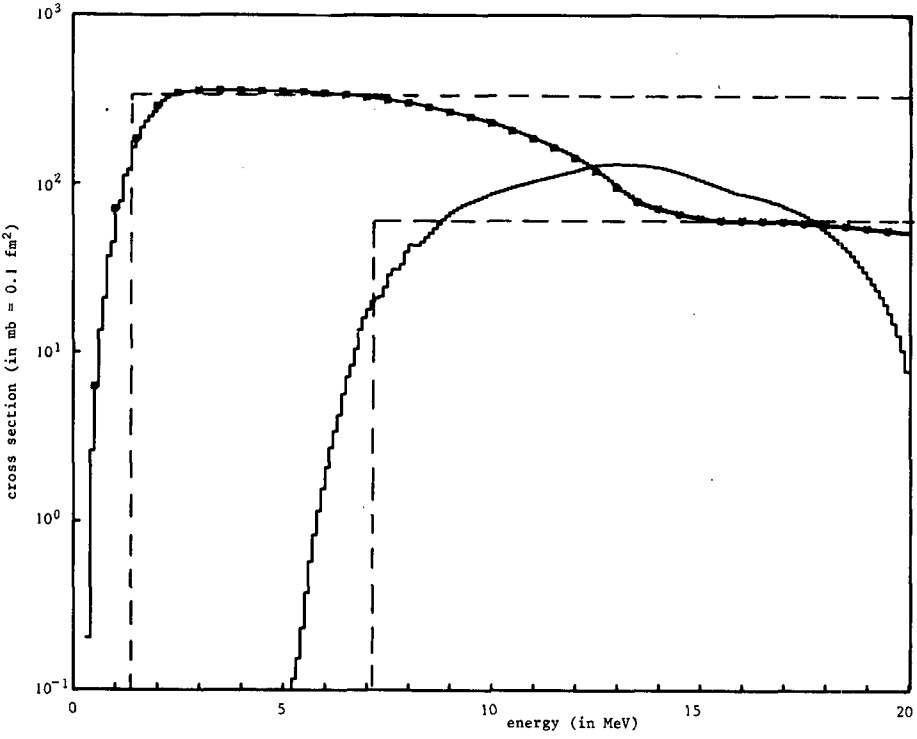

FIG. 3. Effective cross-section compared with actual cross-section curve.

—●— $^{115}In(n,n')^{115m}In$; S = 329.1 mb; T = 1.4 MeV
——— $^{27}Al(n,\alpha)^{24}Na$; S = 57.85 mb; T = 7.1 MeV

5. COMPARISON OF MEASURED OR CALCULATED AVERAGE CROSS-SECTIONS

In Table IV, we compare the evaluated experimental values, as reported by Fabry, and some calculations from evaluated differential data. In the comparison, we consider the well-known evaluated data set of Simons and McElroy [9] supplemented by less recent data from the early SAND-II library. From this comparison, a series of large discrepancies becomes apparent. One of the most urgent problems to be solved is the average cross-section for the reaction $^{63}Cu(n,\alpha)^{60}Co$. Is the observed inconsistency due to a "sub-threshold" contribution to the reaction, to a Co-impurity of the target material, to inaccurate measurements, or to some other systematic error?

The nuclear data for routine thermal and fast flux density measurements are in general quite sufficient, but for spectrum determinations we need integral data which are known to within 5%, not only for the routine reactions but also for other reactions, because of their different energy response. The inaccuracy of the group cross-sections can, however, be somewhat larger (see Appendix for some explanatory remarks).

TABLE IV. COMPARISON OF EVALUATED AVERAGE CROSS-SECTIONS FOR A FISSION NEUTRON SPECTRUM (1973)
Cross-section values refer to a Watt spectrum and are expressed in mb (= 0.1 fm^2)

Reaction	Integral experimental value (Fabry report [24])	Integral calculated value (SAND-II library)	Discrepancy (order of magnitude)
$^{63}Cu(n,\gamma)^{64}Cu$	10.1 ± 1.5	10.9 a	8%
$^{115}In(n,\gamma)^{116m}In$	146 ± 5	-	-
$^{197}Au(n,\gamma)^{198}Au$	88 ± 4.5	82.8 a	6%
$^{24}Mg(n,p)^{24}Na$	1.53 ± 0.03	1.50 d	2%
$^{27}Al(n,p)^{27}Mg$	4.0 ± 0.4	3.84 b	4%
$^{27}Al(n,\alpha)^{24}Na$	0.725 ± 0.02	0.663 a	10%
$^{31}P(n,p)^{31}Si$	36 ± 2	33.0 c	10%
$^{32}S(n,p)^{32}P$	69 ± 2	60.9 c	13%
$^{46}Ti(n,p)^{46}Sc$	12.3 ± 0.5	11.3 a	10%
$^{47}Ti(n,p)^{47}Sc$	20 ± 2	17.2 a	15%
$^{48}Ti(n,p)^{48}Sc$	0.315 ± 0.02	0.236 a	> 25%
$^{55}Mn(n,2n)^{54}Mn$	0.253 ± 0.01	0.154 f	> 25%
$^{54}Fe(n,p)^{54}Mn$	82.5 ± 2	76.3 a	8%
$^{56}Fe(n,p)^{56}Mn$	1.07 ± 0.06	1.08 b	1%
$^{58}Ni(n,p)^{58}Co$	113 ± 2.5 g	102.0 a	10%
$^{59}Co(n,\alpha)^{56}Mn$	0.156 ± 0.06	0.147 a	6%
$^{63}Cu(n,\alpha)^{60}Co$	0.50 ± 0.05	0.356 a	> 25%
$^{63}Cu(n,2n)^{62}Cu$	0.124 ± 0.09	0.0859 b	> 25%
$^{64}Zn(n,p)^{64}Cu$	31 ± 1.5	38.0 d,e	20%
$^{92}Mo(n,p)^{92m}Nb$	7.0 ± 0.4	-	-
$^{93}Nb(n,2n)^{92m}Nb$	0.47 ± 0.03	0.685 f	> 25%
$^{115}In(n,n')^{115m}In$	188 ± 4	184.6 b	2%
$^{127}I(n,2n)^{126}I$	1.09 ± 0.05	0.687 d	> 25%
$^{232}Th(n,f)$ F.P.	83 ± 3.5	71.3 d,i	15%
$^{235}U(n,f)$ F.P.	1250 ± 70 h	1230 a	2%
$^{237}Np(n,f)$ F.P.	1370 ± 75	1293 a,i	6%
$^{238}U(n,f)$ F.P.	328 ± 10	287.0 a,k	15%
$^{239}Pu(n,f)$ F.P.	1859 ± 60	1762 a	6%

a Values taken from BNWL-1312 (1970), see Ref.[33]; this data set is claimed to be a consistent set.
b Values calculated by author (1972), based on data from Ref.[33].
c Values published by McElroy (1969), see Ref.[32].
d Values from SAND-II report by McElroy et al. (1967), see Ref.[31].
e A value of 47 mb is mentioned by McElroy (1972), see Ref.[34].
f Reaction data not present in SAND-II library; value taken from ENDF/B.
g The Fabry report assigns the value of 35.4 ± 1.0 mb to the reaction $^{58}Ni(n,p)^{58m}Co$.
h This value is the reference value.
i The evaluation by Bak (1971) [30] gives 70.2 mb.
j The evaluation by Bak (1971) [30] gives 1289 mb.
k The evaluation by Bak (1971) [30] gives 285 mb.

6. NEUTRON SPECTRUM DETERMINATION

A well-known method of determining the neutron spectrum in an experiment position in a reactor is the multiple-foil activation method. The neutron spectrum is then determined from the measured activities of a set of detectors, irradiated simultaneously at the same location. For this method, a good knowledge of the cross-section curves is required, and also high precision and accuracy in the absolute foil activities. The method consists of unfolding a set of integral equations of the following type:

$$\alpha_i = \int_0^\infty \sigma_i(E)\,\phi_E(E)\,dE$$

where α_i is the experimental saturation activity per target atom of the i-th detector, $\sigma_i(E)$ the cross-section of the i-th detector, and $\phi_E(E)$ the unknown spectral distribution of neutron flux density.

The most promising computer programs for neutron spectrum unfolding are at present SPECTRA [10, 11] and SAND-II [12, 13]. These programs apply a number of energy groups which is much larger than the number of activation reactions. These codes need, therefore, extra physical spectrum information as input, and this information will, to some extent, have an influence on the final output spectrum.

In practice, it turns out that a consistent and well-known set of cross-section values is needed to obtain a physically acceptable and smooth calculated spectrum. Inconsistent data or inaccurate activity data will result in a series of peaks and valleys. This is in particular clear for resonance detectors, where unbalanced detector data give rise to "inverse" resonance structure in the neutron spectrum.

The procedure is sometimes so sensitive to showing unrealistic structure that it can be used to trace inconsistencies.

Since (n, γ)-reactions have their main response in the thermal energy and, to a lesser extent, in the intermediate energy region below 1 keV, and since (n, p), (n, α) and (n, f) are normally threshold reactions with a threshold above 1 MeV, there are often gaps in the response in the region between say 0.01 MeV and 1 MeV (see Fig. 4). To a certain extent, (n, n')-reactions might be used to partly fill in the gaps, but these reactions have a series of problems:

poor knowledge of the cross-sections;
not very well established decay schemes (half-lives and gamma abundances;
difficulty of measuring absolutely low-energy gamma-rays and X-rays (i.e. from ^{103m}Rh and ^{93m}Nb).

The low effective threshold makes the (n, n')-reactions very attractive for neutron spectrum measurements. The problems related to these reactions are discussed in the next section.

7. THE (n, n')-REACTIONS

7.1. The ^{93}Nb (n, n')^{93m} Nb-reaction

The nuclide ^{93m}Nb produced by inelastic neutron scattering, goes to the ground state by a gamma transition with an energy of 28 keV. Since

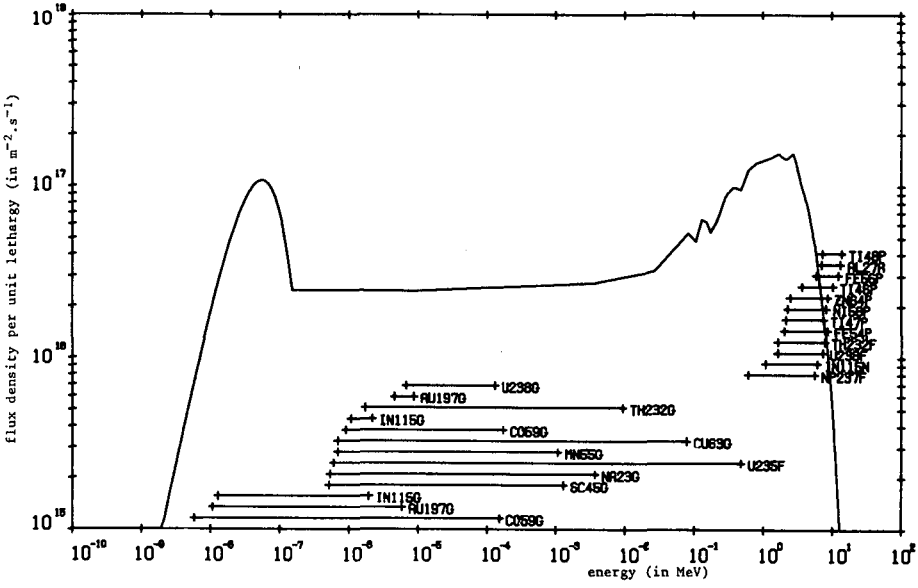

FIG. 4. Representative HFR neutron spectrum in experiment position E5. The horizontal bars denote the energy regions corresponding to 90% detector response.

the effective threshold is, therefore, also of the order of 28 keV, this reaction might be of extreme importance for fast neutron fluence determinations for irradiation damage purpose.

The status report of the IAEA Nuclear Data Section [1] shows that there exists a remarkable discrepancy in the cross-section values below 3 MeV and that there is an appreciable lack of knowledge of the differential cross-section shape for larger energies.

Recently, Hegedüs [14] proposed a histogram curve covering the entire energy region up to 15 MeV. Hegedüs also gave a fission spectrum average value of 97 mb for this cross-section. The status report also mentions another difficulty with using the ^{93}Nb(n, n') reaction as a fluence monitor: the discrepancy in experimental determinations of the half-life of the isomeric state. Measurements before 1954 give about 4 years for the half-life, and later measurements about 12 years or even 16 years (recent communication from Dr. Lloret from CEAN Grenoble). Hegedüs recommends 11.4 ± 0.9 year.

The status report concludes as follows: At the present time, the multi-group cross-sections of Hegedüs should be used as they are the only values spanning the entire energy region, and they are not in disagreement with the other measurements below 3 MeV. However, the group data need confirmation before the reliability of the data can be assured.

7.2. The ^{103}Rh(n, n') ^{103m}Rh-reaction

This reaction is very interesting for fast flux density determinations, because of its low effective threshold (about 0.7 MeV), its relatively large

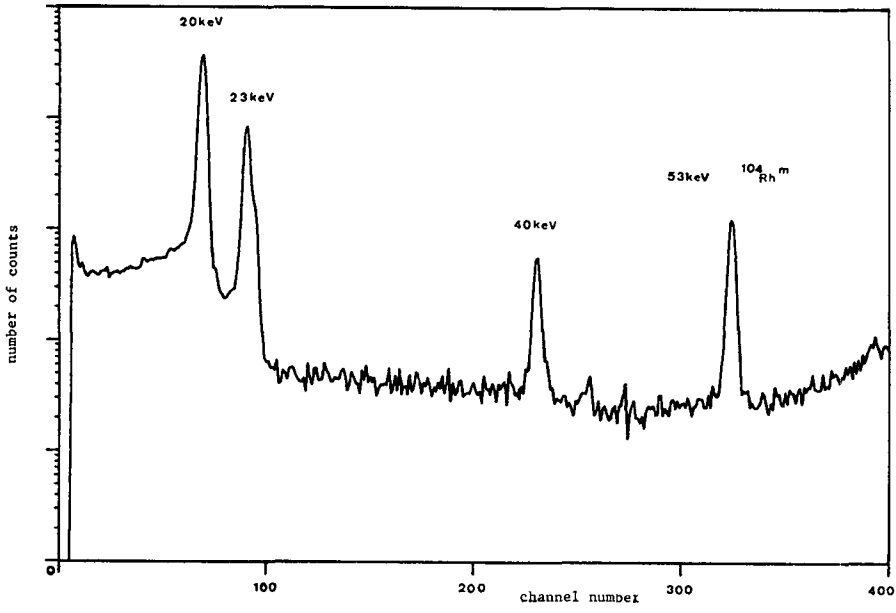

FIG. 5. Photon spectrum of an irradiated rhodium foil.

activation cross-section, and its appropriate half-life (56 min). The energy of the isomeric transition is 40 keV.

With respect to the differential cross-section curve, one has the situation that there are large discrepancies between the results from the different authors (for a review, see Ref. [1]).

There are also wide discrepancies in measured fission-spectrum averaged activation cross-sections. There are very old values near 1100 mb. More recent values give 716 ± 40, 558 ± 32 and 403 ± 40 mb. Review [1] concludes that the uncertainty of between 20 to 40% in the differential data make this reaction unsuitable for use at this time.

The cross-section problem is partly related to difficulties encountered in the absolute activity determination of ^{103m}Rh sources. The 40-keV gamma-ray energy of the isomeric transition shows a very large internal conversion (very near to 100%). The internal conversion leads to the production of Rh K X-rays, with energies of about 20 keV, which have an appreciably larger abundance than the 40 keV radiation.

An illustration of the photon spectrum as measured with a low-energy Ge(Li) detector is given in Fig. 5.

For absolute activity measurements, one has to calibrate the detector efficiency by means of a series of low-energy photon radiations with known abundance emitted by calibrated reference sources (e.g. ^{57}Co (14.4 keV) and ^{241}Am (26.348 keV) from an IAEA radionuclide set). For accurate measurements, one should like to have well known decay schemes for all nuclides used in detector calibration. Moreover, one should like to apply more photon emitters with low energy from available reference sources.

Another problem is related to the purity of the sample. The metal sheet which is obtained commercially will show many radioactive impurities after irradiation. The photoelectric absorption of photons from the radioactive impurities also caused a contribution of K X-rays, which had an influence on the 20-keV peak on which the activity measurement was based. One should, therefore, perform an accurate analysis of the decay curve for this peak, but because of the spread in the half-life of ^{103m}Rh, an unambiguous analysis is difficult.

7.3. The ^{115}In(n, n')^{115m}In-reaction

The isomeric transition of the product nuclide has an energy of about 335 keV. The effective threshold for a fission spectrum, equal to about 1.4 MeV, and the half-life of the product nuclide, equal to 4.50 h, make this reaction very useful for fast flux density determination.

The available evaluated differential cross-section curves [1] show a deviation of about 5% in the average cross-section.

Moreover, it turns out that the available values for the abundance of the 335-keV gamma radiation (46 and 50%) show a relative discrepancy of 8%. Regrettably, the combination of both discrepancies makes this reaction less suitable for accurate fast-neutron measurements.

8. LONG-TERM FLUENCE DETECTORS

In some cases, one wishes to determine the neutron fluence incident on a reactor material during a very long irradiation period. One may think of determining the neutron fluence incident on reactor constructional materials during an extended period of reactor operation. For purposes of determining the radiation damage, e.g. to the reactor pressure vessel, one should like to apply long-term neutron fluence detectors over a period of 10 years or more (e.g. to predict the economic life-time of reactors). The usefulness of a fluence detector as a radiation damage detector is greater since the detector response is more linear and corrections for irradiation history become smaller.

There are only a few techniques which can be considered for use in materials surveillance programs for power reactors. Activation techniques have the advantage of simplicity, the limitations being in the choice of suitable reactions. One might also think of formation of ^{137}Cs by fission of ^{235}U, ^{239}Pu, ^{238}U, ^{232}Th and ^{237}Np. Mass-spectrometric techniques such as the helium accumulation technique for the absolute determination of transmutation products can, in principle, give very satisfactory results although the equipment is less common.

With respect to the activation detectors we have listed some relevant data for reactions yielding product nuclides with a half-life longer than 10 years (see little table on next page).
The cross-sections for thermal burn-up and the average fission neutron cross-section are inaccurate.

The nuclide ^{40}K is a β-emitter (89% β^- with E = 1.314 MeV; EC 11% with gamma rays of 1.460 MeV). One often prefers, however, gamma counting. Moreover, the half-life of ^{40}K is so large that for practical purposes the induced specific activity will, in general, be too small.

Reaction	Half-life (in a)	Radiative capture cross-section		Average cross-section for fission neutrons (in b)
		Target nuclide (in b)	Product nuclide (in b)	
$^{40}Ca(n,p)^{40}K$	1.27×10^{9}	22	70	≈ 0.047
$^{93}Nb(n,\gamma)^{94}Nb$	20×10^{3}	1.00	15	-
$^{94}Mo(n,p)^{94}Nb$	20×10^{3}	?	15	≈ 0.00083
$^{154}Gd(n,p)^{154}Eu$	10	≈ 23	1500	$\approx 2 \times 10^{-5}$

The reaction $^{154}Gd(n,p)^{154}Eu$ seems less favourable from the results listed above. Moreover, this reaction has the disadvantage that it may be perturbed by the side reaction $^{153}Ru(n,\gamma)^{154}Eu$, which has a cross-section of about 390 b; this reaction occurs in europium impurities in the gadolinium detector. A few ppm of Eu in Gd may already perturb the response (both Gd and Eu are lanthanides!)

The nuclide ^{94}Nb emits β- and γ-radiation (nearly 100% β^- with E = 0.49 MeV; two gamma-rays: 0.702 MeV (100%) and 0.871 MeV (100%)). From the nuclear-data point of view the reactions $^{93}Nb(n,\gamma)^{94}Nb$ and $^{94}Mo(n,p)^{94}Nb$ seem to be the most suitable and promising ones.

There is, at present, no practical experience available. Moreover, the cross-section data are not very well established. The value of 0.83 mb for the average fission neutron cross-section of the $^{94}Mo(n,p)$ reaction is based on an old theoretical estimate [15]. It should be mentioned that the target material should be extremely pure. A very small amount of niobium impurity in molybdenum will perturb the response.

The conclusion is that the reactions $^{93}Nb(n,\gamma)$ and $^{94}Mo(n,p)$ should be investigated in more detail, not only with respect to material purity and absolute measurements, but also with respect to cross-section data.

9. FURTHER REMARKS ON NON-NEUTRON DATA

Nearly always the activities induced in activation and fission foil detectors are determined with gamma-ray spectrometers with NaI(Tl) scintillation detectors or with Ge (Li) semi-conductor detector.

For accurate absolute activity determinations one has to know the decay scheme, and especially the gamma abundances of dominant gamma transitions. Especially, since high-resolution Ge (Li) spectrometers have become available, interest in gamma-ray data has increased.

Very valuable and extensive data on gamma-ray energies and abundances have recently been reported by Meixner [16]. Only for low energies, where X-radiation might be of importance, one has to rely on less recent tabulations of X-ray energies (e. g. the Table of Isotopes [17] for the K X-rays, and Crouthamel [18] for L X-rays).

The accuracy of absolute activity determinations is now sometimes determined by the knowledge of the decay scheme of the product nuclide formed in activation detectors. It is reasonable to ask for well-established

and evaluated decay schemes of many activation detector products. As example one could mention: ^{27}Mg, ^{64}Cu, ^{110}Ag, ^{116m}In, ^{152}Eu, ^{165}Dy, ^{176}Lu, ^{182}Ta and ^{187}W.

Similarly justified is the great interest in having well-established fission product yields for some reference spectra, e. g. for ^{131}I, ^{132}I, ^{140}Ba and ^{99}Mo. Even for ^{235}U and ^{239}Pu, variations are serious, sometimes between 5 and 10% (corresponding to an uncertainty of 5%). For ^{99}Mo from ^{239}Pu the uncertainty seems to be even larger, while for fission detectors like ^{232}Th and ^{237}Np the situation is much worse.

10. CONCLUSIONS

In the last few years, one has been able to observe that there is an increasing need for better and more reliable neutron-spectrum data. Advanced computer codes such as SAND-II which are able to unfold the responses from activation and fission detectors, demand consistent cross-section data sets and accurate activity measurements which, in turn, require well-known decay schemes (half-lives and gamma abundances).

There still remains an appreciable number of discrepancies between evaluated measured average cross-sections and integral values derived from evaluated differential-cross-section data (see Table IV).

11. GENERAL CONCLUSIONS OF THE EURATOM WORKING GROUP ON REACTOR DOSIMETRY (EWGRD)

At its 32nd meeting in Rome, September 1972, the EWGRD devoted an essential part of its time to the problem of improvement of detector cross-sections. Some conclusions may be summarized as follows:

1. The problem of cross-sections for the detectors used in reactor dosimetry (irradiation experiments, shield assessment, reactor performance studies) is a very critical one and may appreciably influence the development of competitive nuclear power. This fact has recently been recognized by many organizations, and a certain amount of systematic work has been started, notably in the USA and by the IAEA.

2. The EWGRD considers of primary importance that the improvement in these cross-sections should be paralleled by an effort to arrive at normalized values to be recommended for use in the different laboratories so that reactor experiments, and in particular irradiation experiments, can be directly compared and exchanged, increasing the amount of available information and reducing duplication of efforts. The acceptance of normalized values on as wide a basis as possible, and the internal consistency of such values, are considered by the EWGRD to be potentially more important than the improvement of single cross-sections.

3. In this process of normalization, the role of the IAEA (and especially of its Nuclear Data Section) is essential; the EWGRD acknowledged the important effort already done by the Agency in this field.

4. One important aspect of the need for standard values is the necessity for the free and unrestricted circulation of detector cross-sections (evaluated differential files, adjusted fine group or multi-group sets, integral values), much in the same way as has been done for "standard cross-sections".

5. In the normalization effort, one should first assume a reference data set and use it in the interpretation of integral results. A part of this reference data set could e. g. be the Detector Tape of the ENDF/B now being produced in the USA.

6. New evaluations of detector cross-sections not available in the available data tapes should be encouraged; the results should be compiled in a data file format allowing an easy exchange.

7. Integral experiments in well-known (reference) neutron spectra and in other reactor spectra should be encouraged for the intercomparison of detector cross-sections, for the check of internal consistency, for the identification of discrepancies in shape or in normalization factor of the reference data. The accepted reference data set should be used, solely or in addition to different evaluations, in the interpretation of the integral experiments.

8. Users of these cross-sections should at all stages contact the original evaluators of the cross-sections so that a prompt feedback is provided and improvements are sure to be included in successive evaluations. In the countries participating in the European Community, at least for the moment, the appointed sub-group could collect and co-ordinate the results of these integral evaluations.

9. Integral experiments generally require unfolding codes for their interpretation. The introduction of the reference data into the unfolding codes is no trivial endeavour, and prompt solution of this problem should be encouraged.

10. In a second step, a new intercomparison of integral detectors in various reactor spectra (as already accomplished some years ago by the EWGRD) could be organized in co-operation with the IAEA.

11. By a co-ordination of efforts among countries of the European Community, the IAEA and the other main contributing countries or organizations, one should reach a revised evaluation for detector cross-sections, which would constitute the first standard to be recommended for general use. It seems reasonable to assume that such a stage could be reached in approximately two years from now. The recommended values could then be up-dated periodically in a systematic way by co-ordinated efforts among the various evaluation centres.

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APPENDIX

RELATION BETWEEN VARIANCE OF RESPONSE INTEGRAL AND VARIANCES OF INDIVIDUAL CROSS-SECTION MEASUREMENTS

How precise should points on a cross-section curve be measured in order to yield a given precision of the response integral?

One can imagine that there is an influence of the number of points of the curve, of the spacing between these points, and of the weighting spectral distribution.

The response integral α can be defined as

$$\alpha = \frac{A}{N} = \int \sigma(E)\, \phi_E(E)\, dE = \int \sigma(u)\, \phi_u(u)\, du \qquad (A1)$$

where A is the disintegration rate at saturation, when the flux density is constant in time, N is the number of target atoms, $\sigma(E)$ or $\sigma(u)$ is the cross-section as function of energy or lethargy, and $\phi_E(E)$ or $\phi_u(u)$ is the flux density per unit energy or per unit lethargy, respectively.

For multi-group applications we can write

$$\alpha = \sum_{i=1}^{n} f_i \sigma_i \qquad (A2)$$

where f_i represents $\phi_{E,i}(E)\Delta E_i$ or $\phi_{u,i}(u)\Delta u_i$, the fraction of the spectral distribution in the energy (or lethargy) range under consideration, and i represents the serial number of one of the neighbouring intervals, corresponding to the i-th interval.

According to the law of propagation of errors one has

$$\mathrm{var}\,\alpha = \sum_{i=1}^{n} f_i^2\, \mathrm{var}\,\sigma_i + 2 \sum_{i \neq j} r_{ij}\, f_i f_j \sqrt{\mathrm{var}\,\sigma_i\, \mathrm{var}\,\sigma_j} \qquad (A3)$$

In general, the correlation coefficients will have different signs and different sizes, and we may expect that often in practical cases the second term on the right-hand side is negligible compared to the first term.

We further assume that, on the average, each interval gives the same contribution to the variance in α. This leads to the relation:

$$\mathrm{var}\,\sigma_i \approx \frac{\mathrm{var}\,\alpha}{n f_i^2} \qquad (A4)$$

Here we have to remember that f_i represents a group flux density.

Let us consider the case that all group intervals are equal. If we deal with energy intervals, then $f_i = \phi_{E,i}\, \Delta E$. The number of group intervals is $n = (E_{max} - E_{min})/\Delta E$.

We now may write

$$\mathrm{var}\,\sigma_i \approx \frac{\mathrm{var}\,\alpha}{(E_{max} - E_{min})\, \Delta E\, (\phi_{E,i})^2} \qquad (A5)$$

Since the quantity $\phi_{E,i}$ does not depend on the magnitude of ΔE, we see that the variance is inversely proportional to ΔE, the interval width, and thus direct proportional to n, the number of intervals. Introducing the average cross-section $\langle \sigma \rangle$ by the relation

$$\langle \sigma \rangle = \frac{\Sigma f_i \cdot \sigma_i}{\Sigma f_i} = \frac{\alpha}{\Sigma f_i} \tag{A6}$$

we obtain

$$\frac{\text{var}\,\sigma_i}{\langle \sigma \rangle^2} = \frac{\text{var}\,\alpha}{n\,\alpha^2\,(f_i / \Sigma f_i)^2} \tag{A7}$$

This relation expresses the condition for the variance in group cross-section values, when one has an a-priori knowledge of:

the accuracy to be reached for the total response,
the number of groups contributing to the total response,
the relative shape of the weighting spectral distribution.

The above relation can be used if an acceptable group structure is present so that large variations of σ_i or f_i do not occur inside a group, and if only those groups are considered which contribute significantly to the response integral α.

The latter condition may, e. g. be expressed by the condition that the group contribution $f_i \sigma_i$ is less than the standard deviation of the average contribution to α.

$$f_i \sigma_i \geq \sqrt{(\text{var}\,\alpha / n)} \tag{A 8}$$

Combination of Eqs (A8) and (A4) leads to the relation

$$\frac{\text{var}\,\sigma_i}{\sigma_i^2} < 1 \tag{A9}$$

which implies that for the least contributing group the relative standard deviation never exceeds 100%.

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DISCUSSION

R. NICKS: I should like to make two observations. First, the use of threshold detectors for fast spectrum measurements is somewhat unsatisfactory as the determination of the activation rates leading to the explicit spectrum requires the use of a trial spectrum as an input, and it is not at all certain that the resulting calculated spectrum is independent of the input spectrum. Moreover, I think that the method is inadequate for showing dips (even broad dips) in the spectrum. Threshold detectors do, however, have their value when one is comparing measured activation rates with calculated ones, as a means of checking calculation codes, for instance shielding codes.

Second, as far as the sandwich technique is concerned, this is a cumbersome and sometimes an imprecise procedure (the error of the different activity is composed of the errors of the single foil measurements!). Moreover, if the detector presents more than one resonance, you also need a trial spectrum to determine the contributions of the single resonances.

W.L. ZIJP: The best procedure for determining a neutron spectrum with activation detectors is to cover the whole range, the fast neutron range with special detectors and the low-energy part, with thermal and resonance detectors. One should irradiate a whole set of detectors at the same time and at the same location if possible, so that the whole range is covered. The sandwich technique, I agree, is a fairly special technique but, if you apply it very carefully, you can obtain specific, localized information, although only at the site of the main resonance. In general, however, we want to have the whole spectrum. For this, with the computer technique now available, one gets a trial input spectrum and then calculates the activities for the set of detectors. By comparing the calculated activities and the measured activities one can then modify the neutron spectrum.

L. HJÄRNE: In view of the persistent discrepancies between fission-spectrum-averaged differential and integral cross-section data, which of the two kinds of data do you, in general, personally trust and use most?

W.L. ZIJP: In the present circumstances, I cannot give a soundly reasoned answer to your question. Personally, I have a slight preference for evaluated experimental integral values, for which consistency of a whole set is claimed.

DISTRIBUTION SPATIALE DES FLUX NEUTRONIQUES ET CONSTANTES NUCLEAIRES

Application à l'analyse par activation

B. VORSATZ, É. ZEMPLÉN-PAPP, E. KELEN
Institut central de recherches physiques,
Budapest, Hongrie

Abstract–Résumé

NEUTRON-FLUX SPATIAL DISTRIBUTION AND NUCLEAR CONSTANTS: APPLICATION TO ACTIVATION ANALYSIS.

Separating the interfering activities produced by neutron generator activation is in any case a complicated task, especially when this cannot be done by gamma spectroscopy. The neutron generator itself produces neutrons by a nuclear reaction; several reactions are used for this purpose, but most often the $^{3}T(d, n)^{4}He$ nuclear reaction is used. The products of this nuclear reaction are alpha particles, which are absorbed in the parts of the neutron generator, and "fast neutrons" of 14.7 MeV energy, which leave the generator and therefore can be utilized for activation. For a given irradiation system, neutrons are produced with different energy, depending on the effect of the surroundings. For a given activation reaction, the spatial energy distribution of the irradiating neutrons, as well as the energy dependence of the activation cross-section of the produced nuclear reaction, therefore, provide a means of distinguishing the activities according to their origin.

This principle can be utilized for simultaneous and non-destructive determination of silicon and aluminium contents, and thus has particular significance in the analysis of mineral samples. An example for the application of the method is given.

DISTRIBUTION SPATIALE DES FLUX NEUTRONIQUES ET CONSTANTES NUCLEAIRES: APPLICATION A L'ANALYSE PAR ACTIVATION.

L'activation d'échantillons à l'aide d'un générateur de neutrons peut donner naissance à des activités parasites difficiles à distinguer, en particulier lorsque la spectroscopie gamma n'est pas applicable. Le générateur lui-même produit des neutrons par réaction nucléaire. Diverses réactions sont utilisées à cette fin, mais la plus fréquente est la réaction $^{3}T(d, n)^{4}He$. Cette réaction donne naissance à des particules alpha, qui sont absorbées par les composants du générateur, et des neutrons rapides de 14,7 MeV qui quittent le générateur et peuvent ainsi servir à l'activation. Dans une installation d'irradiation, les neutrons produits ont différentes énergies, compte tenu des effets du milieu. Pour une réaction d'activation donnée, la distribution spatiale en énergie des neutrons incidents et la section efficace d'activation en fonction de l'énergie offrent donc un moyen de discriminer les activités selon leur origine.

Ce principe peut être appliqué à la détermination simultanée et non destructive des teneurs en silicium et en aluminium, et revêt une importance particulière pour l'analyse d'échantillons minéraux. Un exemple d'application de la méthode est donné.

Le principe de la méthode d'analyse par activation consiste tout d'abord à radioactiver l'élément à doser, puis à déterminer cette radioactivité. L'échantillon contenant une certaine quantité de l'élément à doser est irradié, le plus souvent au moyen de neutrons, et des radionucléides apparaissent, suivant les propriétés nucléaires des isotopes de l'élément soumis à l'analyse.

La sensibilité de la méthode dépend de l'intensité du flux de particules, de la section efficace d'activation de l'élément considéré, de la période du radionucléide formé, du type de rayonnement émis par celui-ci et, enfin, de

la présence d'éléments susceptibles de fournir, avec des sections efficaces élevées, des radionucléides interférants.

S'il existe une interférence d'une telle origine, et si le radionucléide n'a pas une période plus courte que le radionucléide produit de l'élément à doser, il sera parfois possible de les distinguer en exploitant la différence entre les rayonnements émis.

En appliquant la méthode non destructive, cette discrimination peut être faite en mesurant les énergies, les périodes et les différents types de rayonnement mentionnés. Mais la discrimination effectuée de cette façon est plus ou moins limitée. L'irradiation de différents isotopes stables peut produire les mêmes radionucléides, comme par exemple l'irradiation du fluor par des neutrons rapides, où le radioisotope ^{16}N se forme par réaction nucléaire (n, α), de même que par réaction $^{16}O(n, p)^{16}N$. C'est pourquoi, lors du dosage de l'oxygène au moyen de l'analyse par activation avec les neutrons de 14 MeV, il est impossible d'éliminer l'activité parasite du fluor par la mesure de l'activité.

Ainsi il y a des cas dans lesquels il n'existe aucune possibilité de séparer les différents radionucléides ou d'éliminer des activités parasites au moyen de la mesure, la séparation n'utilisant que les données caractéristiques des radionucléides produits.

Mais il existe une autre possibilité de supprimer les perturbations mentionnées, en agissant sur l'activation pendant l'irradiation. Pour cela il est nécessaire de connaître les constantes nucléaires des réactions en question.

On sait que pendant l'activation d'un échantillon, l'activité augmente en même temps que le flux de neutrons, et cela d'autant plus que la section efficace est plus grande. Il existe encore une constante nucléaire assez importante, c'est l'énergie de seuil de la réaction nucléaire. La valeur de la section efficace dépend aussi de l'énergie des particules incidentes, et la fonction qui exprime cette corrélation est aussi une donnée très importante pour l'analyse par activation.

Les sections efficaces peuvent en général être calculées à l'aide des lois connues, mais cette manière d'obtenir leur valeurs est assez imprécise et ne peut pas être la base d'une méthode de discrimination.

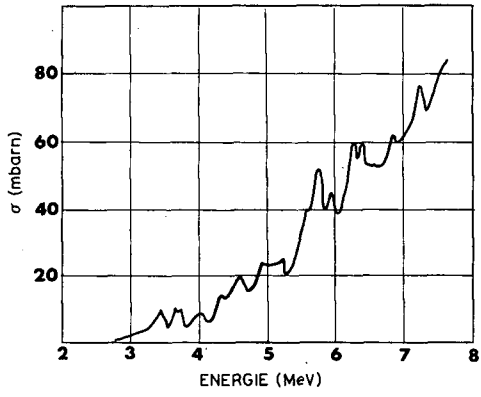

FIG. 1. Section efficace de la réaction $^{27}Al(n, p)^{27}Mg$ en fonction de l'énergie des neutrons.

La figure 1 montre la section efficace σ de la réaction $^{27}Al(n, p)^{27}Mg$ en fonction de l'énergie des neutrons. La figure 2 montre les valeurs des sections efficaces de $^{19}F(n, \alpha)^{16}N$ et de $^{16}O(n, p)^{16}N$.

On trouvera dans le tableau I quelques réactions et leurs constantes nucléaires. Toutes les valeurs des sections efficaces figurant dans ce tableau sont des valeurs mesurées.

Selon les données du tableau I, l'isotope stable ^{16}O irradié avec des neutrons d'énergie inférieure à 10 MeV doit donner une faible activité de ^{16}N, par contre l'isotope stable ^{19}F doit être plus fortement activé. La

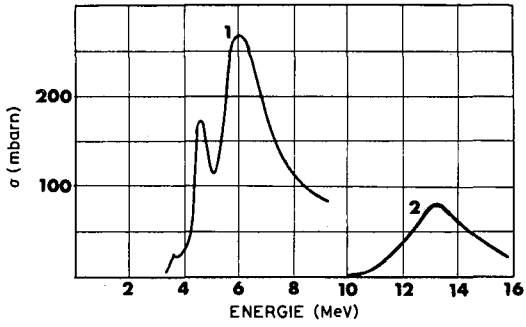

FIG. 2. Section efficace des réactions $^{19}F(n, \alpha)^{16}N$ (courbe 1) et $^{16}O(n, p)^{16}N$ (courbe 2) en fonction de l'énergie des neutrons.

TABLEAU I. CONSTANTES NUCLEAIRES DE QUELQUES REACTIONS (Tiré de [1])

Réaction nucléaire	Section efficace à 14 MeV (mbarn)	Energie de seuil approx. (MeV)	Période de l'isotope produit	Energie du rayonnement γ (MeV)
$^{19}F(n, \alpha)^{16}N$	50	1, 5 [2] 3, 9 [3]	7, 3 s	6,13
$^{41}K(n, p)^{41}Ar$	80	1, 8 [2]	1, 83 h	1, 29
$^{27}Al(n, p)^{27}Mg$	80	1, 9 [2]	9, 5 min	0, 84; 1, 01
$^{41}K(n, \alpha)^{38}Cl$	50	3, 3 [2]	37, 3 min	1, 62; 2, 16
$^{27}Al(n, \alpha)^{24}Na$	115	4, 0 [2]	15, 0 h	1, 37; 2, 75
$^{24}Mg(n, p)^{24}Na$	180	4, 9 [2] 2, 1 [3]	15, 0 h	1, 37; 2, 75
$^{16}O(n, p)^{16}N$	90	10, 2 [2]	7, 3 s	6, 13
$^{19}F(n, 2n)^{18}F$	60	11, 0 [2] 10, 4 [3]	110 min	β^+
$^{14}N(n, 2n)^{13}N$	5	11, 1 [2]	10, 0 min	β^+
$^{39}K(n, 2n)^{38}K$	5	13, 4 [2]	7, 7 min	β^+; 2, 16

figure 2 le montre clairement. En conséquence, s'il y a un ou plusieurs emplacements d'irradiation où l'énergie des neutrons est variable, ou si la distribution spatiale en énergie des neutrons est connue, connaissant les rapports entre les flux rapides et thermalisés on peut séparer certaines activités parasites.

Les expériences et les mesures nous ont permis de constater qu'un générateur de neutrons utilisant la réaction nucléaire neutronigène $t(d, n)\alpha$ produit un flux de neutrons rapides de 14 MeV environ qui diminue très vite en s'éloignant de la cible, alors que le flux de neutrons thermalisés ne diminue pas si rapidement. La figure 3 montre la distribution des flux de neutrons rapides et thermalisés. La courbe 1 représente la variation du flux (exprimé par l'activité des échantillons) des neutrons rapides qui induisent la réaction nucléaire $^{27}Al(n, p)^{27}Mg$ et la courbe 2 de ceux qui induisent la réaction $^{27}Al(n, \gamma)^{28}Al$, la mesure étant faite avec des disques en aluminium de 27 mm de diamètre et de 7 mm de hauteur.

Nous avons placé devant la cible un réservoir d'eau de 300×300×250 mm et avons disposé dedans trois échantillons en trois positions (voir fig. 3).

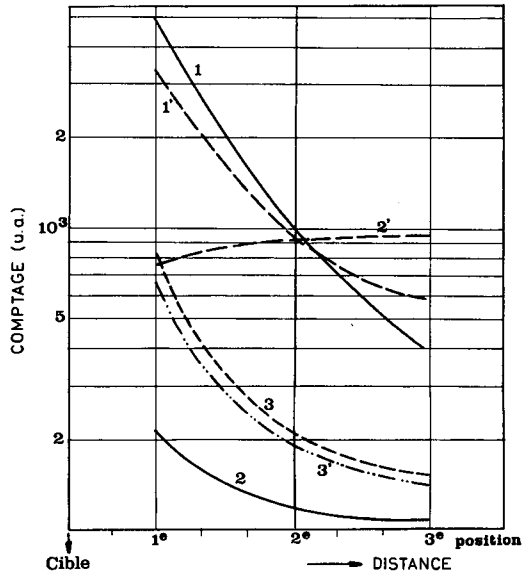

FIG. 3. Distribution des flux de neutrons rapides et thermalisés (voir texte).

Dans ces conditions, les distributions des flux rapide et thermalisé donnent les courbes 1' et 2'. On peut constater que le flux de neutrons rapides diminue un peu moins rapidement que dans le cas 1; par contre le flux de neutrons thermalisés a une valeur sensiblement plus élevée. Les courbes 3 et 3' montrent les distributions des flux de neutrons induisant la réaction $^{28}Si(n, p)^{28}Al$.

Les distributions spatiales des flux de neutrons d'énergies différentes fournissent donc un moyen de distinguer les activités parasites.

Prenons pour exemple le dosage du silicium dans le Silumin, alliage d'aluminium et de silicium. L'activité produite par l'irradiation du silicium à une énergie d'environ 1,78 MeV est perturbée par l'activité du radionucléide ^{28}Al créé par la réaction (n, γ) de l'aluminium. Cette perturbation dépend de la distribution des flux de neutrons d'énergies différentes, c'est-à-dire du rapport entre des flux ayant des énergies différentes. Mais si nous déterminons ce rapport – en irradiant deux disques en aluminium pur en même temps, puis deux capsules remplies de SiO_2, l'une dans la position 1 et l'autre dans la position 3 par exemple (v. fig. 3), et en mesurant l'activité à l'énergie γ de 0,86 MeV et 1,78 MeV – nous obtiendrons des constantes (exprimées par les activités mesurées).

Le rapport des activités, donc le rapport du flux des neutrons rapides à celui de neutrons thermalisés dans les positions mentionnées, dans le cas d'une géométrie fixée, est donné par les expressions:

pour l'aluminium:	$x_1 = c_{0,86}/c_{1,78}$	position 1
	$y_1 = C_{0,86}/C_{1,78}$	position 2
et pour silicium:	$x_2 = c_{0,86}/c_{1,78}$	position 1
	$y_2 = C_{0,86}/C_{1,78}$	position 2

(Les indices 0,86 et 1,78 indiquent que les comptages ont été effectués respectivement aux énergies de 0,86 et 1,78 MeV.)

Lorsqu'on active un échantillon de Silumin pour doser le silicium il faut déterminer l'activité parasite de ^{28}Al provenant de l'aluminium activé par les neutrons thermalisés. Il faut donc connaître d'une part l'activité de l'aluminium et d'autre part l'activité du silicium. On calcule l'activité de l'aluminium en utilisant les valeurs $c_{0,86}$ et $C_{0,86}$ obtenues à l'aide de x et y (compte tenu du fait que l'activité provenant de ^{28}Al peut diminuer par décroissance).

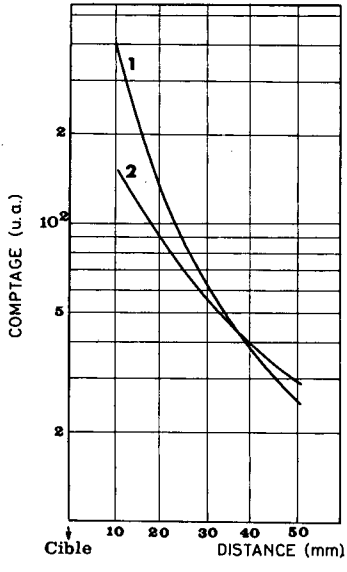

FIG. 4. Distribution des flux induisant les réactions ^{16}O (n, p)^{16}N (courbe 1) et ^{19}F (n, α)^{16}N (courbe 2).

Il convient de noter que, dans le cas des réactions nucléaires ayant un seuil d'énergie, il est aussi important de connaître les distributions des flux de neutrons d'énergies différentes.

On peut trouver dans la littérature de nombreuses constantes nucléaires et des calculs des sections efficaces et des seuils de diverses réactions nucléaires [1, 3-7]. Toutefois les valeurs mesurées avec notre propre appareillage sont aussi importantes, parce qu'elles peuvent contribuer à résoudre des problèmes pratiques. La figure 4 en est une illustration: On y voit la distribution des flux induisant les réactions $^{16}O(n, p)^{16}N$ (courbe 1) et $^{19}F(n, \alpha)^{16}N$ (courbe 2). De telles mesures servent à calculer les corrections à apporter lors du dosage du fluor en présence d'oxygène. Ainsi cette figure montre qu'à l'endroit où la courbe 2 s'est abaissée à la moitié de sa valeur, la courbe 1 a atteint le quart de sa valeur. Les mesures ont été faites sans aucune matière thermalisante.

En conclusion, on constate que les données concernant les sections efficaces et les seuils d'énergie peuvent faciliter, dans le cas de certaines réactions, la séparation de quelques activités parasites, et que l'étude des constantes nucléaires présente aussi des avantages pour l'analyse par activation.

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DISCUSSION

J.A. CZUBEK: When you perform the energy discrimination it seems to me that the accuracy of your measurements would be affected by changing the sample-target distance. In view of the finite dimensions of the sample, I do not think that the sample position is very well defined, and it can lead to greater inaccuracies in the final results than those due to the inaccurate nuclear data which we have been discussing today.

Section XII

COMPILATION AND EVALUATION: DATA CENTRES

Chairman

G. A. BARTHOLOMEW (Canada)

THE EXPANDING ROLE OF THE UNITED STATES NUCLEAR DATA COMMITTEE

W.W. HAVENS, Jr., Columbia University,
G.A. KOLSTAD, United States Atomic Energy Commission,
R. CHRIEN, Brookhaven National Laboratory,
United States of America

Abstract

THE EXPANDING ROLE OF THE UNITED STATES NUCLEAR DATA COMMITTEE.

The need for exchange of information between measurers, compilers, evaluators, and users of nuclear data has long been recognized in the reactor-physics field. Both National and International Advisory Committees have encouraged active co-operation. In the United States, the US Nuclear Data Committee fills the role. On the international level, the European-American Nuclear Data Committee and the International Nuclear Data Committee occupy similar advisory roles under the auspices of the Nuclear Energy Agency of OECD and the International Atomic Energy Agency, a UN body. It has recently become clear that it is necessary to promote close ties between measurers, compilers and users in the non-neutron data fields. In the United States, the USNDC now assists in establishing a feedback from users to measurers by compiling a request list for measurements needed for fission reactors, safeguards, fusion reactors, biomedical and basic-physics applications. The mechanism for doing this was to create a number of specialized sub-committees to survey the needs and establish user-measurer relationships in these specialized fields. These sub-committees will emulate the relationships which have been so successful in the reactor-physics field. To the extent possible, the committee and sub-committee members implement their own recommendations. The committee also makes available to users status reports on measurements currently planned or underway, and sponsors conferences and symposiums.

The USNDC operates under the general auspices of the US Atomic Energy Commission with participation from other government agencies, including the National Bureau of Standards, the Department of Defense and the National Science Foundation.

1. INTRODUCTION

The U.S. Nuclear Data Committee evolved from a series of predecessors starting in 1948 with the Neutron Cross Section Advisory Committee to the Atomic Energy Commission. Later its functions were broadened and it became the AEC's Nuclear Cross Section Advisory Committee. However little attention was paid to the needs of other than neutron cross section data and other related neutron data such as α, ζ, ν and the special properties of the fissionable material. Only recently have the legitimate needs for other than neutron data users been recognized, resulting in the formation of the U.S. Nuclear Data Committee.

The U.S. Nuclear Data Committee is in the process of evolving several mechanisms and procedures for the exchange of information between measurers, compilers, evalutors and users of nuclear data. One of these mechanisms is the publication of documents useful in the neutron cross section field. The forerunner of the cross section compilation known today as BNL-325 was first published under NCSAC auspices in 1952 as AECU-2040. Neutron cross section request compilations were originated by this group and have grown to the point where international request lists are assembled under IAEA auspices. The latest version, INDC(SEC)-276, has recently been distributed. Status reports of relevant nuclear energy programs have been dissem-

inated by the committee under the title of "Reports to the NCSAC" and contain valuable and timely detail of work in progress. These summaries have proved invaluable aids to workers in the neutron cross section field, and an indication of their success is their emulation by analagous organizations in other countries.

USNDC and its predecessors have sponsored many conferences and symposia since the inception of the Committees. The most recent series of conferences have been in the area of Neutron Cross Sections and Technology. The proceedings of these conferences are familiar to most scientists who specialize in neutron physics. The Conferences have probably been one of the most successful methods of improving the communication between measurers and users of nuclear data. Many of the recent conferences on nuclear data have been sponsored by the EANDC and the INDC because of the growing role of international cooperation in the field and the fact that they are so important as a method of disseminating information. The Conference we are presently attending is a good example of an attempt to assemble a group of measurers, compilers and users of nuclear data to improve the communication among these groups and to assess the need for further steps to meet the needs of nuclear data users on a broad scale.

The three documents, the Neutron Cross Section Compilation, the Cross Section Request Compilation and the progress reports from U.S. Laboratories, which have been widely distributed are the public manifestations of the workings of what was first a national, (U.S., NCASC), second a three-nation, (Tripartite Neutron Cross Section Committee, U.S., Canada and the United Kingdom), third a multi-national (EANDC) and finally a world-wide committee (INDC). The documents themselves have proved important because they describe what has been measured in the neutron cross section area, the measurements in progress, and what neutron cross sections need to be measured for neutron physics applications.

Supervising the preparation of these documents is only a small part of the work of these Committees. Most of the time the members devote to the work of the Committees is spent in attempting to assess the needs for nuclear measurement data that will be needed in the relatively distant future (more than two years in advance) and making arrangements for the appropriate manpower, facilities and samples to enable these measurements to be made. The mechanisms and procedures developed by these Committees have been so successful in the neutron nuclear data field that it appears desirable to attempt to assess the feasibility of using these mechanisms and proceudres in the non-neutron nuclear data field as these non-neutron data applications become increasingly important.

2. HISTORY

In the early days of the Manhattan Project it was not necessary to have an organization even as simple as a committee to determine what cross sections were necessary. Fermi understood the physics of chain reactors, knew all the people who were measuring neutron cross sections and what they were doing. He was measurer, compiler, evaluator, and the user of the neutron data all rolled into one. Of course, the data available in 1941 was very meagre, and there were very few scientists in the field so that one man could know all that was going on. However, as the Manhattan Project expanded, even Fermi could not keep up with everything that was being measured. H.H. Goldsmith undertook the task of keeping track of all neutron cross section measurements in progress and of compiling the results. At this time there was no difference between the

measurers and users of neutron data. The only people who knew anything about the field had to make the measurements which were required and the same people used the results for reactor calculations.

As the Manhattan Project expanded it soon became clear that no individual could contribute to all parts of the project. If the project was to be speeded up, some scientists had to specialize in making the measurements required, while others had to specialize in the design of the reactors and other new systems being developed. The objective of the Manhattan Project was clear. All the efforts of everyone working on the project were devoted to the practical application of nuclear energy.

In those days the measurers and users would get together to discuss which measurements should be given first priority. The measurers in general made those measurements the users requested. However, as is usual in the progress of any intelligent research project, the measurers did not measure only those quantities requested by the users. Being close to the measuring equipment, they could see that some potentially useful measurements could be made with a very small investment of time. At the time these measurements were made, the users often did not know that the measurements would be useful and would not have requested them. Some of these "bootleg" measurements paid off handsomely and some disappeared into oblivion. However, the important point for the purposes of this Conference was not that good measurements were made but that the measurers and users had the same background and education and could talk to each other without difficulty. They understood each other's problems and the procedures used to solve them.

There was little difficulty in communications as long as all of the work on the project was being done in the same laboratory. However, when the plutonium part of the Manhattan Project moved from Columbia University to Chicago in the fall of 1942, a communications problem developed. Formal communications mechanisms were set up in the form of bi-weekly progress reports. However, anyone who has been closely connected with a research project knows that very little is accomplished in a two week period. Therefore, these biweekly progress reports became rather repetitive. The repetitiveness of the progress reports and the cumbersome classification system which was used for their distribution tended to decrease the amount of communication through these progress reports. Quicker and more informal methods of communication had to be arranged. H.H. Goldsmith assumed the task of improving communications in the neutron cross section field. He would visit Columbia to find out what had been, and was being measured, and inform the group of the most important quantities to measure for the development of reactors, always giving an explanation of why these particular measurements were needed. Very little cross section information was available at the time and progress on cross section measurements was also limited by the samples which could be obtained. Almost as much time was spent on the procurement, purification and packaging of samples for neutron cross sections measurements as was spent on any other aspect of the neutron cross section program. However, all quantities that have been measured were immediately useful and the results were distributed informally by Goldsmith and other scientists who traveled between New York and Chicago and later Oak Ridge, Hanford and Los Alamos. Of course, this information was more formally distributed by Progress Reports but it was really through informal communication that the results of the more important measurements were rapidly distributed. It was Goldsmith that started the informal compilation of neutron cross sections which later developed into the AECU-2040 and BNL-325. The compilation was, fo course, a classified document at the

time with limited circulation and its first public manifestation was a Review of Modern Physics article in 1947. (H.H. Goldsmith, H.W. Ibser, and B.T. Feld, Review of Modern Physics 19, 259, 1947).

After World War II, most of the scientists in the National Laboratories which had been established during the war returned to their academic posts. It took several years for the Atomic Energy Commission to be organized and become effective. In 1948, the Division of Reactor Development recognized that the cross sections necessary for further development of nuclear energy were not being measured and set up the Neutron Cross Sections Advisory Committee. The Committee consisted of scientists who were actively measuring and using neutron cross sections. It was the first time that the difference between measurer and user was recognized, although in 1948 there wasn't very much difference between the two. Harvey Brooks and Thoma Snyder, then of General Electric Company (G.E.) were definitely recognized as users. There were very few measuring facilities at G.E. at that time and G.E. was one of the major contractors for the design of reactors for the production of power. Westinghouse was represented by Paul Garabedian, who had very little contact with the measurement program. Oak Ridge sent Al Weinberg to represent the users and Art Snell to represent the measurers. Jane Hall and Dick Taschek represented Los Alamos, Don Hughes and Carl Muhlhouse Argonne, W.W. Havens, Columbia and G. Weil, who had been with the original Fermi group at Columbia, represented the Atomic Energy Commission.

The meetings in the early days consisted of the members of the Committee reporting on the measurements which had been made in the laboratories they represented, the measurements in progress and the measurements planned for the immediate future. Attempts were also made to formulate a long range cross section measurements program. Obviously, most of the discussion in the original Committee was about neutron cross section measurements and therefore the measurers were the backbone of the Committee. Since the original Committee reported to the Division of Reactor Development, meetings were held at laboratories where reactor programs where being supported. Users of neutron cross sections would meet with the Committee and give it long lists of measurements required for reactor development. What the users wanted were all cross sections of all isotopes from 10^{-2} to 2×10^{7} eV in the steps of 10^{-3} eV. Obviously an impossible measurement program. The Committee had to sort out these requests and decide which measurements could and should be made. Not only did the Committee decide which measurements could and should be made, but they returned to their home laboratories and either made the measurements or inspired other groups in their laboratories to make them. Those measurements which could not be made by the members of the Neutron Cross Sections Advisory Committee or groups in the laboratories which they represented were listed and the AEC investigated possible mechanisms which could be established to enable them to be measured. It soon became clear that the main purpose of the Neutron Cross Sections Advisory Committee was to implement a measurement program rather than to formulate a specific list of cross sections which should be measured. The cross sections which could be measured with existing techniques and equipment were measured very rapidly. However, most of the needed cross sections could not be measured with existing techniques and equipment. Further progress in the field required developing new techniques and equipment. It was the recognition that measurement of the cross sections needed for the development of nuclear reactors were more in the area of research than in the area of development that led the Atomic Energy Commission in 1950 to transfer this Committee from the Division of Reactor Development to the Division of Research. Dr. George Kolstad, who headed the Research Division program, was representative

on this Committee and the basic framework of our present system began to take form.

3. DATA CENTERS

The first major accomplishment in the communications area of the Neutron Cross Section Advisory Group of the Atomic Energy Commission was establishing the Neutron Cross Section Compilation Group at Brookhaven National Laboratory. The first leader of this group was Donald J. Hughes, who was ably assisted by Irving Kaplan, now at MIT, in producing the first major Neutron Cross Section Compilation AECU-2040. The Brookhaven Neutron Cross Section Compilation Group has grown into the National Neutron Cross Section Center, and the Brookhaven Compilations, BNL-325, are known throughout the world as a source of basic data in the Neutron Cross Section area. The CCDN at Saclay was established in 1963 by tne ENEA upon the recommendation of the European-American Nuclear Data Committee. CCDN was patterned after the Brookhaven Compilation Center and many of the methods for handling Neutron Data which had been developed at Brookhaven were adopted by CCDN. The IAEA Neutron Cross Section Compilation group was set up in the late sixties at the IAEA headquarters in Vienna on the recommendation of the International Nuclear Data Committee. The Obninsk Compilation Center in the USSR fulfills a similar function in the Soviet Union. These four data centers operate as a world-wide net.

Arrangements have been made for transfer of data among these data centers, and the coordination between these centers, while not perfect, is steadily improving.

4. PROGRESS REPORTS

The next major communication vehicle of the USAEC's Neutron Cross Section Advisory Committee to be establihsed was the Progress Reports. It became obvious after the first few meetings that a great deal of time at Committe meetings could be saved if the reports given by the individual members at meetings of the Committee were written up before. Later it was recognized that Progress Reports contain a great deal of information which could be very useful to a much wider community than the Committee members, and the Progress Reports were therefore packaged together and given a relatively wide distribution as a single document. The system of preparing Progress Reports for the USNCSAC meetings, distributing them to members of the Committee, subcommittees and to others who are intimately engaged in making measurements of interest to the Committee, and later packaging the Reports in one document for wider subsequent distribution, is essentially the system which is in operation today. It was later adopted by the EANDC and the INDC, and still appears to be the best rapid communication mechanism for the applied nuclear data field.

The system of preparation and distribution of the Progress Reports is not, however, the really important factor in distributing the information. It's the content of the Progress Reports themselves that makes them valuable. The Progress Reports did not take on their present form without a great deal of argument and wrangling, and sometimes even arbitrary editorial cutting. It was recognized in the early 1950s that the work of the Committee also involved charged particle cross sections, especially for the study of the fission process. The name of the Committee was therefore changed to the Nuclear Cross Sections Advisory Committee, and a somewhat broader spectrum of measurements useful in the development of nuclear energy systems was included in the purview of this Committee. However, when the name of the

Committee was changed from Neutron to Nuclear, the exact content of the Progress Reports became a problem. Some took the broad view that all measurements which improved our knowledge of the nucleus were beneficial to the development of nuclear energy. Others took a narrower view that only those measurements which could be directly related to applied problems in the development of nuclear energy systems should be included in the Reports. After a period of confusion, the matter was settled by limiting the content of the progress reports to information pertinent to the request lists. We now appear to have reached a reasonable balance.

The boundary conditions of what should be included and excluded from Progress Reports has varied over the years and will continue to vary, because what is immediately relevant to the development of nuclear systems will vary with the existing state of knowledge at any particular time and the applications which are on the horizon. It has been our experience that the Reports should be limited to that material which is related to the development of nuclear systems, relatively liberally interpreted, (i.e., where a direct or indirect connection can be made between a measurement and an application). The Progress Reports to the various Nuclear Data Committees are not only helpful in the dissemination of information, but they also serve another useful purpose in requiring the scientists who prepare these reports to examine the measurements which have been made recently to assess the quality of their work and their relevance to the development of nuclear systems. In several cases, this review has led to the reorientation of a measurement program in a laboratory so that the results will be more immediately useful.

One of the important items which came out of the discussions on the content of the Progress Reports was that the description of every experiment should include the name of the individual who was performing the measurement. The scientists who read the Progress Reports could then write to obtain more information about the progress and results of the measurement. The inclusion of the name of the individual performing the measurement described in the Progress Reports has led to a great deal of beneficial informal communication between scientists with similar interests and may have been as important as any other single accomplishment of the cross section committees.

5. THE NUCLEAR CROSS SECTION REQUEST COMPILATION

The Neutron Cross Section Request Compilation originated in the early days of the Manhattan Project when it became obvious that it was not possible to measure all of the cross sections necessary for the development of nuclear systems. A request list was formulated and any useful request list must include a statement of priorities. There was no rigid statement of priorities until the U.S. Cross Sections Report list became a separate document. This occurred after the major declassification of nuclear data in 1955. When the Tripartite Nuclear Data Committee was formed after the first Geneva Conference in 1955, a combined TNCC request list was assembled and discussed by the TNCC. Each of the three countries of the TNCC, (U.S., U.K., and Canada), had its own definition of priority and the definitions had to be made consistent if the request lists were to be meaningful to all parties. The presently accepted U.S. priority definitions are:

Priority 1

Nuclear data which satisfy the criteria of Priority 2 and which have been selected for maximum practicable attention, taking into account the urgency of nuclear energy programme requirements.

Priority 2

Nuclear data which will be required during the next few years in the applied nuclear energy programme (e.g., the design of a reactor or fuel processing plant; data needed for optimum use of reactor fuel and construction materials such as neutron moderators, absorbers and radiation shields; space application and biomedical studies; data required for better understanding of some significant aspect of reactor behavior).

Priority 3

Nuclear data of more general interest and data required to fill out the body of information needed for nuclear technology.

The various nuclear data committees, national and international, have reviewed the nuclear cross section request lists on many occasions, and have made significant contributions to the development of a system which enables the requested measurements to be made. The latest publicly issued World Request List, INDC (SEC)-27L, contains requests for 1,411 measurements. Various estimates have been made about the scientific manyears required to make all these measurements. It's certainly in the high thousands and may even be in the tens of thousands. From the practical viewpoint it appears that the neutron cross section request compilation is much too long. If the request list could be reduced in length by removing duplications and less urgent requests, greater progress might be made. Without question, assembling the request compilations has been of considerable benefit to the international nuclear data committees. These lists may have helped scientists in laboratories not directly connected with the nuclear energy programs to establish a more useful research program. The Request lists appear to have value internationally provided that issuance is rapid. It would be an improvement to reduce the length of this request list and encourage only those measurements which are clearly of significant value.

6. NEW FACILITIES AND PROGRAMS

Soon after the Neutron Cross Section Advisory Committee was formed, the AEC recognized that a communications gap was developing between measurers and users, and assigned the task of preparing and assigning priorities of a list of cross section measurements desired for the development of fission reactors to a separate group more closely related to this technology, the Advisory Committee on Reactor Physics (ACRP). The Data Committee received requests from the ACRP with priorities assigned and discussed the measurements required to fulfill these requests. Those measurements which could be made with existing techniques and equipment were usually made very rapidly, but a large number of high priority measurements required the development of new techniques and equipment. It was in this area that the U.S. Data Committee made one of its greatest contributions to the development of nuclear energy. When all of the information desired by the reactor designers was assembled, it was divided into categories which requred different measurement techniques. The Committee recommended that facilities be established for making measurements which could not otherwise be made. Some of these measurements have only recently come to fruition. It was in 1952 that the Committee first noted that the cross sections of the transuranic elements would be important in the long range development of nuclear energy and recommended that the Atomic Energy Commission provide for production of the transuranic isotopes in quantities large enough to enable cross section measurements to be made. Fast time of flight systems were first discussed at Committee meetings in the early fifties and the recommendations to the AEC contributed to the

establishment of the fast time of flight facilities at Argonne, Los Alamos, Oak Ridge and Westinghouse. The use of large scintillators for capture cross section measurements was discussed at Committee meetings in the early fifties and many large scintillators were subsuquently constructed throughout the world to make neutron measurements. It was the USNCASC that recommended to the Atomic Energy Commission that the calutrons, the electromagnetic uranium isotope separation facilities at Oak Ridge, which were about to be scrapped be adapted to the separation of all isotopes so adequate samples would be available for cross section measurements. These facilities now provide the bulk of the stable isotopes for a variety of measure users throughout the world.

7. CONFERENCES AND SYMPOSIA ON NUCLEAR DATA

In 1948 communications and information exchange among AEC laboratories and scientists were cumbersome and difficult because of the classification system inherited from the Manhattan Project. One of the first tasks the U.S. Nuclear Data Committee assumed was to improve communications among the relevant AEC scientists. An effective way of doing this was by organizing information Meetings which were held at the various National Laboratories at approximately one year intervals. The meetings were very similar to what are now Topical Meetings of the American Physical Society. There were invited papers, which gave a review of a specific area, and contributed papers, which gave details on the latest measurements. The "Proceedings" of these neutron cross section information meetings were assembled by the scientists in the host laboratory and distributed as a report of that laboratory. These meetings were generally successful and invitations to attend them were sought after by many of the scientists working in the field.

After the declassification of most nuclear data in 1955 and the complete declassification of the remaining data in 1958 the Information Meetings became public although less frequent. The first public neutron cross section information conference was the Oak Ridge "Fast Time of Flight" Conference in 1956. The EANDC took on the sponsorship of international conferences in the nuclear data field in the early 1960s. A list of conferences sponsored by the EANDC is given in Table I.

The sponsorship of conferences has been an important function of the IAEA since its inception and when it assumed an important role in the nuclear data field, it sponsored a number of such conferences as an excellent way of increasing the communications between scientists. The IAEA has probably sponsored more conferences per year in the nuclear energy area than any other single organization. Some of the conferences sponsored by the IAEA of particular interest to the INDC are listed in Table II.

After the very successful EANDC - sponsored conference on Neutron Time of Flight Methods which took place in Paris, 24-27 July, 1961, the U.S. Nuclear Data Committee thought that national neutron cross section information meetings were no longer necessary. However, it soon became obvious that communications and the cross fertilization among measurers, compilers and users of nuclear data was not taking place. To meet this problem, the Committee reinstituted its information in meetings in a different form under the name "Neutron Cross Section and Technology Conferences." The first one of which was in Washington in March of 1966, the second in Washington in March, 1968, and the third in Knoxville in March, 1971. The Committee has found that such conferences serve an important function in the nuclear data area and organizing such meetings is one of the Committee's important tasks.

TABLE I. INTERNATIONAL CONFERENCES SPONSORED BY EANDC

Conference	Location	Date
Symposium on Neutron Time of Flight Methods	Saclay	24-27 July, 1961
Isotope Supply and Sample Preparation	Geel	6-8 August, 1963
Symposium on Absolute Determination of Neutron Flux in the Energy Range 1 to 100 keV	Oxford	10-13 Sept., 1963
Automatic Acquisition and Reduction of Nuclear Data	Karlsruhe	20-24 July, 1964
Round Table Conference on High Precision Chemical Analysis of Substances of Interest to Nuclear Energy	Brussels	18-22 Jan., 1965
International Conference on Study of Nuclear Structres with Neutrons	Antwerp	19-23 July, 1965
Seminar on Preparation and Standardization of Isotopic Targets and Foils	Harwell	20-21 Oct., 1965
2nd Round Table Conference on High Precision Chemical Analysis of Substances of Interest to Nuclear Energy	Brussels	November, 1965
Seminar on Intense Neutron Sources	Santa Fe	19-23 Sept., 1966
Symposium on Neutron Standards and Flux Normalization	Argonne	21-23 Oct., 1970

8. THE EXPANDING SCOPE OF THE U.S. NUCLEAR DATA COMMITTEE

As the use of nuclear data has expanded into wider and wider segments of society and in more and more technical areas, so has the need for cross section measurements. The original U.S. Neutron Cross Sections Advisory Committee devoted itself only to the measurements of cross sections for the development of fission reactors. However, it became obvious that nuclear data was needed in many other areas of science and technology.

9. SPACE

One of the first major perturbations in the technical cognizance of the USNDC was the request submitted by scientists working for the National Aeronautics and Space Administration (NASA) for high energy cross sections which

TABLE II. A FEW OF THE IAEA CONFERENCES OF INTEREST TO THE INDC

1. Pile Neutron Research in Physics	Vienna, Austria, October, 1960
2. Physics and Chemistry of Fission	Salzburg, Austria, August, 1965
3. Nuclear Data for Reactors	Paris, France, October, 1966
4. Neutron Standards for Neutron Measurements	Brussels, Belgium, May, 1965
5. Physics and Chemistry of Fission	Vienna, Austria, August, 1969
6. Neutron Capture Gamma Ray Spectroscopy	Studsvik , Sweden, August, 1969
7. Nuclear Data for Reactors	Helsinki, Finland, June, 1970
8. Standards for Nuclear Data Measurements	Vienna, Austria, November, 1972
9. Application of Nuclear Data in Science and Technology	March, 1973

would be important in the design of shields for space vehicles. The data needed for cross sections for shielding calculations for neutrons of energy less than 20 MeV were immediately available because these data had already been developed for reactor shielding. The shielding of space vehicles, on the other hand, requires a knowledge of cross sections of particle interactions at energies up to housands of MeV. The data available for shielding calculations in this energy region was very sparse indeed. Accordingly, the cross section request compilation increased somewhat when the requests for high energy data were added. There is no doubt that these cross sections will be necessary for the design of long range space vehicles. However, at this time the construction of such space vehicles appears to be relegated to the relatively distant future so that nuclear data problems associated with deep space flight have been given low priority by the NASA.

10. SAFEGUARDS

The next major perturbation in the activities of the USNDC was the request for nuclear information which could be used in the safeguards program. Safeguards is a very important area and will receive a great deal of attention and support for a long period of time. The scientists developing safeguards systems are only beginning to reach the stage where they can see what nuclear data are needed for the long range improvement of present systems and the design of new systems. The nuclear data needed for the development of the safeguards program are more varied than the nuclear data needed for reactor development. The safeguards program requires a large component of photonuclear and charged particle data along with neutron data. The problem of how best to handle the safeguards data needs has been discussed at great length in both the international and national nuclear data committees. A satisfying solution appears to be evolving in the U.S. by having a specialized subcommittee cognizant of this area.

11. FUSION

A third area in which nuclear data will play a very important part is in the development of fusion reactors. The problem of attaining and confin-

ing an adequately hot plasma has so dominated research in controlled thermonuclear fusion that thus far relatively little attention has been given to the engineering development of the systems which will be necessary to utilize the energy produced. The first fusion reactor prototypes will probably utilize the (d,t) reaction which emits 14 meV neutrons. The cross sections of the materials which will be necessary for the construction of a fusion reactor are not well enough known in this higher energy region to intelligently design a fusion reactor system. The role of charged-particle nuclear fusion chain reactions at MeV energies may be important for fusion reactor development. A need for accurate absolute reaction cross sections exists for the proper assessment of these chain reactions. Increasing attention has been given to this problem in the U.S. and there is no doubt that a great deal more attention will have to be given to it before economic, safe fusion reactors can be built and operated.

12. MEDICINE

The use of nuclear systems in medicine has also expanded very rapidly in the last few years. For example, the Anger camera for radiographic imaging, developed at the Lawrence Laboratory, has had an enormous sale in hospitals in the United States. There are many ways in which this camera can be improved and obviously improvement of our knowledge of nuclear radiation will help in the improvement in its design. The use of radioactive and stable isotopes in diagnostic techniques in medicine has also expanded rapidly. Physicists in radiology departments are asking for details of nuclear decay schemes which previously have been of interest only to theoretical specialists studying the particular excitation energy of a particular isotope in which the transition occurred.

Electronuclear machines have long been used in the treatment of cancer and it's quite obvious that the data on the radiations from these electronuclear machines is adly inadequate for some of the proposed theraneutic applications. In fact, the whole area of medical physics appears to be expanding rapidly and may in the future utilize the talents of many of the physicists who are not now working in this area of applied nuclear science.

13. REORGANIZATION OF THE U.S. NUCLEAR DATA COMMITTEE

Recognition of the increasingly wide use of nuclear data in applications other than fission reactors has prompted the reorganization of the U.S. Nuclear Data Committee (USNDC) into a group more broadly conversant with the increased need for nuclear data. The Committee operates under the USAEC's Division of Physical Research, in cooperation with the Office of Standard Reference Data Systems of the National Bureau of Standards.

In reorganizing the USNDC it was obvious that the range of expertise in the Committee must be increased. If the expertise in neutron cross sections that existed in the Committee were to be matched in a balanced way by all of the new areas of interest, it was obvious that the size of the Committee would grow so as to become unmanageable. The scheme devised was to have the USNDC consist of regular members chosen to broadly represent the areas of interest, ex officio members, chosen to represent key management organizations, and subcommittee members, serving on disciplinary and applications subcommittees. The latter are appointed by the Chairman with the advice and consent of the Committee. Representatives of the AEC, National Bureau of Standards, Department of Defense, and National Science Foundation are all included in the ex officio membership along with U.S. participants in international nuclear data Committees. Committee members are drawn from

TABLE III. ORGANIZATION OF THE NEW USNDC

MEMBERS

Robert E. Chrien, Chairman, BNL
Harold E. Jackson, ANL
Harry Alter, AI
Robert C. Block, RPI
Charles D. Bowman, NBS
Randall S. Caswell, NBS
Herbert Goldstein, Columbia U.
Daniel J. Horen, ORNL
Melvin H. Kalos, N.Y.U.
Michael S. Moore, LASL
Henry W. Newson, Duke U.
Francis G.J. Perey, ORNL
Gerald C. Phillips, Rice U.
James Robertson, BNL
George L. Rogosa, USAEC
Edward M. Smith, U. of Miami
Donald Steiner, ORNL

EX OFFICIO MEMBERS

William W. Havens, Jr., Columbia U.
Philip B. Hemmig, Reactor Division, USAEC
George A. Kolstad, Research Division, USAEC
David R. Lide, Jr., NBS
William Bartels, USAEC
William S. Rodney, NSF
Alan B. Smith, ANL
Richard F. Tauschek, LASL
Robert W. Wood, Bio. & Env., USAEC

ALTERNATES

(For Dr. Kalos, DNA)
Dean C. Kaul, USAF

(For Dr. Lide, NBS)
L. Gevantman, NBS

SUBCOMMITTEES

Separated Isotopes
F. Perey, ORNL, Chrm.
W.M. Good
H.W. Newson, TUNL
R.C. Block, RPI
C.L. Rogosa, USAEC
G.A. Cowan, LASL
E.M. Smith, U. of Miami

Charged Particles
H.W. Newson, TUNL, Chrm.

Standards
E.S. Caswell, NBS, Chrm.
W.W. Havens, Columbia U.
W. Poenitz, ANL
L. Stewart, LASL
B. Leonard, PNL

Elastic & Inelastic Scattering
A.B. Smith, ANL, Chrm.

Gamma-Ray Production
H.E. Jackson, ANL, Chrm.
H. Motz, LASL
R.E. Chrien, BNL
J.K. Dickens, ORNL
V.J. Orphan, GRT

Total Capture Cross Sections
R.L. Macklin, ORNL, Chrm.
R.C. Block, RPI
M.P. Fricke, GRT
W. Poenitz, ANL
J.B. Czirr, LLL

Fission
M.S. Moore, LASL, Chrm.

Resonance Paramaters
Integrals & Total Cross Sections
R. Block, RPI, Chrm.

Fast Neutron Reactions
H. Alter, AI, Chrm.
D. Barr, LASL
D. Gardner, LLL
J.L. Brownlee, LLL
G. Butler, LASL
D.L. Smith, ANL

Medium Energy Cross Sections
R.E. Chrien, BNL, Chrm.
D. Cochran, LASL
G.C. Phillips, Rice U.

Controlled Thermonuclear Research
D. Steiner, ORNL, Chrm.
H. Goldstein, Columbia U.
D. Dudziak, LASL
C.F. Barnett, ORNL
V. Orphan, GRT
L. Stewart, LASL

Nuclear Data for Materials & Environmental Analyses
D.J. Horen, ORNL, Chrm.
F. McGowan, ORNL
G. Gordon, U. of Maryland
J. Mayer, Cal. Tech.
T. Cahill, U. of Cal., Davis

Biomedical Applications
J.S. Robertson, BNL, Chrm.
E.M. Smith, U. of Miami
G.L. Brownell, Mass. Gen. Hosp.
J.S. Laughlin, Memorial Hosp. N.Y.
R.J. Shalek M.D., Anderson Hosp., Houston
R.S. Caswell, NBS
D.J. Horen, ORNL

Photonuclear Reactions
C.D. Bowman, NBS, Chrm.
E.G. Fuller, NBS
T. Godlove, NRL
W. Miller, U. Notre Dame
R.L. Bramblett, GRT
B.L. Berman, LLL

a representative cross section of the basic and applied research community and include representatives of other professional groups such as the American Physical Society, the American Nuclear Society and the Society of Nuclear Medicine. The scope of Committee activities may be gauged from the list of Committee members, ex officio and subcommittee members, which is given in Table III. It is evident that controlled thermonuclear research, safeguards, medical and biological application and industrial analysis have all been given special emphasis.

The USNDC hopes to develop a "request list" of nuclear data needs for each of the specialties of its subcommittees. Status reports describing recent experiments in progress in the broader range of activities are also expected to be produced for each of the meetings of the USNDC. The USNDC intends to continue to emphasize its role in the neutron and fission physics area since fission reactors remain a prime consumer of nuclear data, and if feasible extend its role to include non-reactor nuclear data and the needs for basic science. One important function is the coordination of nuclear compilation efforts on a national and international basis. In this effort the UNSDC will cooperate with the International Atomic Energy Agency (IAEA), the International Nuclear Data Committee (INDC), and the European-American Nuclear Data Committee (EANDC). The USNDC will provide, for example, its request list to aid the IAEA in the preparation of its world-wide nuclear measurements request lists. Some sections of this list will contain non-fission reactor data such as requests related to controlled thermonuclear research and safeguards.

The USNDC through its ties with the nuclear medicine community hopes to promote user-measurer symposia in nuclear data for medicine and attempt to identify the nuclear data needs for medical applications. The Committee will also act as a broker for suggestions from medical users on the best ways of compiling nuclear data and presenting them in a more useful form. The Committee will also examine the question of making nuclear compilations useful to other kinds of users.

Although the application of nuclear data is a primary concern of the USNDC, the Committee is cognizant of the pivotal role of basic research in the overall scientific effort. A large proportion (about 1/3), of present NDC membership consists of scientists engaged in basic research. The NDC recognizes the importance of the nuclear compilation effort for basic science as well as for application. The makeup of the USNDC membership suggests that it is becoming better qualified to represent the interests of both the pure and applied research communities in the United States.

DISCUSSION

L. HJÄRNE: Professor Havens' paper is the single one at this Symposium to deal with what the Weinberg Report* called "Scientific Intelligence" or what may be understood as science management information. A scientist in charge of a research institute, large or small, is indeed a manager in addition to being a scientist and he needs management information. He can get such information from two sources:

(a) Progress reports, which tell him what work is in progress and who is doing it;

* Science, Government and Information: a report of the President's Science Advisory Committee, US Government Printing Office, Washington, D.C., 10 January 1963.

(b) Request lists, which tell him what work needs to be done – what products he can sell, so to speak.

Much discussion in the past has centred on the reorganization of request lists so that account can be taken of the greatly increased scope of non-neutron data and the greatly enlarged range of applications. I should, therefore, like to address myself to one specific aspect of the problem and ask how, in your opinion, we should correlate the priorities assigned to different applications. How is it going to be done in the United States and how is it going to be done internationally? My contention is that you cannot do this satisfactorily, even though priorities are useful and very much needed within each field of application.

W. W. HAVENS, Jr.: There are no criteria as yet for establishing priorities in the selection of measurements which are required in different fields. For the present, the USNDC will have to discuss measurements on an individual, ad hoc, basis and decide which ones should be performed first, taking into account the difficulty of the measurement and the equipment and manpower available to make it. Our experience has been that required measurements are made very rapidly when the equipment and techniques are available. It is when new developments are required that the measurements take a long time to make.

G. A. KOLSTAD: I should like to make a few comments by way of amplifying the remarks of Professor Havens. You will have noted that three of the sub-committees to which he referred consisted of only one person each. This reflects the fact that the USNDC is still in an early stage of its re-organization and that, at the time the paper was prepared, all sub-committee members had not yet been appointed. Secondly, with respect to priorities, as you may know, in the field of fission reactors we have the Advisory Committee on Reactor Physics, whose function is to screen the requests from the reactor community and to define and assign priorities within the fission reactor field. It is our hope that the other areas which require nuclear data, such as biomedicine, safeguards and controlled thermonuclear reactors, will establish similar groups to recommend definitions of priorities within their spheres of responsibility and to assign priorities to specific measurements needed. Thus, I would not expect the development of a common set of priorities but rather of different sets of priorities within each applied area. Lastly, I might mention that we have decided to bring together a set of "sanitized" versions of the reports of the sub-committees into a single volume and to distribute them on a world-wide basis. The first such volume reached my desk a week ago and is on its way to the printers.

D. J. HOREN: When there are priorities in many different areas, the person who makes the decision as to what actually gets done must take into consideration the effect of the priorities in one area on those in other areas. There is a very strong coupling in some cases. The compilation of basic data is a pertinent example.

J. J. SCHMIDT: What are the sponsoring agencies of the USNDC?

G. A. KOLSTAD: The USNDC operates under the general auspices of the USAEC. This is for reasons of convenience, so that it can have a single administrative point of contact with the Government. There are other participating Federal agencies – the Office of Standard Reference Data Systems of the National Bureau of Standards, as mentioned by Professor Havens, the National Science Foundation and the Department

of Defense, i.e. the Defense Nuclear Agency (DNA). NASA also contributes some input but does not formally participate — this input is supplied through its laboratories.

W.W. HAVENS, Jr.: The list of ex officio members given in the paper includes the representatives from the other agencies.

J.J. SCHMIDT: Dr. Havens referred to the usefulness of the so-called RENDA Request List, which is directed to nuclear data needs for fission reactors. The IAEA is now editing this list on a world-wide scale and is developing similar nuclear data request lists for thermonuclear fusion and for nuclear materials safeguards purposes. There is the question whether request lists of this type should be developed also for other fields, such as biomedicine or activation analysis. This brings me to the following general question: what do you consider to be the most useful means of getting the data needs of the users known to potential producers so that these needs can be satisfied within reasonable time limits compatible with, for example, design deadlines the users must observe?

W.W. HAVENS, Jr.: The best way to have a measurement made is to get hold of the measurer and convince him of the importance of doing it. To be sure, this is not a formal mechanism. However, the request lists have — we believe — had some effect on the setting up of research programs. They have helped establish priorities in those cases where techniques and facilities were available for making measurements. In addition, they have drawn attention over the years to areas in which facilities and techniques do not exist. For example, there is the case of capture cross-sections, which is now well on the way to being solved. If you go back to 1966 and look through BNL 325, you will find that there were very few Γ_γ's in the compilation, the capture cross-section measurements were very poor and the discrepancies were very, very large. But capture cross-sections are quite important, especially where fast reactors are concerned, and therefore a real effort was made to get some good measurements. This was done by the European-American Nuclear Data Committee and I think the number of capture measurements has increased substantially since that time. There are still discrepancies and probably always will be, but the data have improved a great deal and are still improving.

G.A. KOLSTAD: It should also be emphasized that the U.S. Nuclear Data Committee is not an advisory committee. Rather, it can be looked upon as an operational group. Members are chosen on the basis of their technical competence and their broad responsibilities and authority in implementing the conclusions of the group. Thus, the actions of the group are largely self-implementing. It makes recommendations only incidentally to its main responsibilities for information exchange and program coordination and then only when they are inseparable from its other functions. Thus, there is a rather short and direct relationship between the users of the data and the measurers.

T. FUKETA: What account are the sub-committees taking of nuclear data in connection with the waste disposal problem?

W.W. HAVENS, Jr.: The Advisory Committee on Reactor Physics advises the USNDC of the cross-sections and priorities which are necessary for reactor developments. It would be the responsibility of the Advisory Committee on Reactor Physics (ACRP) to inform the USNDC of measurements which would be necessary for the waste disposal problem.

ЦЕНТР ПО СБОРУ, ОЦЕНКЕ И РАСПРОСТРАНЕНИЮ НЕНЕЙТРОННЫХ ЯДЕРНЫХ ДАННЫХ ГКАЭ СССР

Ф.Е. ЧУКРЕЕВ
Институт атомной энергии им.И.В.Курчатова,
Москва,
Союз Советских Социалистических Республик

Abstract–Аннотация

CENTRE FOR COLLECTION, EVALUATION AND DISSEMINATION OF NON-NEUTRON NUCLEAR DATA.

In accordance with a recommendation made by the International Atomic Energy Agency, the USSR State Committee on the Utilization of Atomic Energy decided in 1972 to establish a centre for the collection, evaluation and dissemination of non-neutron nuclear data. The author describes the scope of the centre's operations and the plans for carrying out its tasks.

ЦЕНТР ПО СБОРУ, ОЦЕНКЕ И РАСПРОСТРАНЕНИЮ НЕНЕЙТРОННЫХ ЯДЕРНЫХ ДАННЫХ ГКАЭ СССР.

В соответствии с рекомендацией МАГАТЭ Государственный Комитет по использованию атомной энергии СССР (ГКАЭ СССР) в 1972 году принял решение создать Центр по сбору, оценке и распространению ненейтронных ядерных данных. В докладе будет рассмотрен круг задач, решаемых Центром, и дана информация о планах работы.

В соответствии с рекомендациями Международной рабочей группы по данным о структуре ядра и ядерным реакциям [1] (IWGNSRD) ГКАЭ СССР принял решение организовать Центр по данным о строении атомного ядра и ядерным реакциям ГКАЭ. Этот Центр организован в Институте атомной энергии им.И.В.Курчатова (г.Москва). Одновременно Академия наук СССР приняла решение об организации аналогичного Центра в Ленинградском институте ядерной физики (г.Гатчина). Оба Центра, согласовав программы своей работы, действуют в тесном контакте друг с другом. Поскольку оба Центра расположены в городах, вблизи которых сконцентрированы наибольшие научные силы и наиболее хорошо оборудованные лаборатории, то мы рассчитываем, что не будет возникать трудностей при привлечении научных сотрудников высокой квалификации к решению задач, стоящих перед вновь созданными Центрами. Кроме того, такое расположение Центров позволит легко получать много дополнительной информации от авторов работ путем личного общения.

Центры собирают всю информацию о ядрах, не вводя какого-либо предварительного элемента оценки. Сбор информации – одна из главнейших задач Центров; она решается путем разумного разделения труда между ними с последующим обменом всей собранной информации. Без достаточно полной информации, записанной в таком виде, который обеспечивает оценщику легкость доступа и возможность автоматизации рутинной части его работы, трудно рассчитывать на привлечение высококвалифицированных физиков к работам по оценке ядерных данных, необходимых для решения научно-технических задач.

Предполагается, что Центр по данным о строении атомного ядра и ядерным реакциям не только собирает информацию, но и организует в рамках ГКАЭ работы по оценке тех данных, которые необходимы как для решения научно-технических задач ГКАЭ, так и для фундаментальных исследований.

В своей деятельности по оценке ЦАЯД ГКАЭ поддерживает и будет поддерживать тесные связи с ЦАЯД АН СССР.

ЦАЯД ГКАЭ должен обеспечивать ненейтронными ядерными данными следующие основные прикладные области:

1. Реакторостроение.
2. Исследования в области управляемого термоядерного синтеза.
3. Технические методы системы гарантий.
4. Элементный анализ вещества с использованием ядерно-физических методов.
5. Производство и применение искусственных радиоактивных изотопов.
6. Защита и дозиметрия.
7. Ядерная физика, астрофизика и т.д.

Более подробное изложение потребностей каждой из этих областей дано в работе [2].

Сейчас нам трудно более четко определить тематику деятельности ЦАЯД ГКАЭ вследствие того, что анализ запросов пользователей еще не завершен и пока не выработан список первоочередных потребностей. Тем не менее уже сейчас можно сказать, что в области реакторостроения существенная часть работ по оценке ядерных данных будет связана с задачами контроля нейтронных потоков внутри реакторов. Для этих задач необходимо знать:

а) сечения активации тепловыми и резонансными нейтронами элементов, применяемых в качестве детекторов нейтронов (Na, Mn, Cu, ^{103}Rh, V, Dy, ^{177}Lu, W, Ag, Pt, Au), времена жизни и схемы распада ядер продуктов. Для учета фона необходимы эти же данные для элементов конструкционных материалов (Ni, Al, Fe, Cd) и ядерного горючего (^{235}U, ^{238}U, ^{239}Pu);

б) сечения активации элементов, применяемых в качестве пороговых детекторов (O, P, S, Al, Mg, Fe), времена жизни и схемы распада ядер-продуктов.

Одной из главнейших задач ЦАЯД ГКАЭ является задача обеспечения ненейтронными ядерными данными интенсивно ведущихся в институтах ГКАЭ исследований по управляемым термоядерным реакциям. Для этих исследований необходимы данные о сечениях взаимодействия легких ядер при энергиях в несколько МэВ. Нужны также и данные о спектрах гамма-лучей и временах жизни ядер, рождающихся в результате взаимодействия продуктов термоядерных реакций с элементами конструкционных материалов и материалов, которые могут быть использованы для воспроизводства трития (Li, Be, C, F, Nb, Mo, Zr).

Подробный анализ необходимых ядерных данных для термоядерных исследований приведен в работах [3,4].

Приведенный выше список основных прикладных областей науки и техники, которые необходимо обеспечивать ненейтронными ядерными данными, не может, конечно, считаться окончательным. Наблюдающаяся сейчас тенденция к внедрению методов, созданных для решения ядерных

задач, в других областях, несомненно, поставит новые, может быть, даже не чисто ядерные задачи перед оценщиками.

В качестве примера можно привести очень интересный и,по-видимому, многообещающий метод элементного анализа микроколичеств вещества по характеристическому рентгеновскому излучению, возбуждаемому в нем пучками тяжелых заряженных частиц, такими, как протоны и альфа-частицы.

Этот метод использует германиевые и кремниевые полупроводниковые спектрометры и ускорители средних энергий, т.е. типичное оборудование современной ядерно-физической лаборатории. Мы думаем, что если этот метод найдет широкое применение, то главенствующая роль в оценке необходимых данных будет принадлежать физикам-ядерщикам, хотя этот метод в своей основе не ядерный, а атомный.

Широкое развитие работ по сбору, оценке и распространению ненейтронных ядерных данных невозможно без международного объединения усилий Центров разных стран. В первую очередь международное сотрудничество необходимо в области сбора информации о работах по ядерной физике - основных поставщиках ненейтронных ядерных данных. В этой области,по-видимому,наибольший опыт имеет Проект по Ядерным Данным (Nuclear Data Project),руководимый Д.Дж.Хореном (D.J.Horen). Принятая в Ок-Ридже система реферирования в ключевых словах обладает тем несомненным достоинством, что рефераты работ, полученные этим путем, понятны и человеку, и вычислительной машине. Мы считаем, что будущее развитие информационной системы мирового масштаба по данным о строении атомного ядра и ядерным реакциям будет происходить на базе рефератов в ключевых словах. Необходимо, конечно, расширять круг работ, реферируемых в этой системе, и прежде всего за счет расширения классов реферируемых работ. На наш взгляд, необходимо также сделать работу по составлению этих рефератов общей работой, разделив ее между физиками многих стран. Такое разделение при обязательном условии обмена всей собранной информации выгодно по двум причинам:

1. Увеличится скорость поступления сведений о выполненных работах к оценщикам и,следовательно, ими будет охвачено больше первичных данных.

2. Исчезнут языковые трудности, которые сейчас иногда приводят к досадным погрешностям при реферировании за рубежом работ советских ученых.

Если все редакторы ядерно-физических журналов примут рекомендации IWGNSRD [5] и обеспечат реферирование работ в ключевых словах, то это, конечно, ускорит поступление данных о выполненных работах в информационную сеть и ликвидирует языковые трудности. Но необходимо, по нашему мнению, обратить внимание на то, что если рефераты будут составляться самими авторами работ, то на содержании рефератов может заметно отразиться личная точка зрения автора, и тогда реферат получится субъективным. Такая субъективность может проявиться особенно сильно в разделе "комментарий" (где рассказывается о параметрах экспериментальной установки) и осложнить последующую работу оценщика. Поэтому особенно желательно, чтобы при выполнении работы по оценке оценщик имел возможность легко установить контакт с автором для уточнения сведений об использованной методике.

До тех пор пока не все редакции публикуют рефераты в ключевых словах, работу по реферированию следовало бы разделить между рядом

региональных центров в соответствии с языком и местом публикации. Такое разделение позволило бы включать в библиотеки всех тех групп и организаций, которым необходимы ядерные данные, рефераты ядерно-физических работ одновременно с их публикациями или даже несколько раньше.

Советские Центры могли бы взять на себя реферирование работ, публикуемых на русском языке.

Для организации работы в международном масштабе по реферированию статей неообходимо, на наш взгляд, уточнить правила реферирования, так как опыт показывает, что правила, изложенные в работе [6], допускают неоднозначное толкование. Организацию уточнения правил составления реферата могла бы взять на себя Секция Ядерных Данных МАГАТЭ.

Обмен рефератами наиболее целесообразно осуществлять через Секцию INIS, возможности которой, по-видимому, достаточны, чтобы предоставить каждой группе оценщиков рефераты ядерно-физических исследований на том носителе, который максимально удобен в услових данной группы.

Мы столь подробно останавливаемся на проблеме сбора информации о работах потому, что без развитой системы международного сотрудничества в этой области едва ли возможно включить большие новые группы физиков в деятельность по оценке. Необходимость разделения труда в международном масштабе еще более очевидна при выполнении работ по оценке. Их очень трудно выполнять без непосредственных контактов с авторами ядерно-физических исследований, так как публикации не всегда содержат достаточно подробные сведения.

В заключение следует еще раз подчеркнуть важность инициативы МАГАТЭ в решении проблемы сбора, оценки и распространения нейтронных данных. Можно надеяться, что эта инициатива будет понята и поддержана всеми заинтересованными лицами и организациями. Учитывая сложность решения всей проблемы в целом, следует приветствовать любой частичный успех в этом направлении.

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DISCUSSION

M. LEDERER: Does the work of your centre help to support, or is it in other ways connected with, the compilations of Peker and Dzhelepov?

F.E. CHUKREEV: Yes, such support is of course provided. In fact, the work of our centres would be impossible without close contacts with our country's scientists.

F. FRÖHNER: What is the relationship between the new centre and the Nuclear Data Centre at Obninsk? How much overlap do you expect, or will there be a clear division of tasks?

F.E. CHUKREEV: We are co-ordinating our activities with those of the Obninsk Centre and shall do everything possible to avoid duplication.

J.J. SCHMIDT: Is there any organization in the Soviet Union which corresponds to the Nuclear Data Committee of the United States?

F.E. CHUKREEV: Yes, there is. The body in question is the Nuclear Data Committee of the USSR State Committee on the Utilization of Atomic Energy.

NUCLEAR DATA PROJECT: OPERATIONS, STATUS AND PLANS

D.J. HOREN,
Nuclear Data Project,
Oak Ridge National Laboratory,*
Oak Ridge, Tenn.
United States of America

Abstract

NUCLEAR DATA PROJECT: OPERATIONS, STATUS AND PLANS.

A description of the operations of the Nuclear Data Project is presented. This includes a discussion of the Project's reference systems, compilation and evaluation efforts, generation of a nuclear structure data file, and the types of publications and services offered. The status of the mass-chain compilations for $A > 44$ is reviewed. Interactions between the Project and other compilation groups will be noted. Some time will be devoted to future plans for handling the abundance of nuclear data being generated, and the contemplated modifications to the Project's methods of communicating with producers and users of nuclear data.

Historically, the Nuclear Data Project has been primarily concerned with the problem of providing evaluated summaries of nuclear structure data, mainly for those nuclei with mass number $A \gtrsim 45$. During the process of performing this function, a need arose for a convenient manner in which to supply compilers with pertinent references, and this led to the development of a nuclear keyword system. In this paper, we will discuss 1) the Project's reference systems and usage, 2) some of the aspects involved in the preparation of mass chain compilations published in Nuclear Data Sheets, and 3) some interactions with applied data users and some future plans.

The Nuclear Data Project scans[1] and prepares keywords for all papers (i.e. about 3,000 per year) published in low to medium energy nuclear physics. The keyword system used is an extended version of that originally developed by K. Way and collaborators and initially adopted by the journal Nuclear Physics. It is based upon utilization of descriptors commonly used in the field. The first figure shows examples of keyword strings that have been used to describe the contents of a few selected papers. Notice that each begins with a major category, e.g., Radioactivity, Fission, Nuclear Reaction, etc., and contains both information as to what has been measured as well as what has been deduced. Computer programs have been written which allow various types of sorting and listing. A major use of this system is in the preparation of Recent References which appears three times per year as issues of Nuclear Data Sheets. In this publication, the keywords are ordered according to atomic mass number, which is convenient for those interested in experimental papers which contain nuclear structure information on specific nuclides.

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* Operated by Union Carbide Corporation for the US Atomic Energy Commission.

[1] Since 1968, this function has been performed by David West of the Data Project.

RADIOACTIVITY ^{62m}Co; measured $T_{1/2}$, Eβ, $\beta\gamma$-coin, $\beta\gamma$-CP; deduced log *ft*, Q. ^{62}Ni deduced levels, J, π, ICC. Ge(Li) detector, 3.5 keV at 660 keV.

NUCLEAR REACTIONS ^{12,13}C(d,d),(d,p), E = 0.4-0.85 MeV; measured σ(E;θ); deduced optical model parameters. ^{13,14}C levels deduced S. Enriched targets, DWBA analysis, resolution 100 keV; θ = 20-120°, $\Delta\theta$ = 4°.

NUCLEAR REACTIONS, FISSION ^{235}U(n,F), E = 1-1000 eV; measured σ(E), (fragment) (fragment) (E;θ).

FIG. 1. Examples of keyword strings.

At the Data Project, other types of sorting are also generated. These include listings by specific type of reaction [e.g., (d, p), (n, α), etc.] and by topic [e.g., $T_{1/2}$, B(E2), Doppler shift, polarization, etc.].

In addition to using the system to publish Recent References, the Data Project provides selected reference lists periodically to the following.

Table of Isotopes (J.M. Hollander and C.M. Lederer, USA) magnetic tapes
Atomic Mass Adjustment (A.H. Wapstra, The Netherlands; N.B. Gove, USA)
Nuclear Moments (G.H. Fuller and V. Shirley, USA)
Photonuclear Cross Section Center (E.G. Fuller, USA)
Nuclear Information Research Associates (22 compilers, USA)
Non-Project Compilers (H. Verheul, The Netherlands; S.C. Pancholi, India; A. Artna-Cohen, USA; B.S. Dzhelepov and L.K. Peker, USSR)
National Neutron Cross Section Center (BNL, USA)

The manner in which the Project interacts with these compilers/evaluators and data producers and users is depicted in the second figure.

The Project also scans and writes keywords for numerous unpublished literature that it receives, and maintains a separate (secondary sources) file which is also used to inform most of those listed above. Numerous selected reference lists have been provided on request.

Nuclear Data Compilations

Although compilations published in the Nuclear Data Sheets are primarily oriented toward the basic researcher working in nuclear structure, much of their content has applied usage as is evident from the contributions to this symposium. In the following, we will attempt to present an indication of what is entailed in such a compilation.

A typical mass chain compilation involves consideration or some 200 or more papers which deal with a variety of topics and experimental techniques in nuclear physics. We have made no attempt to estimate the actual quantity of numerical data that must be considered by the compiler in doing such a compilation, but it is certainly staggering.

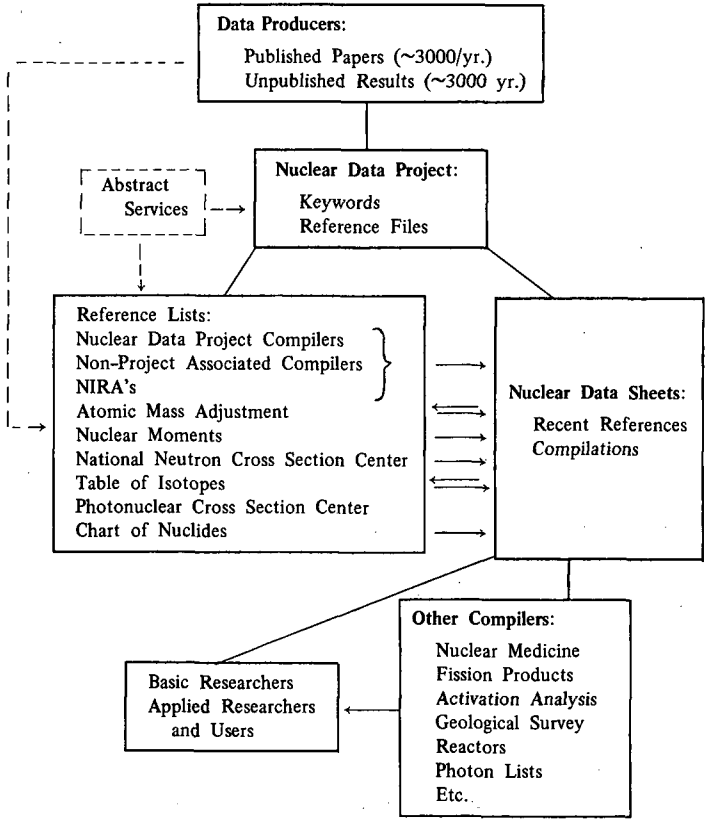

FIG.2. Efforts of Nuclear Data Project in effecting communication between producer, compiler/evaluator, and users of nuclear data.

The function of the compiler is to "thoroughly" examine and "evaluate" the data content of these papers, correlate the results obtained from different experimental methods and present a consistent and "best" set of data describing nuclear structure characteristics. In addition, he should clearly denote discrepancies in the data, as well as focus attention on those areas for which data are lacking. In view of the complexity and magnitude of such a task, the compiler is forced to make various compromises such as amount of time he should expend on any given paper, how much reanalysis is warranted, etc., and this depends largely upon the details of specific experimental techniques, the type and quality of data involved, and the background of the compiler. Obviously, the real value of such a compilation to any user depends very much upon his particular application as well as his understanding of the effort that went into producing the compilation. This is true even though he may only be interested in a set of "best" values.

By thoroughly reading a paper, the compiler usually acquires information that assists him in his evaluation. In addition, he is liable to discover errors

and/or inconsistencies that might have gone undetected had he only extracted the data from tables or figures contained therein. Unfortunately, it is not uncommon to find comments in the text which sometimes reflect upon the interpretation of the tabulated data. In numerous instances, the compiler's conclusions can differ from those of the author due to the availability of information which may not have been available to the latter, or possibly due to bias on the part of the author. It might also be noted that in the evaluation effort for the Data Sheets use is made of a variety of theoretical tools (e.g., theoretical conversion coefficients, nuclear models, ϵ/β^+ ratios, etc.). These may also lead the compiler to conclusions which differ from an author's.

In view of the broad interest in the properties of nuclear energy levels (e.g., energy, half-life, spin and parity, etc.) and their decay (e.g., modes of decay, energies, branching ratios, etc.), particular emphasis is placed upon these quantities. For example, the compiler meticulously examines the decay schemes as well as the bases for absolute normalizations. Trying to determine a legitimate normalization factor can sometimes prove rather frustrating, especially if there is direct decay to the ground state of the daughter. The main reason for this is that in those cases a reasonably accurate measurement might involve considerable effort on the part of the measurer. If the latter is a nuclear structure scientist (which he usually is), he might conclude that the amount of physics to be derived may not be worth the work required. For example, most of the structure physics can usually be learned from just a measurement of the energies and intensities of the photons following radioactive decay. In many cases, an accurate (better than 10%, e.g.) measurement of the normalization factor provides little additional insight as far as the physics is concerned. However, the absolute photon branches might be of crucial importance to the applied worker. It is not uncommon to find that in some cases there is little or no justification for an adopted normalization. In fact, compilers have, in a few cases, found incorrect normalizations that were propagated for some years. With the decrease in the number of research groups interested in measuring beta spectra, we would expect the situation to worsen, especially as regards the applied user, in those cases for which there is direct decay to the ground state.

Another aspect of mass chain compilations, which might be of interest here, is the process of the correlation of the results obtained from measurements of different types. For example, the results from reaction studies might reveal a different photon branching ratio than that inferred from radioactive decay. Should it be concluded that the former was correct, this would require reexamination of the absolute photon branchings in the decay. In some cases, spin and parity information derived from reaction studies might have significant bearing upon interpretation of the decay data. The point that we wish to make here is that if one is really interested in the "best" values, it may be extremely important to examine all existing data that may have a bearing on such values.

The relative importance of any of these possible situations to the applied user certainly will depend upon his intended application.

Besides presenting "best" decay schemes, the compiler also correlates the data and presents "adopted" level energies and properties for each nuclide. Included are the experimental bases upon which spins and parities are assigned. During the course of the compilation, he also checks the input data used by Wapstra and Gove[1] in their compilation of atomic masses.

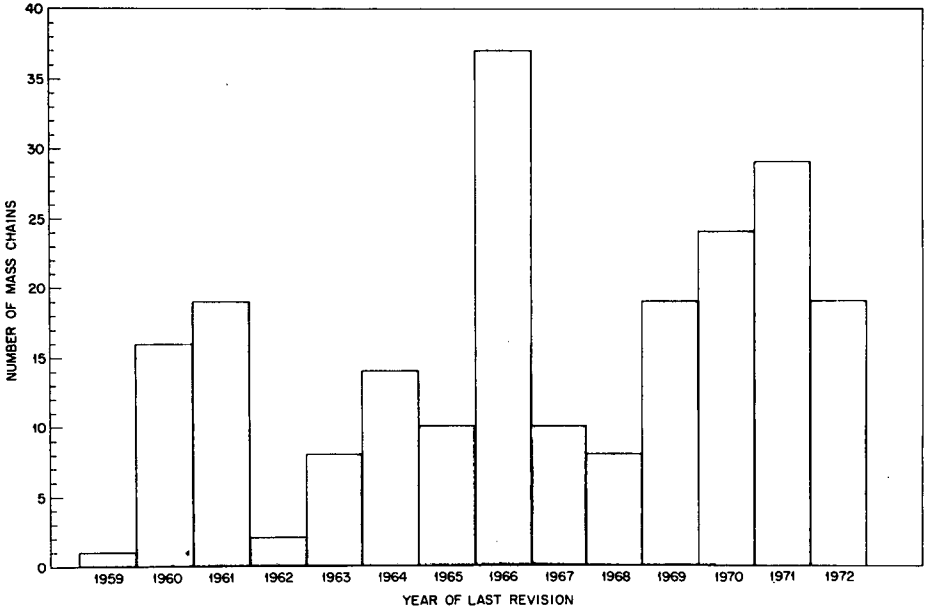

FIG. 3. Number of mass-chain compilations for A ≥ 45 versus year of last revision.

Status

In the third figure we show the number of mass chain compilations for $A \geq 45$ versus the year in which they were last revised. A current index to mass chain compilations is included in each issue of the Nuclear Data Sheets. The Nuclear Data Project presently has a staff of about eight compilers, and we attempt to revise about 25 mass chains per year. In the fall of 1971, a program was initiated by an ad hoc committee of members of the Division of Nuclear Physics of the American Physical Society which obtained funds from the National Science Foundation to support approximately 48 man-years of effort (Nuclear Information Research Associates or NIRA's) to assist the Data Project in updating these compilations. Hopefully, by the end of 1974 most of the mass chains will have been revised and be current within a three or four year period.

Users and Future Plans

Basically, one can categorize the various users of nuclear data into two broad groups, e.g., those involved in basic research (both experimental and theoretical) and those in applied areas. Furthermore, in each of these categories there exist varying degrees of competence (or for that matter, even interest) to self-evaluate the experimental data. There are few nuclear theorists, for instance, who have the experience to do so and in general they prefer to have experimentalists do the compilation and evaluation tasks. Likewise, the reactor engineer generally utilizes tables or files of data generated by others upon whom he depends for their reliability.

The specific needs of each user are usually somewhat different. For example, experimental physicists probably do not require an exhaustive compilation which results in "best" evaluated data. However, this might be crucial for the nuclear theorist or applied user. The quantities needed for specific users also vary considerably, and even the degree of accuracy required for the same quantity may vary, depending upon the intended usage. An example of the latter is the use of a radioisotope as a routine tracer in an industrial application for which the absolute photon intensities may be unimportant. When the same isotope is used in a medical application, the absolute radiation intensities may be essential.

Although the Nuclear Data Sheets contain evaluated data of broad usage, we realize that the presentation is not suitable for all users. For instance, in some medical applications it is not only important to know absolute photon and β-ray intensities, but also conversion electron, X-ray, and Auger intensities. It was in recognition of this fact that a compilation of these quantities was produced for some 105 commonly used radionuclides by M.J. Martin and P.H. Blichert-Toft.[2] The title of this compilation is "Radioactive Atoms: Auger-Electron, α-, β-, γ-, and X-Ray Data". Here, the authors present decay schemes as well as tables of all the radiations emitted. The latter includes energies and absolute branchings and their associated errors. Martin has recently compiled similar data for some 13 additional radionuclides, including many fission product gases.

During the past year, we have been responding to numerous requests for information varying from references to specific data from persons at a variety of establishments, including universities, government agencies, private industry, foreign scientists, etc. A few months ago, The Data Project announced the availability on magnetic tape of the 1971 Atomic Mass Adjustment by A.H. Wapstra and N.B. Gove.[1] We are presently exploring steps to establish links to persons interested in preparing radioactive decay data in a more useful format for medical usage.

Two points are obvious from these exchanges:

1. There exists a variety of workers who are interested in extensive radioactive decay data, as well as other nuclear structure information. The specific quantities, relative accuracies, and particular formats largely depend upon intended applications. These conclusions have clearly been supported by the contributions to this symposium.

2. An apparent need may exist to increase the direct communication between those applied users with little or no knowledge of the basic physics which underlies their usage and nuclear structure scientists and/or compilers/evaluators. It seems to us that as the applications of nuclear data in science and technology increase, some effort should be expended to help ensure its efficient and proper usage.

In recognition of the needs noted in point one, we have long felt that a versatile computer file of evaluated nuclear structure data would be extremely useful. Hence, we have begun working to generate such a file. The basic philosophy will be twofold:

1. The input–output should be simple, and therefore we plan to utilize a "card image" storage system which will be easily accessible and transferable.

2. The system should involve a minimum of inputting. To succeed in this respect, we are developing programs which will be able to operate on measured data in order to produce derived data. Consider for example radioactive decay. Usually, the most accurately measured data consist of photon energies and intensities. For most applied purposes, relative transition intensities can be calculated by using theoretical conversion coefficients. Assuming the level structure and absolute normalization factor are known, one can then generate various types of tabulated derived data such as absolute photon intensities, absolute intensities of all the radiations emitted, etc., and arrange this information in almost any desired format. Obviously, it would be trivial to produce drawings of complete or partial decay schemes in such a manner, and we have already begun to do so.

Now once the programming has been completed, an efficient mechanism will have been developed to transfer evaluated as well as raw data from producers to compilers/evaluators, and vice versa, on both a national and an international level. It is anticipated that such an evaluated data file would also be highly useful in the basic sciences for retrieving specific data as well as for horizontal type compilations.

Summary

We have tried to present a brief summary of the functions performed by the Nuclear Data Project, and how these interact with producers, compilers/evaluators, and users of nuclear data. In addition, we have tried to convey some feeling for the type of effort which goes into a Nuclear Data Sheet mass chain compilation, as well as to point out some of the realistic and mundane factors involved in the evaluation and usage of nuclear data. We have mentioned a possible mechanism (i.e., computerization) to improve the efficiency of data transmittal and reformatting. However, we would certainly be derelict if we did not at least note that it takes humans to ensure the proper production, compilation/evaluation, and usage of nuclear data.

References

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DISCUSSION

M. LAMMER: As a user of the Recent References issues of Nuclear Data Sheets, I find the keyword system very useful. However, sometimes when the indication "measured E_γ, I_γ" appears and I look up the reference I cannot find any data on E_γ and I_γ. Would it be possible to include in the keywords the notation, for example, "no data given"?

D.J. HOREN: We would appreciate being notified of specific cases where you find that the keywords do not properly reflect the content of a paper.

Z. SUJKOWSKI: With reference to your histogram (Fig. 3 of the paper) illustrating the revisions of the mass chains, what is the average recycling time, i.e. after what time would you recommend making a revision? What is the ultimate goal?

D.J. HOREN: We believe the most efficient way to handle this matter is to have one person with continuing responsibility for monitoring X mass chains and revising each as the need arises.

D. BERÉNYI: I would like to emphasize the importance of close collaboration between the compilers of nuclear and atomic data. This is particularly important in the case of X-ray data, where it is sometimes rather difficult to get the required information on X-ray intensities. The problem is complicated because the form (pattern) of an actual X-line (e.g. an L-line) will be different depending on the resolution and type of detector used (scintillation, semi-conductor, etc.). In the Martin-Blichert-Toft table, for example, the intensity of L-X-lines (^{241}Am) is assigned to one of the L-series, but if the measurement is made with a scintillation counter (as is very common in applied work), the energy of the L-peak will be at a quite different place (according to the weighted average of the lines in the L-series). All of this is important from the point of view not only of applied problems but also from that of basic research (X-rays generated by heavy ions).

H. GRUPPELAAR: Have you tried to convince the editors of journals other than Nuclear Physics to use the keyword system?

D.J. HOREN: Yes, we have, and Physical Review C is now planning to include them. We have written letters to editors of other journals also, but so far have not received positive replies.

H. GRUPPELAAR: I think it would be very useful if the authors of an article were asked by the editors to publish their data in a standard format, if possible, in order to facilitate evaluation tasks. Can you comment on this?

D.J. HOREN: Professor K. Way has written on this subject over the past ten years or so, and we as well as the IWGNSRD have brought the matter to the attention of editors. Physical Review C has accepted a few suggestions.

H. GRUPPELAAR: Your intention is to publish decay schemes based on computer programs. Is it also your intention to publish separate lists of γ-ray energies and intensities in the Nuclear Data Sheets?

D.J. HOREN: Essentially, we are already doing this in the Nuclear Data Sheets. This should become trivial when we develop our computer programs.

J.J. SCHMIDT: What are the chances of keywords being accepted by journals other than those you have already mentioned? Can their adoption by Physical Review C be expected to have a favourable impact in this respect? Also, are the keywords used by the ORNL Nuclear Data Project identical with those used by the Nuclear Physics and Physical Review C journals?

D.J. HOREN: Physical Review C is going to include keywords, which are essentially the same as those used in Nuclear Physics and by the Nuclear Data Project. It is to be hoped that this will help induce other journals to do likewise.

J.J. SCHMIDT: Are there any plans in the Soviet Union for adopting this keyword system, say, for Jadernaja Fizika or Atomnaja Energija?

I. A. KONDUROV: The policy of Soviet journals as regards keyword abstracts is discussed in my paper (SM-170/25).

G. A. KOLSTAD: I would like to ask a question with regard to Fig. 3 of your paper. You indicate that the NIRA program was started in 1971, yet Fig. 3 shows a drop in the output of mass chains by 35% in 1972. What is the explanation for this?

D. J. HOREN: In part it is due to our diverting some efforts to participation in the NIRA program. Moreover, the compilation of some mass chains is more time-consuming than that of others.

ПРИНЦИПЫ РАБОТЫ ЦЕНТРА ДАННЫХ ПО СТРУКТУРЕ ЯДРА В ЛЕНИНГРАДСКОМ ИНСТИТУТЕ ЯДЕРНОЙ ФИЗИКИ

И.А. КОНДУРОВ
Ленинградский институт ядерной физики
им.Б.П.Константинова,
Союз Советских Социалистических Республик

Abstract–Аннотация

PRINCIPLES OF OPERATION OF THE CENTRE FOR DATA ON NUCLEAR STRUCTURE AT THE LENINGRAD INSTITUTE OF NUCLEAR PHYSICS

The Data Centre at the Leningrad Institute of Nuclear Physics is intended to free research workers from routine tasks when handling data, whether the latter are the results of their own research or the findings of others. The basic principles of operation of the Centre may be summarized as follows: the establishment and constant expansion of an automated reference system on the structure of the nucleus and on elementary particles, with operational access to the data and to the system of programs for working therewith. It is intended to accumulate data and calculations within a framework of broad co-operation and exchanges with related regional centres.

ПРИНЦИПЫ РАБОТЫ ЦЕНТРА ДАННЫХ ПО СТРУКТУРЕ ЯДРА В ЛЕНИНГРАДСКОМ ИНСТИТУТЕ ЯДЕРНОЙ ФИЗИКИ.

Деятельность центра данных Ленинградского института ядерной физики (ЛИЯФ) направлена на то, чтобы освободить исследователя от рутинных операций при работе с информацией, причем имеется в виду как информация, полученная им самим в ходе исследований, так и результаты других авторов. Основные принципы работы центра данных могут быть определены следующим образом: создание и постоянное пополнение автоматизированного справочника по структуре ядра и элементарным частицам с оперативным доступом к данным и к системе программ для работы с данными. Накопление данных и работу по оценке предполагается проводить в рамках широкой кооперации и взаимного обмена с заинтересованными региональными центрами.

Прогресс современной науки в значительной мере зависит от организации системы информации, особенно в условиях экспоненциального роста объема научных исследований и при значительном возрастании заинтересованности специалистов в информации об исследованиях и разработках в смежных областях науки. Традиционные формы обмена информацией уже не удовлетворяют требованиям современной науки и техники. Это обусловило появление в течение последнего десятилетия большого числа научно-информационных центров, основной задачей которых является реализация более эффективного обмена количественно нарастающей научной информацией, чем это допускают печать и почта.

Ядерная физика и физика элементарных частиц не являются здесь исключением. Развитие реакторов и ускорителей, появление прецизионных полупроводниковых детекторов ядерных излучений и пропорциональных камер для регистрации заряженных частиц и, наконец, широкое внедрение компьютеров и автоматизированных измерительных систем в эксперимент обусловили интенсивный рост количества данных, которые необходимо быстро и полно доставить пользователям. Сказанное можно подтвердить небольшим частным примером. На рис.1 показано, как с годами меняется полное число ядерных уровней, времена жизни которых измерены различными экспериментальными методами [1].

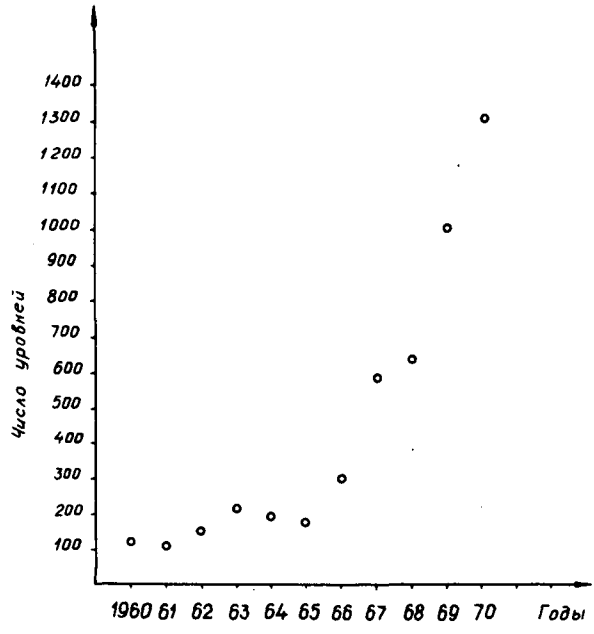

Рис.1. Изменение полного числа ядерных уровней, времена жизни которых измерены различными экспериментальными методами.

Перечисленные выше факторы определяют два нарастающих во времени информационных потока, которые должен переработать исследователь: результаты собственных экспериментов и данные, полученные другими авторами. Основной способ помочь исследователю в решении этой проблемы – освободить его от рутинных операций при работе с информацией. Это означает, что необходимо автоматизировать и максимально упростить:

а) поиск необходимых данных и

б) обработку полученных данных.

Эти две задачи и составляют основу деятельности центра данных по структуре ядра и элементарным частицам Ленинградского института ядерной физики им.Б.П. Константинова АН СССР.

Основная тематика института – фундаментальные исследования в области ядерной физики и физики элементарных частиц и использование ядерно-физических методов в физике твердого тела и радиобиологии – определила главную цель работы центра:

создание и постоянное пополнение автоматизированного справочника по структуре ядра и элементарным частицам с оперативным доступом к данным и к системе программ для работы с данными.

Под данными здесь понимается информация трех различных уровней:

1. систематизированные, оцененные и справочные данные;
2. данные конкретных работ (авторские оценки);
3. "сырые" экспериментальные результаты (спектры, распределения, таблицы, графики и т.д.).

Ниже обсуждается работа центра применительно к исследованиям по структуре атомного ядра.

Оцененные данные необходимы буквально на всех этапах исследовательского процесса. При изучении структуры ядра возникает потребность в следующих категориях оцененных данных:

1. ядерные характеристики – массы ядер, энергии переходов, схемы возбужденных состояний ядер, сечения различных реакций и т.д.;
2. атомные характеристики – энергии и интенсивности рентгеновских переходов, коэффициенты поглощения излучений в веществе, коэффициенты внутренней электронной и парной конверсии и т.д.;
3. фундаментальные константы – скорость света, гравитационная постоянная и т.д.

Эти данные накапливаются центром на магнитных лентах (МЛ) и будут предоставлены пользователям в режиме прямого доступа.

Особое внимание уделяется накоплению данных по схемам распада радиоактивных ядер и данных о структуре ядерных уровней из реакций. В настоящее время эти данные сконцентрированы, в основном, в сборниках Джелепова и Пекера [2], Ледерера и др. [3] и в "Nuclear Data Sheets" [4], однако во всех случаях отставание в оценках составляет сегодня в среднем несколько лет. Ясно, что ликвидировать это отставание возможно лишь в рамках международного разделения труда по оценке свойств ядерных уровней и по приведению этих данных к машинному формату. Центр данных ЛИЯФ, работая в контакте с советскими группами по оценке и при тесном сотрудничестве с центром по ядерным данным Института атомной энергии им.И.В.Курчатова [5], записывает на МЛ имеющиеся на сегодня данные по схемам уровней и готов принять участие в любой международной кооперации по этому вопросу.

Данные конкретных работ представляют значительный интерес при проведении исследований и разработок, однако в настоящее время не представляется возможным хранить эти данные в ЭВМ. Центр предоставляет пользователям возможность автоматического поиска интересующих их данных по ключевым словам, причем как по отдельным запросам, так и в режиме избирательного распределения информации.

В основу нашей информационно-поисковой системы положено аннотирование в ключевых словах, осуществленное в журналах "Nuclear Physics" и в "Nuclear Data Sheets". Эта система ключевых слов существует уже давно и к ней привыкли.

В настоящее время центры данных ЛИЯФ и ИАЭ совместно аннотируют в ключевых словах все советские периодические издания, в которых публикуются работы по структуре ядра, и записывают эти данные на МЛ в согласованном формате. Явным преимуществом такого регионального аннотирования является то, что информационные документы могут быть обработаны еще до их опубликования, пока они находятся в портфеле редакции. Так, например, было проведено аннотирование и запись на МЛ в ключевых словах тезисов докладов на XXIII ежегодном совещании по ядерной спектроскопии и структуре атомного ядра (Тбилиси, 1973 год). Препринт с аннотациями вышел в свет одновременно со сборником тезисов [6].

Нам представляется, что такое региональное аннотирование, если оно проводится в рамках международной кооперации с обменом информацией на МЛ, поможет преодолеть коммуникационные и языковые барьеры,

которые стоят сейчас на пути скорейшего доведения свежей информации до потребителя.

Сырые экспериментальные данные, вообще говоря, представляют наибольший интерес для исследователей, решающих одинаковые проблемы. Существенно, что эти данные нужны относительно небольшому числу исследователей, но им нужны максимально полные и именно "сырые" данные, так как они содержат первичную информацию, не искаженную окончательной обработкой и субъективными авторскими оценками. Однако, при резком возрастании объема сырой информации существует очевидная тенденция сокращать эти данные в публикациях из-за ограниченного объема последних.

Использование машинных носителей информации снимает ограничения, связанные с объемом экспериментальной информации, и позволяет осуществить эффективный обмен данными, однако здесь возникают трудности, определяемые необходимостью выработки единого международного обменного формата. Эти трудности усугубляются тем, что даже при наличии такого формата эффективный обмен будет возможен лишь тогда, когда сами исследователи примут и будут широко и свободно использовать этот формат.

Центр данных ЛИЯФ ведет работу по выработке формата для записи на МЛ полных результатов работ, которые ведутся в институте.

Обработка "сырых" экспериментальных результатов в исследованиях по структуре ядра может быть условно разделена на два этапа:

1. Учет характеристик экспериментальной установки (декомпозиция спектров, анализ распределений и т.д.) с целью получения характеристик ядер (энергии, спины, четности уровней, мультипольности переходов и т.д.) и построения схемы возбужденных состояний ядра.

2. Сравнение полученных значений величин с предсказаниями в различных модельных предположениях.

Оба эти этапа в настоящее время проводятся с использованием ЭВМ, однако программы обработки часто разбросаны и не сопрягаются друг с другом, и очень часто между исследователем и ЭВМ стоит программист, который служит, по существу, тормозом в процессе взаимодействия исследователя с полученной им информацией.

Центр данных ЛИЯФ ведет работу по созданию системы сопряженных и простых в обращении программ для анализа экспериментальных распределений, построения схем уровней ядер по результатам экспериментов и для сравнения полученных характеристик ядер с предсказаниями различных ядерных моделей.

В заключение следует еще раз отметить, что только международное разделение труда, столь характерное для науки, сможет двинуть вперед дело организации всестороннего и эффективного обмена научной информацией.

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DISCUSSION

W. BEDERT: On the subject of keywords and the establishment of a standard system, I think it would be very important to take into account the valuable work which Chemical Abstracts has done in this respect.

Miss K. WAY: I would like to make a comment on the origin of the keyword system. The idea was first developed in 1961, I think, in Vienna at the IAEA in discussions between Professor Buras of Poland and myself. These discussions were initiated by Professor Buras and, although not a nuclear physicist, he contributed much to the idea and the specific plans which were worked out in very much the same form as that used today. I would like here to give recognition to his contributions.

J.J. SCHMIDT: In his presentation Dr. Kondurov called for the development of a single international computer format for nuclear structure data. I think it is quite legitimate to ask what are the chances of success for such an undertaking. As far as I know, apart from international documentation systems, the only existing data information exchange in computer medium so far is in the neutron data field. I am referring here to EXFOR. The development of this system took many years of effort by all the parties involved; it is now functioning reasonably well and on a truly international scale in exchanges between the neutron data centres at Obninsk, Saclay, Brookhaven and Vienna. It might be useful to point out that it required agreement on the part of the Obninsk centre to use Western magnetic tapes and tape units and to adopt keywords, symbols and conventions in the English language. This is one of the problems that had to be faced and could be successfully solved. I feel therefore sure that similar problems, once they arise, could also be overcome in the field of nuclear structure data.

Finally, I would like to ask Drs Kondurov, Chukreev and Horen what types of computers they use or are going to use for their computer systems of evaluated data.

I.A. KONDUROV: I think the difficulties involved in achieving an international format are well known to everyone here, so there is hardly any need to discuss them. On the technical side, we think, like Dr. Schmidt, that use can be made of the wide experience of the IAEA and the four international centres. As regards questions of format, we think these can be solved by international agreement.

In reply to Dr. Schmidt's question, we are planning to use computers which operate on-line, i.e. in the conversational mode, similar to those which other countries are experimenting with. This is a slower system, relatively speaking. We are using the BECM-6 computer which has considerably greater possibilities for work in FORTRAN.

F.E. CHUKREEV: I would like to supplement briefly the remarks of Mr. Kondurov. For data recording, processing and retrieval and for

solving a number of problems in connection with compilation, we are at present using a PDP 15.20 computer. Eventually, we are hoping to get a new, and perhaps better, machine, which will be connected with the BESM-6 at our Institute.

M. LEDERER: It should be one of the major advantages of computers that format conversions are easy to do. We at Berkeley have found this to be true, using Control Data computers at our laboratory.

L. HJÄRNE: When we speak about problems with agreements on formats, we are not really concerned so much with difficulties in connection with computer format as – and this has certainly been our experience in the neutron data area – with difficulties in achieving agreement on content. Agreements on all the various keyworded items are very hard to reach.

W.B. LEWIS: I would like to ask the Agency's scientific secretaries if the facilities established by the Agency could not be used to satisfy an international desire for a common format for nuclear structure and decay data. Would not the International Nuclear Data Committee (INDC) be an appropriate channel?

J.J. SCHMIDT: International links have been established, e.g. in the EXFOR system, which I have already mentioned, and in the neutron reference system CINDA. In principle, depending on the availability of manpower and funding, I can certainly foresee that some co-ordination assistance by the IAEA would be feasible. Dr. Lewis also mentioned the INDC, which is the advisory body to the Agency in matters of all nuclear – not only neutron – data. Perhaps Dr. Kolstad as a member of INDC would like to comment at this point.

G.A. KOLSTAD: I agree with Dr. Lewis' statement that, with the INDC the mechanisms already exist within the Agency to implement further international co-operation in the non-neutron nuclear data field. The two problems that arise will be, first, organizing the INDC in such a way that it will be able to cope with this much broader problem and, second, providing the Agency with additional staff to cope with this problem. This latter difficulty is further increased by the recent devaluation of the dollar.

J.J. SCHMIDT: I should like to add that, in the case of EXFOR, the centres involved had to hold technical meetings in order to arrive at the desired detailed agreements. Without making a commitment on the Agency's part at this moment, it is conceivable in my opinion that similar technical meetings of centres and groups engaged in the compilation and evaluation of "non-neutron" nuclear data would be able to develop the kind of international system for nuclear structure data that Dr. Kondurov was asking for.

D.J. HOREN: Our first objective is to develop computer programs to assist the compiler in doing his compilation, and in reducing the amount of such routine work as number manipulations, etc. Our second purpose is to build a nuclear structure file. We would not like to get bogged down in endless conversations on formats and the like. Rather, we want to develop a system which we find acceptable and try to make it useful for others.

Section XIII

LARGE-VOLUME COMPILATIONS

Chairman

A.H. WAPSTRA (Netherlands)

NEUTRON-CAPTURE GAMMA-RAY COMPILATIONS

G.A. BARTHOLOMEW
Chalk River Nuclear Laboratories,
Atomic Energy of Canada Limited,
Chalk River, Ontario, Canada

Abstract

NEUTRON-CAPTURE GAMMA-RAY COMPILATIONS.

Neutron-capture gamma-ray compilations are discussed with particular attention to the Chalk River–Kurchatov compendium. Possible future developments are considered in relation to applied needs.

We shall begin by reviewing the present status of neutron capture compilations and next discuss their adequacy in meeting both fundamental and applied needs. Finally some ways of effecting improvements will be suggested.

Recent major (n,γ) compilations are listed chronologically in Table I with an indication of the scope and motivation of each work. The Oak Ridge National Laboratory Nuclear Data Sheets[8] have not been included although they embrace all (n,γ) data in their broad coverage. Several earlier (n,γ) compilations[9-11] are also not included.

Entry (1) is a comprehensive compilation of (n,γ) data from all sources available at the time of writing both published and unpublished. It includes tables of energies, E_γ, and absolute intensities, I_γ, of resolved lines from the more complete tabulations by different experimenters, decay schemes, representative graphs of the better measured spectra, and graphs of absolute yields as a function of E_γ for elements throughout the mass table. References are given to other (n,γ)-type measurements such as coincidence and angular correlation work and to early measurements not tabulated. No evaluations, except the minimum necessary to present the most complete and self-consistent decay scheme for each spectrum, were carried out.

Entry (2) is a compilation of absolute γ-ray yields in tabular form for Na, Al, Si, S, Cl, K, Ca, Ti, Fe, Ni, Cu, Zn, Ba and stainless steel as measured by the authors with a large NaI detector. Although individual lines are not resolved, the spectrum envelope was analyzed with high-resolution data from other authors as a guide, and individual line intensities and intensities summed over 0.5 MeV intervals extracted. The yield data from other authors are presented similarly for comparison.

Entry (3) gives, in tabular form, E_γ and I_γ for resolved γ-rays from 75 stable, natural, elements from Li to Bi inclusive (omitting gases except nitrogen). The results are the authors' own measurements with a Ge(Li)-NaI coincidence detector system operated in Compton-suppression and pair spectrometer modes. Line recognition and measurements of γ-ray energies and intensities were carried out from the original data, by computer.

Entry (4) is an extension of (3) containing in addition to discrete line information, an analysis of the unresolved continuum. The yields

TABLE I. NEUTRON CAPTURE GAMMA-RAY COMPILATIONS

Entry	Compilation Title	Authors	Date	Content	Source and Motivation	Ref.
1	Compendium of Thermal Neutron Capture γ-Ray Measurements:			Energies, intensities of resolved γ-rays, yield distributions, decay schemes, all elements, some isotopes.	Authors' own data plus literature search; fundamental orientation.	
	Part I, $Z \leq 46$	G.A. Bartholomew, et al.	1967			1
	Part II, $47 \leq Z \leq 67$	L.V. Groshev, et al.	1968			2
	Part III, $68 \leq Z \leq 94$	L.V. Groshev, et al.	1969			3
2	Gamma-Ray Spectra Arising from Thermal-Neutron Capture in Elements Found in Soils Concretes, and Structural Materials.	R.E. Maerker and F.J. Muckenthaler	1969	Gamma-ray yields as function of energy for individual lines and summed in 0.5 MeV intervals, 14 materials.	Authors' own results plus some data from literature; applied orientation.	4
3	Thermal Neutron Capture Gamma-Ray Spectra of the Elements.	N.C. Rasmussen, et al.	1969	Energies, intensities of resolved γ-rays; all stable non-gaseous natural elements Li to Bi plus N.	Authors' own data; mostly applied orientation.	5
4	Line and Continuum Gamma-Ray Yields from Thermal-Neutron Capture in 75 Elements.	N.C. Rasmussen, et al.	1970	Item 3 plus total yields, including continuum in 0.25 MeV intervals.	Authors' own data; applied orientation.	6
5	Compilation of keV Neutron Capture Gamma Ray Spectra.	J.R. Bird, et al.	1973	Energies, intensities of resolved γ-rays, spectra, decay schemes, 42 elements F to Bi, neutron energies 5-300 keV.	Authors' own data plus literature search; fundamental orientation.	7

for discrete lines, for the continuum and their sum are tabulated in 0.25 MeV intervals of E_γ, and the original γ-ray spectra are shown for each element. The total yield has also been written onto magnetic tape in ENDF format.

Entry (5) is a compilation of all measurements of E_γ and I_γ of resolved lines in tabular form for capture of neutrons with energies $5 \leq E_n \leq 300$ keV in 42 elements between F and Bi, inclusive. Thermal capture γ-rays are listed for comparison, and relevant (d,p) reaction data are included in the tables. Decay schemes are given for resolved resonances and typical spectra are given to show the quality of the original measurements.

The motivations for preparing these five compilations differ as indicated in the sixth column of Table I. Compilations (1), (3) and (5) give prominence to high-resolution and high-precision energy measurements, (1) and (5) being primarily, but not entirely, motivated by interests in level structure and other matters in fundamental nuclear physics while (3), motivated largely by applied requirements, is also of great value to fundamental physics in supplementing and confirming the high precision data in (1). On the other hand, the principal objective in compilations (2) and (4) is to provide complete γ-ray yield information with the fine details of the spectra or their use in nuclear physics of secondary concern.

On the subject of compiler motivation, the Kurchatov-Chalk River Compendia, entry (1), have attracted interest as an international compiling program and a short digression to outline how this developed may be of value.

Both Groshev and his colleagues at the Kurchatov Institute and the Chalk River (n,γ) group had simultaneously, about 1958, produced (n,γ) compilations[12,13] primarily to consolidate and summarize large bodies of data that had been produced at each laboratory. For the Chalk River group, it was partly the urge to see this body of work collected together; partly the need to make minor updates and extensions (not otherwise easily published) to earlier data; and also, of course, partly the desire to provide a convenient data package for other users, that stimulated the initial compiling work. For completeness, available data from other authors were also included. Because of a strong interest in comparing results of different authors, a listing of all known measurements at face value was preferred to an evaluation or synthesis. About 1966, both groups independently considered it time to update their respective compilations and, upon realizing that duplication was about to recur, decided to combine efforts. With each group benefiting from the experience of the earlier compilations, it happened that the updated compilations, begun independently, were very similar in format, and merging of the work was therefore remarkably easy. The whole merging operation was effected with the inter-change of only two rough drafts (one each way) and less than a dozen letters to settle all matters concerning content, format, referencing, and mode of publication. Because of the unpublished (private communication) data available to each group separately, the combined compilation was much more complete and up to date than either component would have been alone.

The main uses of (n,γ) compilations are listed in Table II. The largest group of users are undoubtedly the fundamental nuclear physicists while the largest applied group are the shielding designers. The third group, using neutron capture γ-rays for prompt activation analysis, is small. So far, not many applications have arisen in which this technique has advantages over other methods of analysis. The principal applications

TABLE II. PRINCIPAL USES OF NEUTRON CAPTURE γ-RAY DATA

1. Fundamental nuclear physics
2. Radiation shielding design
3. Prompt activation-analysis

developed to date are for measurement of concentrations of certain elements in coal and other ores[14]. For prompt activation analysis, as in the usual kind of activation analysis, one must know the energies and absolute intensities of at least the stronger γ-rays in the spectrum.

Turning now to the adequacy of the available compilations for satisfying the various needs, we consider first fundamental physics. A major consideration is to establish level schemes and for this one needs the most complete possible information on the energies and intensities of individual lines in spectra from single isotopes. The feasibility of measuring the (n,γ) spectra of a separated isotope depends on the cross section and availability of that isotope. In the Compendia comprising item (1), Table I, decay schemes for somewhat less than half of all possible isotopes are reported and most of these are far from complete. In order to appraise the present state of the art in resolving the spectrum into its individual lines, we make use of the comprehensive high resolution studies of Rasmussen et al.[6]. These authors deduced for each element the fraction of the total energy emitted that appears as resolved γ-rays when measured with their spectrometer which had a resolution of 0.5% at 1 MeV and 0.1% at 7 MeV. Their results, renormalized so that 100% corresponds to emission of the neutron binding energy once per capture, are plotted as a function of atomic number in Fig. 1. Although the spectra are, for most cases, 100% resolved below Z=30 (zinc) there are regions in heavier elements where only a small fraction is resolved, e.g. only 2% at Z=63 (europium). There are regions where nearly 100% is resolved at the closed neutron shells near Zr, Ce, and Pb. It would appear, then, that the tables are far from complete from the point of view of the fundamental physicists but rectifying this is a job for spectroscopists, not compiler-evaluators. On the other hand, examination of the resolved γ-ray tables in item (1), Table I, for consistency shows wide discrepancies between the work of different authors, particularly in intensities, and instances where γ-rays are listed that would appear to be from contaminants or background are also not uncommon. It is clear, therefore, that there is also room for critical evaluation of the data now available.

Let us now consider whether the compilations in Table I satisfy the applied users, in particular the large community of shielding designers. We note in passing that the needs of the prompt activation analysts for strong, well-resolved lines, probably are adequately met for most elements by compilations (1) and (3). Two questions stand out:

(a) Are the γ-ray yield tables for thermal neutron capture sufficiently reliable and complete for shielding design?
(b) Is the coverage of resonance- and fast-neutron capture adequate?

For an assessment of reliability of γ-ray absolute yield data we turn again to Rasmussen et al.[6] who compared data from several experimenters

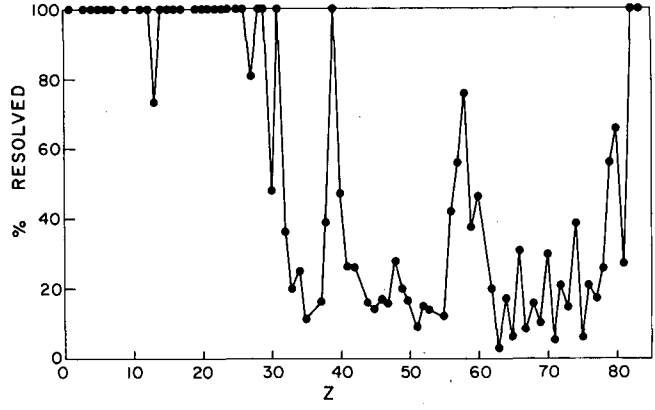

FIG.1. Percentage of total energy emitted that appears as resolved γ-rays in thermal-neutron-capture spectra measured with a Ge(Li)-NaI Compton-suppression/pair spectrometer [6]; renormalized data.

with their own results. Table III, for titanium, is one case considered in which all experimenters listed covered the entire energy range; we show Rasmussen's own results as originally measured, not renormalized, as he presented them. As can be seen from the last column, the fact that the neutron binding energy, BE, must be emitted once per capture provides a valuable check on the overall accuracy and completeness of the yield measurements. Clearly an uncertainty of ±10% or so in the overall yield is representative of this kind of data. Comparing yields in the individual 0.5 MeV intervals, one sees still wider variations, particularly for low intensity intervals. However, if the results of each group are renormalized so that $\sum_i E_{\gamma i} I_{\gamma i} = BE$, the agreement for individual intervals improves for all but a few of those at lower energies. This is shown in Table IV. The RMS deviation from the mean of all five entries is better than ±15% for all but the weaker-intensity intervals. This is consistent with precisions quoted by the experimenters. Maerker and Muckenthaler[4] estimate their accuracy for intensities in any interval to be ±15%. Other experimenters usually quote larger uncertainties for the stronger individual lines, e.g. 10-30% depending on energy (Bartholomew and Higgs[13] and ±20% Rasmussen et al.[5]) so that, when averaged over γ-rays in a 0.5 MeV interval, ±15% is probably representative. Accuracies of ±15 or ±20% are usually required by shielding designers submitting requests for (n,γ) yield data to RENDA. Therefore we must conclude that, at present, the accuracies available are adequate, but only just so.

It would not be appropriate here to review in detail the several contributory causes of these large intensity uncertainties but one particular point should be brought out: The relative intensity determinations depend strongly on knowing the efficiency function of the spectrometer. In all of the work in Tables III and IV, the detection efficiency at high γ-ray energies was established, at least in part, by calibrating on (n,γ) lines known from earlier absolute measurements made at Chalk River and Kurchatov laboratories. The crux of the problem is that, even today, there are few spectrometers with absolute, accurately-known, efficiency functions capable of completely independent intensity determinations and very few high energy γ-rays of known absolute intensity available for calibrating any other kind of spectrometer. The early Chalk River and Kurchatov data

TABLE III. MEASURED YIELDS OF THERMAL NEUTRON CAPTURE γ-RAYS IN TITANIUM[a], PHOTONS/100 CAPTURES

Experimenter Year	Gamma-Ray Energy Interval, $E_{\gamma i}$ (MeV)[b]																	$\Sigma E_{\gamma i} I_{\gamma i}$ %BE
	1.25-1.5	1.5-2.0	2.0-2.5	2.5-3.0	3.0-3.5	3.5-4.0	4.0-4.5	4.5-5.0	5.0-5.5	5.5-6.0	6.0-6.5	6.5-7.0	7.0-7.5	7.5-8.0	8.0-8.5	8.5-9.0	>9.0	
Groshev, et al.[12] 1958	85.0	25.7	5.2	3.0	6.7	5.7	0.6	9.9	0.0	0.8	27.5	42.0	0.6	0.3	0.2	0.0	0.2	93
Knowles, et al.[15] 1959	87.6	14.8	0.6	1.4	3.6	3.9	0.6	9.7	0.4	0.8	29.2	47.4	1.0	0.2	0.3	0.0	0.2	93
Draper & Bostrom[16] 1963	88.6	23.6	3.3	6.0	8.7			10.2		1.7	90.6		1.1	0.5	0.4			110
Maerker & Muckenthaler[4] 1969	83.8	25.7	3.6	5.0	6.2	4.6	0.7	9.5	0.4	1.0	33.1	56.0	0.9	0.2	0.3	0.0	0.2	106
Rasmussen et al.[6] 1970	69.3	24.0	4.4	3.6	8.5	6.1	0.9	12.0	1.2	1.6	37.9	61.3	0.4	0.0	0.2			117[c]

a) Derived from Table II ref. 6

b) Upper energy limit of each group not included in summation

c) Includes contribution below 1.25 MeV

TABLE IV. RENORMALIZED YIELDS OF THERMAL NEUTRON CAPTURE γ-RAYS IN TITANIUM, PHOTONS/100 CAPTURES

Experimenter Year	Gamma Ray Energy Interval (MeV)																	$\Sigma E_{\gamma i} I_{\gamma i}$ %BE
	1.25-1.5	1.5-2.0	2.0-2.5	2.5-3.0	3.0-3.5	3.5-4.0	4.0-4.5	4.5-5.0	5.0-5.5	5.5-6.0	6.0-6.5	6.5-7.0	7.0-7.5	7.5-8.0	8.0-8.5	8.5-9.0	>9.0	
Groshev et al.[12] 1958	90.9	27.5	5.6	3.2	7.2	6.1	0.6	10.6	0.0	0.9	29.4	44.9	0.6	0.3	0.2	0.0	0.2	100
Knowles et al.[15] 1959	93.7	15.8	0.6	1.5	3.9	4.2	0.6	10.4	0.4	0.9	31.2	50.7	1.1	0.19	0.32	0.0	0.20	100
Draper & Bostrom[16] 1963	80.5	21.4	3.0	5.5	7.9			9.3		1.5	82.4		1.0	0.45	0.36			100
Maerker & Muckenthaler[4] 1969	79.1	24.2	3.4	4.7	5.8	4.4	0.7	9.0	0.4	0.9	31.2	52.8	0.8	0.17	0.30	0.0	0.20	100
Rasmussen et al.[6] 1970	59.4	20.6	3.8	3.1	7.3	5.2	0.8	10.3	1.0	1.4	32.5	52.5	0.3	0.0	0.19	0.0	0.0	100

were not very accurate; for the Chalk River pair spectrometer efficiency function, uncertainties of ± 20% at 5 MeV, ± 10% at 7 MeV,[17] and unknown, probably ± 30% at 9 MeV[13] were claimed, all relative to unit efficiency at 2.76 MeV. To the extent that the calibrations on which many present yield measurements are based rely on that earlier work, those inaccuracies are still incorporated and these measurements which we have compared as independent determinations in Tables III and IV therefore may not be entirely independent. Agreement between the early Chalk River and Kurchatov intensity determinations, together with later relative intensity calibrations based on internal consistency of intensities in (n, γ) decay schemes in light elements [18, 19] have led to the conclusion that the accuracies of the relative intensities of the γ-rays usually used as intensity standards are better than ±15%, perhaps as good as ±5% below 7.5 MeV, but an exhaustive examination of the whole question, which would be basic to a thorough evaluation of (n, γ) intensities over the entire range to 11 MeV has not been made. A paper by Sokolovsky in these Proceedings may have more to say on this subject. In any uprating of compilations in Table I, particularly of item (1), that may be undertaken, some attention to intensity evaluations would be desirable. In the long term, there may be a need for further accurate absolute intensity measurements with an instrument not requiring external calibration. No spectrometer now available seems ideally suited to this task.

The γ-ray yield tables[4,6] together cover the full energy range and all targets of interest in reactor technology. An exception might be ^{135}Xe.

Turning next to the question of the adequacy of the resonance and fast neutron capture γ-ray compilation, entry 5 in Table I, it is quite obvious that although this is a most valuable summary of all currently available data, it does not meet requirements for fast reactor design. To name the principal omissions, it covers only 42 elements, it treats resolved lines with little information on the complete (resolved plus unreasolved) radiation yields, and it avoids the region below 5 keV neutron energy. The last omission arises from a practical compiling problem; many measurements in that region have been made with high resolution γ-ray spectrometers on individual resonances and the task of assembling and tabulating that detailed data would be monumental. The necessary poor-neutron-resolution measurements in the range 0–5 keV that would give γ-ray yields to 15% precision summed over resonances, and summed over 0.5 MeV intervals in E_γ, have not been made. Similar total yield measurements for energies above 300 keV are also largely lacking. The problems in providing the needed resonance and fast neutron capture γ-ray compilations are thus not so much problems of compiling and evaluating as of a shortage of the appropriate measurements. What really is most needed by shielding designers in the fast-neutron energy range are the neutron non-elastic-interaction γ-ray spectra which include neutron capture γ-rays plus γ-rays from inelastic neutron scattering. Most requests for γ-ray data in the 1972 issue of RENDA[20] are for this kind of information. An important step in satisfying such requirements, for the materials treated in entry (2) of Table I, has been made by Maerker and Muckenthaler [21].

To summarize, several recent compilations of thermal neutron capture γ-rays exist, some of which are oriented more to satisfying fundamental physics needs than applied needs. While the available high-resolution data do not satisfy all the wants of the fundamental users, the needs of applied users for thermal capture γ-ray total-yield data seem to be

adequately met for the time being. However, the uncertainties ∿±15% in the total yields appear only just adequate for shielding design and, while some headway in reducing these uncertainties by an exhaustive evaluation and review of intensity calibrations might be profitable, there seems to be a need for further accurate absolute yield measurements independent of calibration intensities from earlier capture γ-ray results. Much experimental work is yet needed to satisfy shielding design requirements for γ-ray yields in resonance- and fast-neutron capture.

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DISCUSSION

R. NICKS: I would like to confirm the need of shield designers for γ-ray data on non-elastic interactions in the fast neutron range. In the present situation, one is often forced to make approximations, such as introducing thermal capture γ-ray spectra into the calculations. I think that an accuracy of about 10 - 20% of the production cross-section should be enough for making heat calculations. Lastly, it would be interesting to take into account for the continuum contribution in the compilation.

G.A. BARTHOLOMEW: Yes, I agree. To my knowledge, very little of the kind of data you mention is available.

A.H. WAPSTRA (Chairman): Are you or anybody else working on updating the Bartholomew-Groshev compilation?

G.A. BARTHOLOMEW: We are contemplating this but all we can imagine doing in the foreseeable future is simply to update the existing compendia. Even for this I don't know where we will find the resources. To go any further and include evaluation is more than I would want to think about.

D.J. HOREN: Wherever (n, γ) data pertinent to a given mass chain are available, the compilers of Nuclear Data Sheets try to "evaluate" them prior to their inclusion. I use this term fairly loosely because evaluation of neutron capture gamma rays is a very difficult process.

G.A. BARTHOLOMEW: Yes. I should have mentioned in my presentation that, while I did not include the Data Projects work in my review, this is in fact the only work which involves some evaluation.

INTENSITE DES TRANSITIONS ELECTROMAGNETIQUES DANS LES NOYAUX (A > 40)

F. BERTRAND*, M. MARTINOT**, N. VERGES*
Commissariat à l'énergie atomique,
France

Abstract–Résumé

INTENSITY OF ELECTROMAGNETIC TRANSITIONS IN NUCLEI WITH A > 40.

Evaluations of spectra of neutrons and gamma rays emitted by nuclei are generally based on the use of a statistical model. For calculations one needs to know the partial widths Γ_n et Γ_γ relative to the emission of neutrons and gamma rays. There are fairly satisfactory ways (optical model) for estimating (Γ_n), but there is no single model for interpreting radiative widths correctly. The study published by Skorka et al. (Nuclear Data) for light nuclei ($A \leq 40$) showed in particular that the reduced transition probabilities B(E1) and B(E2) are, on an average, of the order of 0.0026 and 2.5 Weisskopf units. The present paper is complementary to that of Skorka et al. More than 10 000 data items on electromagnetic transition intensities in nuclei of mass number greater than 40 have been assembled, and the mean lives τ_γ, the reduced transition probabilities and the transition intensities (in Weisskopf units, with a statistical factor S = 1) have been deduced from them. A mean value of these quantities is proposed for each of the transitions studied. The frequency functions of transition intensities of given multipolarity and parity have been studied. The influence of the mass number on these frequency functions has been examined.

INTENSITE DES TRANSITIONS ELECTROMAGNETIQUES DANS LES NOYAUX (A >40).

Les évaluations de spectres de neutrons et de rayons gamma émis par les noyaux sont généralement fondées sur l'utilisation d'un modèle statistique. Les calculs nécessitent la connaissance des largeurs partielles Γ_n et Γ_γ relatives à l'émission des neutrons et des rayons gamma. On dispose de moyens d'estimation relativement satisfaisants (modèle optique) pour les premières (Γ_n). Mais il n'existe pas de modèle unique permettant d'interpréter correctement les largeurs radiatives. L'étude publiée par Skorka et al. (Nuclear Data) pour les noyaux légers ($A \leq 40$) a montré notamment que les probabilités de transition réduites B(E1) et B(E2) sont, en moyenne, de l'ordre de 0,0026 et 2,5 unités Weisskopf. Ce travail est complémentaire de celui de Skorka et al. Plus de 10 000 données sur les intensités des transitions électromagnétiques dans les noyaux de masse supérieure à 40 ont été rassemblées. Les vies moyennes τ_γ, les probabilités de transition réduites, et les intensités des transitions (en unités Weisskopf, avec un facteur statistique S = 1) en ont été déduites. Une valeur moyenne de ces quantités est proposée pour chaque transition étudiée. Les fonctions de fréquence des intensités des transitions de multipolarité et parité données ont été étudiées. L'influence du nombre de masse sur ces fonctions de fréquence a été examinée.

INTRODUCTION

Les évaluations de spectres de neutrons et de rayons gamma émis par les noyaux nécessitent la connaissance des largeurs neutroniques et radiatives Γ_n et Γ_γ. Parce que le champ électromagnétique est un champ plus faible que le champ nucléaire, les probabilités d'émission radiative sont beaucoup plus sensibles que les probabilités d'émission neutronique aux détails des configurations nucléaires. Pour cette raison, il n'existe pas de modèle simple permettant d'estimer – même à deux ordres de grandeur près – l'ensemble des largeurs radiatives. Pour pallier cette difficulté, il est apparu nécessaire de se fonder sur une large systématique concernant l'ensemble des noyaux.

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* Centre d'études nucléaires de Limeil.

** Centre d'études nucléaires de Saclay.

Plusieurs systématiques concernant des transitions de type déterminé ont déjà été publiées, notamment celles de Perdrisat [1] sur 143 transitions dipolaires électriques (E1) et de Stelson [2] sur 155 transitions quadrupolaires électriques (E2). Une étude plus générale, portant sur 547 transitions de types variés, a été publiée par Skorka et al. dans Nuclear Data [3]; elle concernait les noyaux de masse inférieure ou égale à 40.

BUT DU PRESENT TRAVAIL

La présente systématique est complémentaire de celle de Skorka: elle concerne les noyaux de masse supérieure ou égale à 40. Plus de 10 000 données sur les intensités de plus de 3000 transitions électromagnétiques ont été rassemblées. Pour limiter les risques d'erreur, il est nécessaire de procéder à de multiples vérifications sur la vraisemblance et la cohérence de ces données. Cette analyse n'étant pas tout à fait terminée, les résultats présentés aujourd'hui ne concerneront que 2503 transitions dans des noyaux classés par charge et masse croissantes entre l'argon-40 et l'erbium-170.

Pour chaque transition sont précisées: la charge et la masse du noyau, les énergies, spins, et parités des niveaux haut et bas, le caractère et la multipolarité de la transition, la méthode de mesure, la largeur radiative Γ_γ en électron-volts, l'erreur sur Γ_γ, l'intensité en unités Weisskopf, la probabilité de transition réduite B (en $e^2\,b^L$ pour les transitions EL, en $\mu_0^2\,b^{L-1}$ pour les transitions ML), la vie moyenne en secondes et la référence.

Si plusieurs données concernent une même transition, une valeur moyenne et une incertitude globale sur Γ_γ sont proposées, chaque donnée sur la largeur radiative étant affectée d'un poids égal à l'inverse de l'incertitude sur cette donnée.

CRITIQUE DE L'UNITE DE COMPARAISON

Pour analyser cet ensemble de données, il convenait de choisir une unité de comparaison incluant le maximum de ce qui est connu sur le noyau, mais contenant le moins possible de paramètres arbitraires.

On peut exprimer les largeurs radiatives au moyen de la relation:

$$\Gamma_{L,\pi} = \frac{8\pi\,(L+1)}{L[(2L+1)!!\,]^2}\left(\frac{E_\gamma}{\hbar c}\right)^{2L+1} B(L,\pi)$$

où $B(L,\pi)$ est la probabilité de transition réduite

$$B(L,\pi) = \frac{1}{2J_i+1}\,|\langle J_f \,||\, O_{L\pi} \,||\, J_i\rangle|^2$$

L'unité Weisskopf est obtenue en considérant les transitions d'un proton de la couche j_i, l_i vers la couche j_f, l_f en admettant:

- que le noyau est une sphère uniformément chargée de rayon $1,2\cdot A^{1/3}$ fermis
- que la partie angulaire de l'élément de matrice réduit (ou facteur statistique) a une valeur égale à 1.

L'influence de ces deux hypothèses a été examinée. En utilisant une distribution de charge trapézoïdale compatible avec un modèle en couche[1], soit avec un rayon moyen de $1,1\ A^{1/3}$ fm et une épaisseur de 3,2 fm, on a trouvé des renforcements maximaux de 1,13 pour les transitions E1, 1,35 pour les E2, 4,2 pour les E6, 1 pour les M1. En faisant varier dans des limites très larges le rayon moyen et l'épaisseur, on n'a trouvé que des renforcements très inférieurs à la dynamique des valeurs expérimentales sur les intensités des transitions.

Les facteurs statistiques ont été calculés en s'affranchissant des hypothèses restrictives habituellement consenties sur les moments orbitaux ℓ_i, ℓ_f.

Pour les transitions électriques:

$$S = (2L+1)\left|\left\langle J_i \tfrac{1}{2} L\,0 \middle| J_f \tfrac{1}{2}\right\rangle\right|^2 \left(1 - \mathrm{Mod}\,(\ell_i + \ell_f + L, 2)\right)$$

Pour les transitions magnétiques:

$$S = \frac{3}{2}(2j_f+1)(2\ell_f+1)(2\ell_i+1)\frac{(2L+1)^2}{L}(2L-1)$$

$$\times\left[\begin{Bmatrix} \ell_f & \frac{1}{2} & j_f \\ \ell_i & \frac{1}{2} & j_i \\ L-1 & 1 & L \end{Bmatrix} + \frac{(-)^{j_f+\ell_f+\frac{1}{2}}\sqrt{\frac{2}{3}(2j_i+1)(2j_f+1)}}{L(L+1)\mu\rho - L}\right.$$

$$\left.\times\begin{Bmatrix} \ell_f & j_f & \frac{1}{2} \\ j_i & \ell_i & L-1 \end{Bmatrix}\begin{Bmatrix} j_f & L & j_i \\ 1 & j_i & L-1 \end{Bmatrix}\right]^2$$

Si l'on se limite à la couche $j^{15/2}$, on trouve des facteurs S généralement de l'ordre de 1, avec les valeurs extrêmes suivantes:

Transition	S_{min}	S_{max}
E1	0,015	2
E2	0,036	3
E3	0,081	4
M1	0,0036	88,9
M2	0,26	60,0
M3	0,26	93,3

En se restreignant à des transitions pour lesquelles S est voisin de 1, on observe néanmoins une grande dynamique des intensités des transitions. Pour cette raison, et compte tenu du peu de réalisme du modèle à une particule, le facteur statistique n'a pas été introduit dans les calculs.

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[1] Beiner (Orsay) nous a aimablement fourni le programme de modèle en couche.

TABLEAU I. DONNEES SUR LA DISTRIBUTION DE LOG ($B/B_{Weisskopf}$) DANS DIFFERENTES ZONES (moyenne m, écart-type σ)

		E1	E2	E3	E4	E5	E6	M1	M2	M3	M4	M5
A – Co	n	38	256	65	25	3	2	177	12	7	0	1
	m	-3,42	0,15	0,18	-0,59	-0,32	-4,77	-1,61	-1,36	1,85		-1,89
	σ	1,27	1,29	0,92	1,03			0,97	1,04	1,91		
Ni – Mo	n	44	243	57	16	3	0	130	15	3	15	0
	m	-4,10	0,68	-0,38	-0,13	-0,31		-1,60	-1,70	-2,78	-0,46	
	σ	0,86	1,02	2,40	0,51			1,00	1,34	1,08		
Mo – Te	n	52	265	43	9	0	0	100	8	5	14	0
	m	-4,85	0,77	0,23	0,087			-2,01	-3,05	-0,85	-0,36	
	σ	0,99	0,95	2,11	0,86			0,90	1,46	1,35	0,70	
Te – Sm	n	23	223	24	3	1	0	114	10	1	17	0
	m	-4,33	0,88	0,98	0,84	3,73		-2,15	-1,22	0,40	-0,52	
	σ	1,49	1,05	1,58	1,20			1,19	2,40		0,81	
Sm – Er	n	70	245	67	0	0	0	76	19	1	1	0
	m	-4,94	0,95	0,18				-2,01	0,084	-1,38	-0,83	
	σ	1,85	1,34	1,58				1,87	2,65			
A – Er	n	227	1232	256	53	7	2	597	64	17	47	1
	m	-4,44	0,68	0,14	-0,25	0,26	-4,77	-1,83	-1,20	-0,037	-0,46	-1,89
	σ	1,50	1,17	1,81	0,96			1,18	2,20	2,32		

RESULTATS

Les données sur les intensités des transitions ont été examinées pour l'ensemble des corps et, séparément, dans cinq régions obtenues en classant les noyaux par Z et A croissants:

de ^{40}A à ^{60}Co; de ^{57}Ni à ^{94}Mo; de ^{95}Mo à ^{123}Te;

de ^{124}Te à ^{152}Sm; de ^{154}Sm à ^{170}Er.

Chaque fois qu'il était possible de le faire, on a, pour chaque région et chaque type de transition, calculé la valeur moyenne et la dispersion relatives au logarithme décimal de l'intensité. Un histogramme des intensités a été tracé en coordonnées logarithmiques. Le tableau I donne les valeurs moyennes m et les dispersions σ obtenues. Le nombre de transitions n est également précisé.

On constate que les transitions E1 sont très retardées. Le facteur de retardement moyen, de l'ordre de 40 pour les noyaux légers, varie ici d'environ 3000 à 1 000 000 suivant les zones.

Les transitions M1, retardées environ 10 fois pour les noyaux jusqu'à l'argon, le sont à peu près 40 fois de l'argon au tellure et 100 fois du tellure à l'erbium.

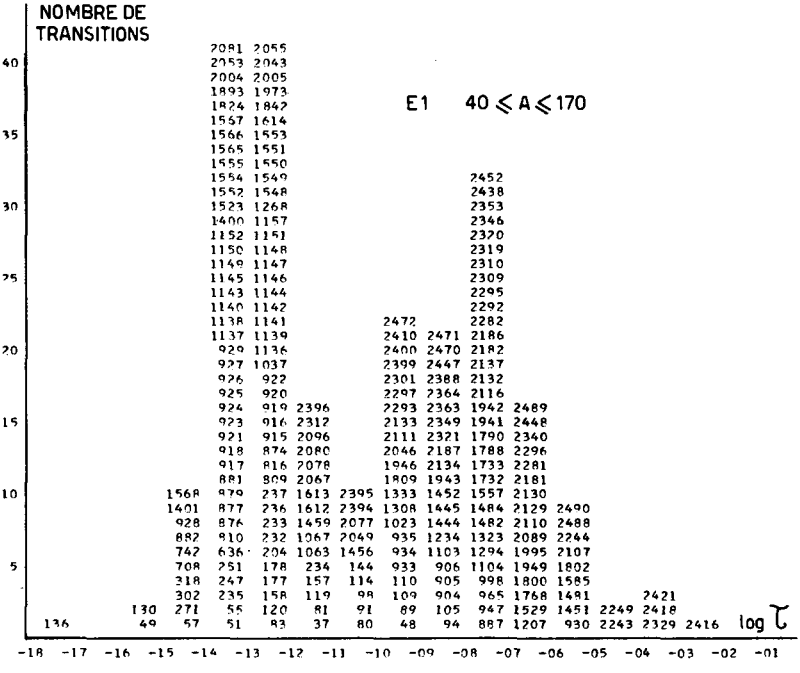

FIG.1. Histogramme des vies moyennes pour l'ensemble des transitions E1.

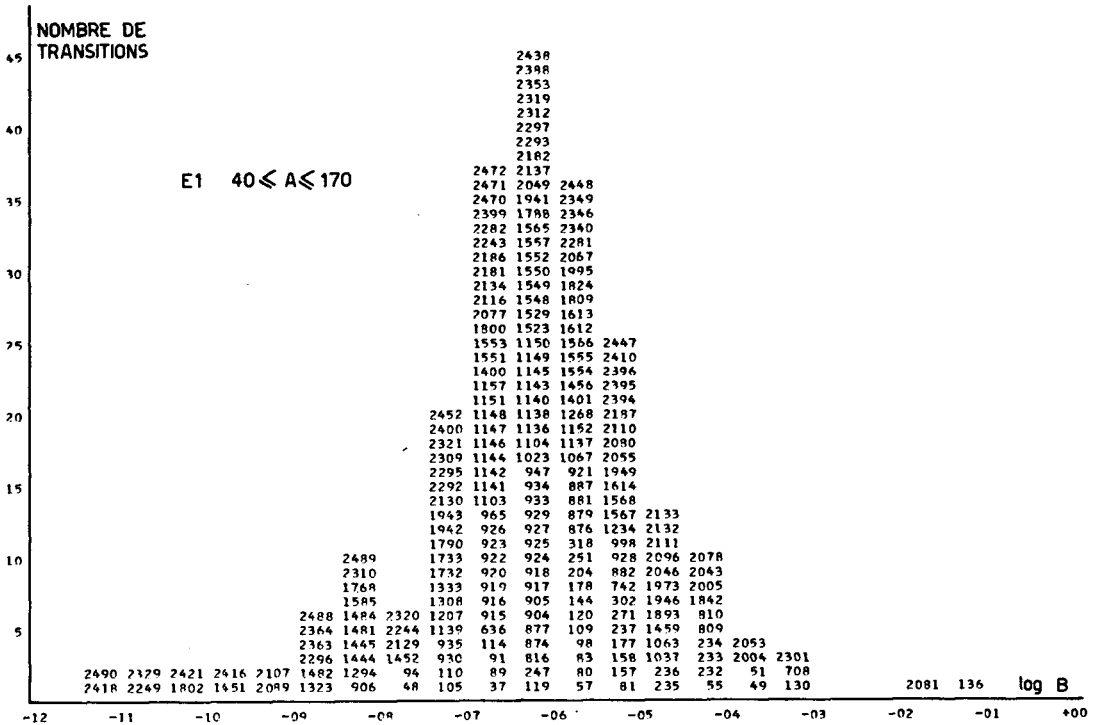

FIG. 2. Histogramme des valeurs des probabilités de transitions réduites B des transitions E1.

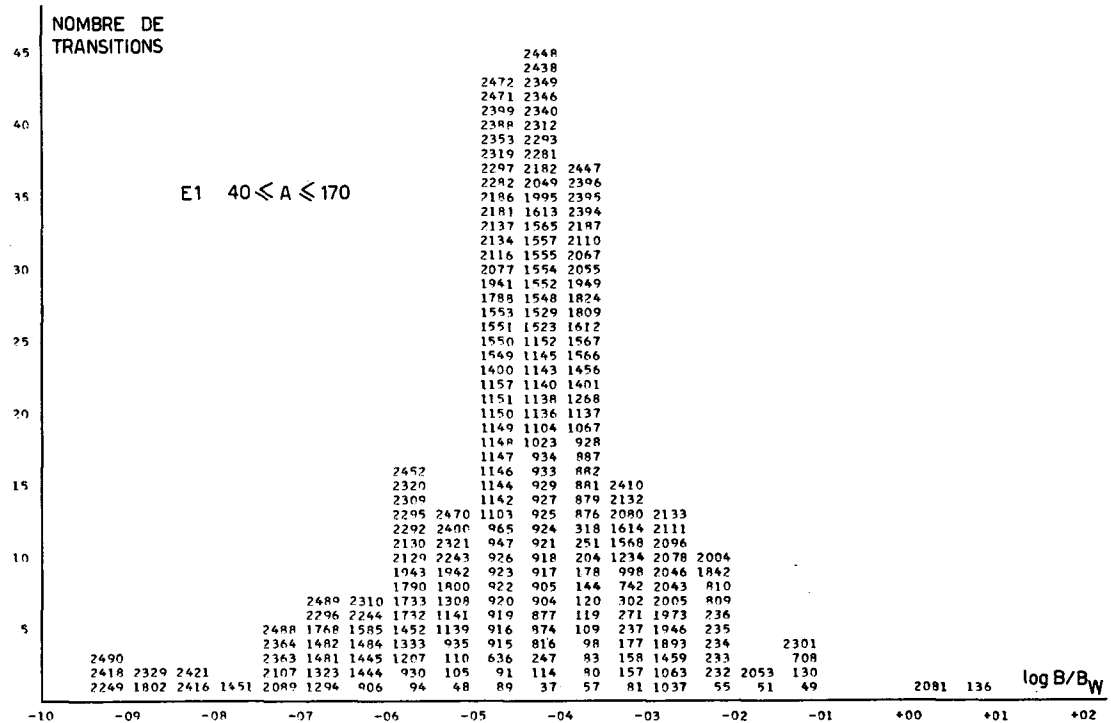

FIG. 3. Histogramme des intensités des transitions E1.

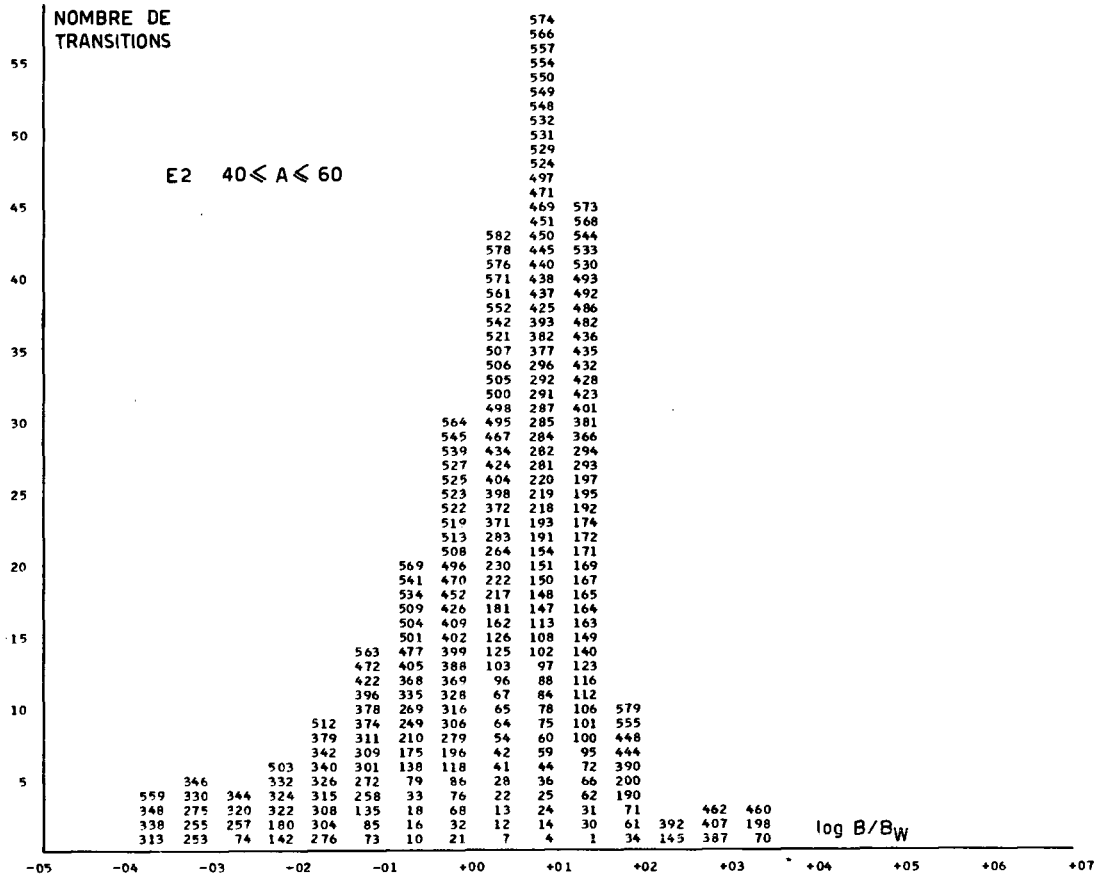

FIG.4. Histogramme des intensités des transitions E2 dans la région comprise entre l'argon et le cobalt.

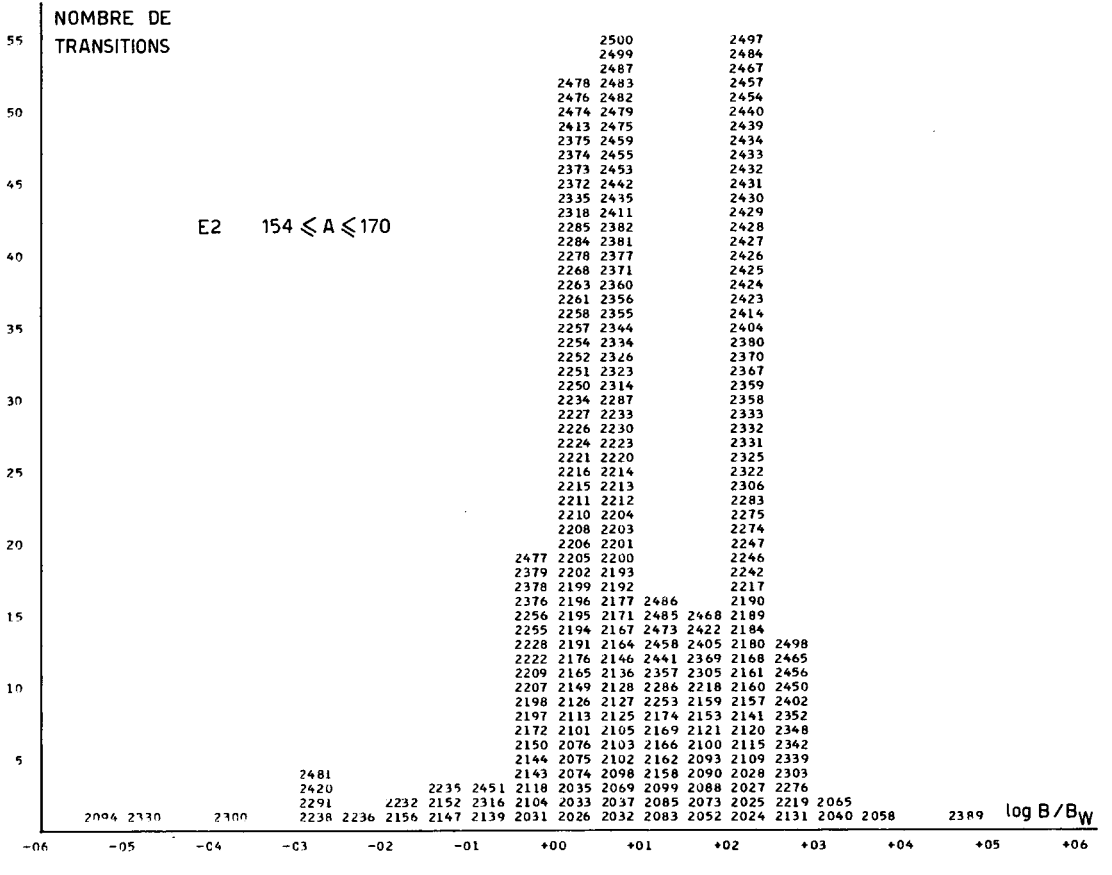

FIG. 5. Histogramme des intensités des transitions E2 dans la région comprise entre le samarium et l'erbium.

Les transitions M2 sont environ 15 fois retardées.

Par contre, les autres transitions sur lesquelles on possède une statistique significative ont un comportement assez proche des prédictions de Weisskopf.

Examinons maintenant quelques histogrammes concernant les transitions E1 et E2. La figure 1 montre l'histogramme des vies moyennes pour l'ensemble des transitions E1, chacune d'entre elles étant repérée par son numéro d'ordre. On constate une grande dispersion; l'écart-type sur le logarithme est de l'ordre de 3. Les structures observées ne sont pas très significatives, parce que les méthodes expérimentales de mesure des durées de vie τ privilégient certaines valeurs de τ, et que par ailleurs τ dépend fortement des énergies des transitions, lesquelles ne sont pas uniformément distribuées.

La figure 2 montre l'histogramme des valeurs des probabilités de transitions réduites B des transitions E1. L'écart-type sur log B est inférieur à 1,5, et les structures non significatives ont disparu.

La figure 3 montre un histogramme analogue pour les intensités des transitions E1. L'écart-type sur log $(B/B_{Weisskopf})$ est à peu près égal à celui sur log B. Ce fait est général. Il résulte de ce que le modèle à une particule est très sommaire et ne fait intervenir que des quantités variant lentement d'un noyau à l'autre.

La figure 4 montre l'histogramme des intensités des transitions E2 dans la région comprise entre l'argon et le cobalt. On observe une dissymétrie suggérant la superposition de deux distributions.

Les deux distributions se séparent nettement lorsqu'on aborde la zone des noyaux déformés; on peut le voir sur la figure 5, montrant l'histogramme des intensités des transitions E2 pour les noyaux compris entre le samarium et l'erbium. Le facteur de renforcement est de l'ordre de 3 pour le premier pic, 200 pour le second. Cette structure est significative d'un renforcement collectif.

L'influence d'autres paramètres tels que l'énergie de la transition, les spins des niveaux, leurs parités, celle du noyau, sera examinée au cours des prochains mois, sur l'ensemble des noyaux compris entre l'argon-40 et les transplutoniens.

REFERENCES

[1] PERDRISAT, C.F., Rev. Mod. Phys. 38 (1966) 41.
[2] STELSON, P.H., Nuclear Data 1 (1965) 1.
[3] SKORKA, S.J., HERTEL, J., RETZ-SCHMIDT, T.W., Nuclear Data 2 (1966) 347.

DISCUSSION

G.A. BARTHOLOMEW: I would like to know what energy range you covered. Did you limit yourselves to γ-rays of low energy from β-decay or did you include γ-rays of high energy from charged particle reactions and neutron capture?

M. MARTINOT: In order to have statistics with as little bias as possible, we used data obtained by a wide variety of methods: Doppler,

fluorescent resonance, β-γ coincidence, neutron capture, inelastic scattering of electrons and charged particles. The transition energies ranged from a few keV to 11 - 12 MeV.

A.H. WAPSTRA (Chairman): In your paper you use the shell model. Would not use of the collective model be more appropriate for the heavier nuclides?

M. MARTINOT: These systematics were intended for the use both of evaluators and physicists. It is true that the single-particle model is not very meaningful for deformed nuclei, e.g. in the region A = 152 - 194. When the systematics have been completed the data collected will be compared with those for a single-particle model when the latter is not too unsatisfactory and with collective models when these are more suitable.

For evaluations, on the other hand, we need a unit of comparison which, while remaining sufficiently simple and general, includes elements of the physics of gamma ray emission. The shell model was used for the purpose of defining this unit of comparison, not of comparing the data and a theory.

THE GAMMA-RAY SPECTRUM CATALOGUE – A USER DATA FILE*

R.L. HEATH
National Reactor Testing Station,
Aerojet Nuclear Company,
Idaho Falls, Idaho,
United States of America

Abstract

THE GAMMA-RAY SPECTRUM CATALOGUE – A USER DATA FILE.

The Gamma-Ray Spectrum Catalogue has been a continuing effort at the National Reactor Testing Station for the past ten years. The purpose of this effort is to provide a collection of experimental X-ray and gamma-ray spectra for general laboratory use in the analysis of data obtained with laboratory gamma-ray spectrometers. The Second Edition of the Catalogue, which was issued in 1964, contained over 300 pulse-amplitude spectra obtained with NaI scintillation spectrometers. These data represented an internally consistent set which was obtained under specified laboratory experimental conditions. Extensive text and supplementary material were supplied for general laboratory use of the techniques of gamma-ray spectrometry. The effort over the past several years has been to update this collection of data to include experimental spectra obtained with lithium-drifted Ge and Si detectors. The improved energy resolution of these detectors and associated techniques for analysis of complex gamma-ray spectra greatly increased the magnitude of this task. The current edition of the Catalogue, which contains experimental spectra of over 200 radioisotopes and extensive collections of gross fission product gamma-ray spectra as a function of irradiation time and decay, is currently in the printing process. This set of experimental data has been analysed to obtain precision values for gamma-ray energies and intensities for each nuclide. These data are available in graphic form and as magnetic data files for library applications in computer codes used for analysis of pulse-amplitude spectra. The experimental techniques employed for acquisition and analysis of data in the Catalogue will be described in detail. In the development and application of these data to problems in activation analysis, fuel safeguards and reactor technology, considerable insight into the requirements for evaluated nuclear data has been obtained. The application of these data in a variety of user areas will be discussed and needs for improved nuclear data outlined.

1. INTRODUCTION

The Gamma-ray Spectrum Catalogue has been a continuing effort of this laboratory for the past ten years. The purpose of this effort has been to provide a collection of experimental x-ray and gamma-ray spectra obtained with pulse-amplitude spectrometers for general laboratory use in the analysis of gamma-ray spectra. The first two editions of the Catalogue [1,2] contained pulse amplitude spectra obtained with NaI scintillation spectrometers. These data were intended to present an internally consistent set of response functions obtained under specified laboratory conditions. The 2nd Edition also contained extensive text and supplementary material for general laboratory use of the techniques of gamma-ray spectrometry.

The effort during the past several years has been to update this collection of data to include experimental data obtained with semiconductor detectors, principally lithium-drifted Si and Ge devices. The improved

* Work performed under the auspices of the US Atomic Energy Commission.

energy resolution afforded by these detectors, together with refined electronics, offers the ability to measure energies and intensities of gamma rays with high precision. Since the utility of the high-resolution gamma-ray spectrometer is related to the quantitative analysis of complex spectra and isotopic assay, experimental techniques and reference data including precision energies and intensities of gamma rays emitted in the decay of radionuclides is essential. The current edition of the Catalogue, which is currently in publication, contains experimental spectra for about 300 nuclides obtained with state-of-the-art gamma-ray spectrometers. These data include results of experimental determination of the energies and intensities of all detectable photon transitions. This work carried out over a period of the past seven years, represents the results of a long-range effort to develop experimental techniques required to successfully apply high-resolution photon spectrometry as a laboratory tool for both basic and applied physics.

A typical experimental spectrum, as it appears in the Catalogue, is shown in Figure 1. This experimental pulse-amplitude spectrum represents the experimental response of a large-volume lithium-drifted Ge detector to radiations emitted in the decay of 115-day ^{182}Ta. The data are presented on an 11-in. x 17-in. computer data plot, suitably annotated to indicate the major features of the spectrum. The experimental values obtained for gamma-ray energies and relative intensities are listed on the reverse side of each plate as shown in Figure 2. In certain cases the low-energy region of the photon spectrum has been measured using lithium-drifted Si detectors. A typical example of such a measurement is shown in Figure 3.

As previously indicated the 2nd Edition of the Catalogue consisted of two volumes. The first volume contained extensive text covering the experimental techniques of gamma-ray spectrometry using NaI scintillation detectors. The second volume contained the experimental spectra and associated experimental data. The current edition will follow a similar format. The second volume contains over 300 experimental spectra with results of gamma-ray energy and intensity measurements. This volume is presently in publication and will be available as a U.S. AEC Report (ANCR-1000-2).

The first volume of this edition will contain a description of important experimental considerations in the use of Ge(Li) and Si(Li) spectrometers for the quantitative and qualitative measurement of gamma-ray spectra. This will include a discussion of the factors which influence detector response (i.e., resolution, counting rate effects, electronic stability and linearity), and the experimental techniques used for the analysis of gamma-ray spectra to obtain precision energies and intensities. The characteristics of all spectrometers used for the experimental results contained in the Catalogue will be included. In addition the first volume will contain selected sorted information from the data file of gamma-ray energies and intensities. This will include principal gamma-rays from all nuclides ordered by energy for different ranges of half-life and mode of nuclide production. These selected subsets of the data set form the basis for table look-up in computer techniques for automated analysis of spectra. This volume is planned for publication within the next year. Recognizing the existing need for the large volume of experimental data represented by the Catalogue effort, it was felt that the second volume should be made available at this time and not be held up by the tedious preparation required for Volume 1.

In view of the many applications of Ge(Li) gamma-ray spectrometers for isotopic analysis of radioactivity, a number of specialized experimental spectra have been included in the Catalogue. These include gamma-ray spectra of gross fission products, rare gases and their daughter nuclides associated

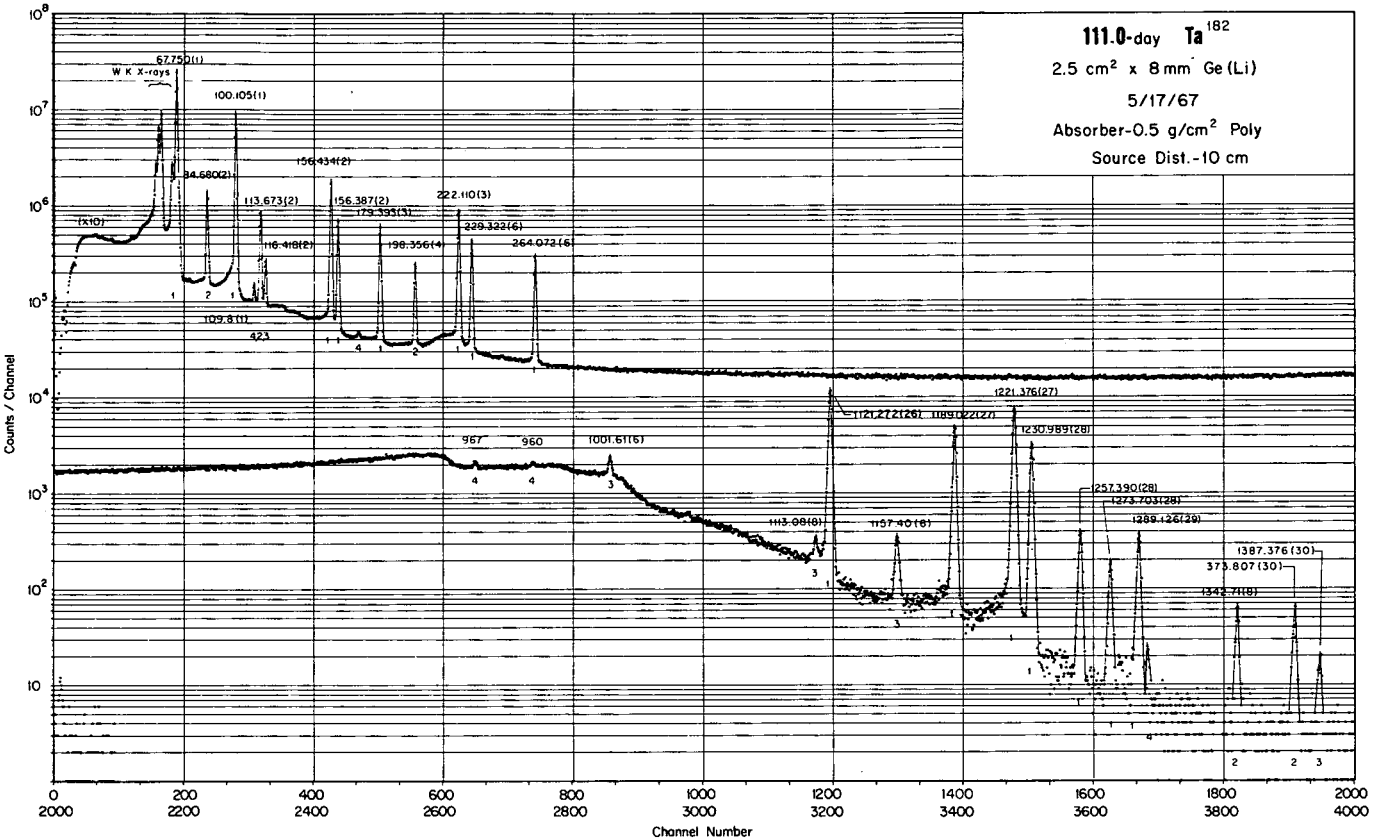

FIG. 1. Plot of a typical experimental gamma-ray spectrum as it appears in the Catalogue. This spectrum is for 115-day ^{182}Ta measured with a 65 cm^3 Ge(Li) detector.

GAMMA-RAY ENERGIES AND INTENSITIES

Nuclide: ^{182}Ta Half Life: 115.0 day

Detector: 55 cm^3 coaxial (c) Ge(Li) Method of Production: Ta(n,γ)

E_γ(KeV)	ΔE_γ	I_γ (relative)	ΔI_γ	S
W x-rays				
67.750	± 0.001	≡100	± 3.0	1
84.680	± 0.002	6.04	± 0.51	2
100.105	± 0.001	30.40	± 2.1	1
109.8	± 0.1	0.27	± 0.03	4
113.673	± 0.002	3.93	± 0.18	2
116.418	± 0.002	0.90	± 0.10	3
152.434	± 0.002	15.62	± 0.85	1
156.387	± 0.002	6.01	± 0.42	1
179.393	± 0.003	7.04	± 0.50	1
198.356	± 0.004	3.40	± 0.20	2
222.110	± 0.003	17.05	± 0.90	1
229.322	± 0.006	8.42	± 0.64	1
264.072	± 0.006	8.40	± 0.62	1
927.6	± 0.1	1.50	± 0.15	4
1001.61	± 0.06	5.34	± 0.42	3
1113.08	± 0.08	0.83	± 0.09	3
1121.272	± 0.026	79.94	± 3.2	1
1157.40	± 0.06	2.22	± 0.20	3
1189.022	± 0.027	37.41	± 1.9	1
1221.376	± 0.027	62.10	± 2.4	1
1230.989	± 0.028	26.02	± 1.2	1
1257.390	± 0.028	3.50	± 0.2	1
1273.703	± 0.028	1.49	± 0.11	1
1289.126	± 0.029	3.24	± 0.2	1
1342.71	± 0.08	0.61	± 0.06	2
1373.807	± 0.03	0.51	± 0.05	2
1387.376	± 0.03	0.15	± 0.04	3

FIG.2. Experimental values for gamma-ray energies and intensities for the ^{182}Ta spectrum. These values are tabulated on the reverse side of each plate.

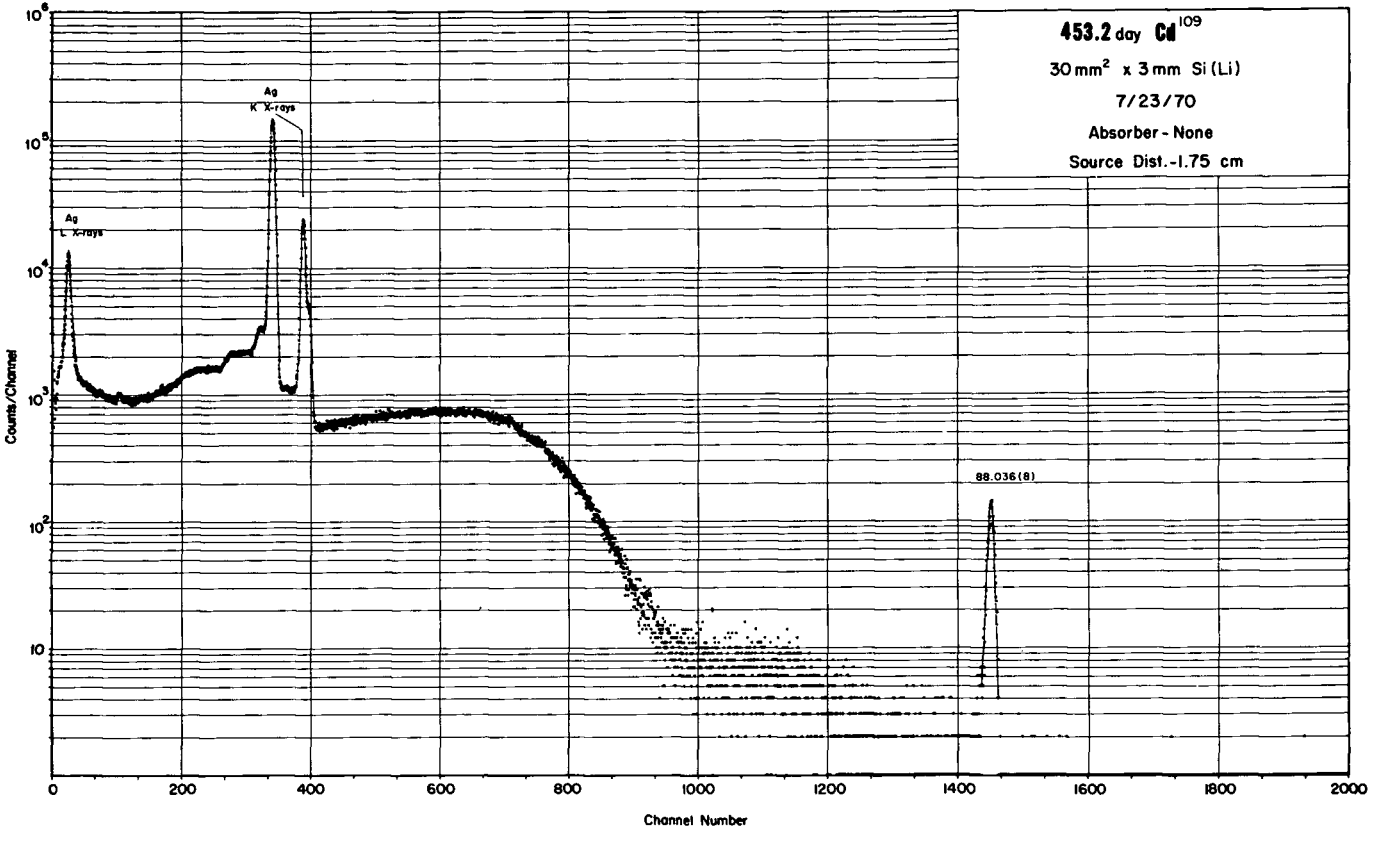

FIG. 3. Plot of the experimental spectrum of low-energy photons emitted in the decay of ^{109}Cd. These data were obtained using a high-resolution Si(Li) spectrometer.

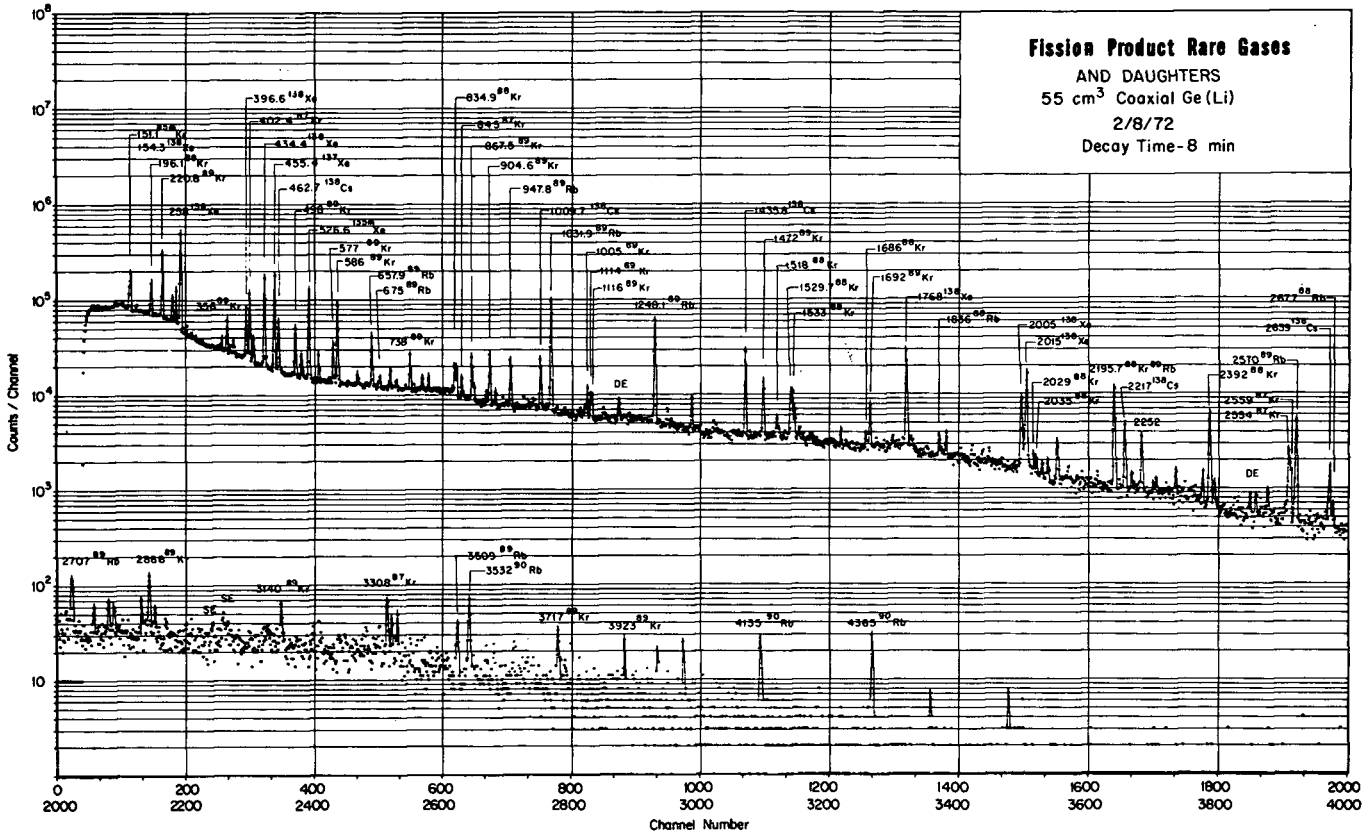

FIG.4. Gamma-ray spectrum of the rare-gas fraction of gross fission products measured after an 8-minute decay with a 65 cm^3 Ge(Li) spectrometer.

with fission, and the natural radioactive decay chains. The fission product gamma-ray spectra have been measured as a function of irradiation time and decay time for ^{235}U thermal fission. A typical example of this class of experimental data is shown in Figure 4, which presents the gross gamma-ray spectrum of short-lived fission product rare gases and their associated daughter nuclides. This particular spectrum represents data obtained from a sealed gas sample, measured 8 minutes after collection. These composite spectra are valuable in determining possible interferences in the analysis of complex spectra of gross fission product nuclides.

The large volume of data obtained from this experimental effort is utilized in many ways. It provides a base for the generation of files of gamma-ray energies and intensities for automated analysis of gamma-ray spectra. Present plans are to include it in the input to the data bank of the Nuclear Data Group at the Oak Ridge National Laboratory and to provide sorted output of selected data in magnetic tape format for general user applications. It is presently intended that the effort will continue to obtain data on additional nuclides and to improve the precision of energy and intensity measurements on nuclides presently in the file. It is felt that this approach to the measurement and compilation of selected types of nuclear data represents an appropriate method of obtaining specialized data sets for particular applications. It is essential that user acceptance of these data be the indicator for justification of continued or expanded effort in base technology for experimental nuclear gamma-ray spectrometry.

2. EXPERIMENTAL MEASUREMENTS

In the development of experimental techniques for the use of NaI scintillation spectrometers, the concept of a standard detector and experimental source-detector geometry was developed. The 3-in. diameter x 3-in. cylindrical NaI(Tl) detector was adopted in a standard geometry and the detector response characterized with considerable precision. With a known sensitive volume and standard geometry it was possible to achieve a laboratory standard spectrometer which has been adopted in many laboratories throughout the world. Unfortunately the 3-in. x 3-in. semiconductor radiation detector has eluded us to the present and the state-of-the-art has not made it possible to produce detectors with precisely defined sensitive volume. Over the past 6 or 7 years we have steadily progressed through small volume planar devices of 1 or 2 cm^3 volume to high-quality coaxial-drift detectors with sensitive volumes approaching 100 cm^3. For this reason and other factors related to the performance of low-noise electronics required to achieve good energy resolution, it has not been practical to consider a standard semiconductor laboratory spectrometer concept. Since the data contained in the present data file have been collected from early in 1966 to the present time, a variety of high-quality spectrometer systems have been used in these experimental measurements. The characteristics of the detectors used are presented in Table I. During this period considerable effort has been expended at this and other laboratories in the development and refinement of electronics to utilize the energy resolution afforded by semiconductor detectors and to develop techniques for the analysis of pulse-height data to obtain precision values for the energies and intensities of gamma rays. The experimental techniques developed at this laboratory have been described in the literature [3,4,5].

All data in the Catalogue were obtained using spectrometer systems which have been carefully calibrated to establish electronic system linearity and

TABLE I. CHARACTERISTICS OF DETECTORS USED FOR CATALOGUE DATA

No.	Type	Sensitive Volume	Applied Voltage	Resolution (FWHM) (electronic)	Resolution (FWHM) (1.332 MeV)	Drift Depth
1	Ge(Li) planar	2.5 cm^2 x 4 mm	800 V	1.3 keV (ext. FET)	2.5 keV	8 mm
2	Ge(Li) planar	2.5 cm^2 x 8 mm	800 V	0.65 keV (cooled FET)	1.78 keV	8 mm
3	Ge(Li) planar	4.55 cm^2 x 8 mm	1500 V	0.85 keV (cooled FET)	1.85 keV	8 mm
4	Si(Li) planar	30 mm^2 x 3 mm	100 V	0.22 keV (cooled FET)	(Fe K x-ray)	3 mm
5	Ge(Li) closed coaxial	35 cm^3 3.3 cm diam. x 4.0 cm	1750 V	0.98 keV (cooled FET)	1.90 keV	12 mm
6	Ge(Li) closed coaxial	55 cm^3 4.0 cm diam. x 4.9 cm	3000 V	0.71 keV (cooled FET)	1.88 keV	18 mm
7	Ge(Li) closed coaxial	65 cm^3 4.3 cm diam. x 5.5 cm	3300 V	0.85 keV (cooled FET)	1.88 keV	19 mm
8	Ge(Li) closed coaxial	70 cm^3 4.5 cm diam. x 5.2 cm	3500 V	0.87 keV (cooled FET)	1.90 keV	18 mm
9	Ge(Li) closed coaxial	80 cm^3 4.6 cm diam. x 5.44 cm	3100 V	1.0 keV (cooled FET)	1.93 keV	18 mm
10	Ge(Li) open coaxial	50 cm^3 4.03 cm diam. x 4.30 cm	2500 V	1.3 keV (ext. FET)	2.0 keV	12 mm

stability using both computer-controlled pulse generators [6] and multiple source techniques. All energy and intensity measurements are made using techniques developed at this laboratory and described in recent publications [7]. The analysis of all data to obtain energies and intensities was accomplished using the computer code GAUSS V, which has been evolved from non-linear regression techniques developed for the analysis of NaI data [4,8].

The determination of precision values for gamma-ray energies is accomplished using internal source calibration techniques. Over the past several years, a program has been developed at this laboratory to re-examine

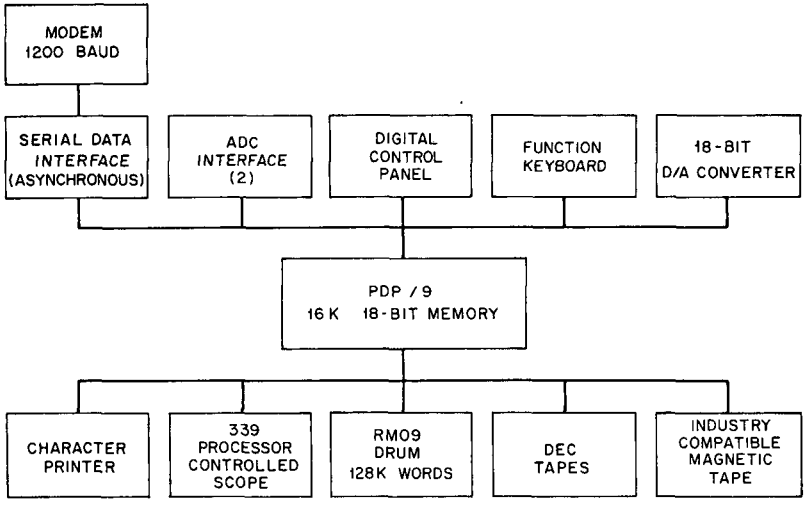

FIG. 5. Block diagram of an on-line data acquisition and analysis system used for processing of data for the Spectrum Catalogue. This system employs a Digital Equipment Corporation PDP-9 computer.

the reference energy scales used in gamma-ray spectroscopy and to provide a series of precisely-measured gamma-ray transitions for use as calibration standards. To date two groups of energy measurements covering the energy range from 30 keV to 1300 keV have been published [9,10]. These measurements provide a large number of precise gamma-ray transition energies for use in energy measurements. At higher energies, a few well measured transitions exist and work is progressing to provide more reference line up to 5 MeV. Above the pair threshold, the field-increment effect [5] introduces many geometric problems which affect the precision of energy determination. This is particularly true when measurements are made using large volume closed-end coaxial detectors. Because of this, all energy measurements are made using only full-energy peaks in the spectrum.

A typical laboratory data system in use for the simultaneous acquisition and analysis of pulse-amplitude spectra is shown in block form in Figure 5 and as a photograph in Figure 6. This system employs a processor-controlled graphics display oscilloscope for the application of interactive graphics techniques to the analysis of data and editing of data files. A 1200 Baud asynchronous data link provides access to other laboratory spectrometers and will provide a link to the laboratory 360/75 system for remote batch terminal operation.

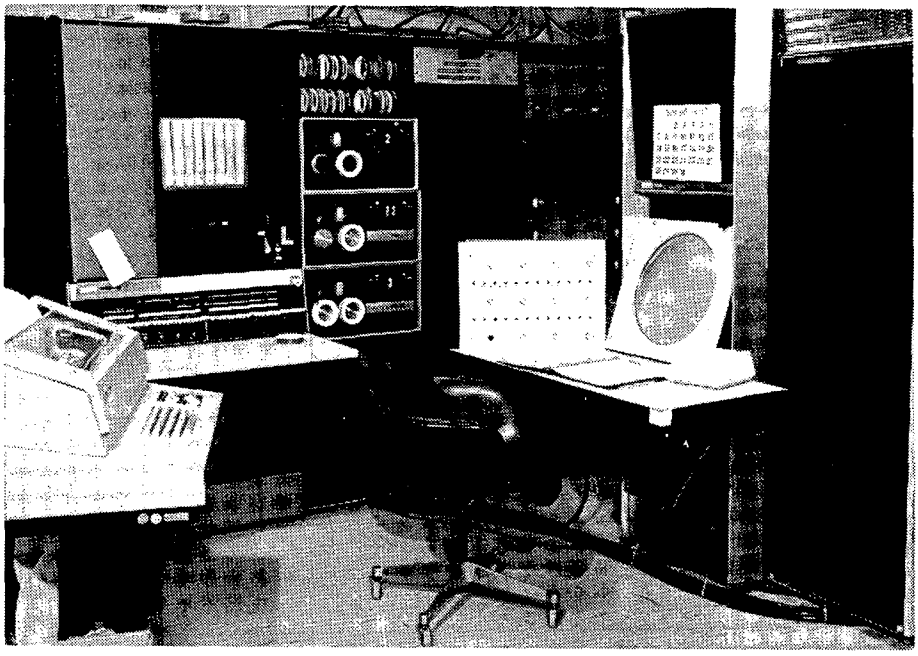

FIG.6. Photograph of computer data analysis system showing interactive display console with function keyboard and control plant.

3. SOURCE PRODUCTION

The measurement of gamma-ray spectra of over 300 individual nuclides has involved the use of a number of large experimental facilities. To produce a relatively pure sample of a given nuclide usually requires a selection of one of several possible nuclear reactions, a tailored radiochemical procedure for purification of the desired nuclide, and a selection of measurement time to optimize the experiment. Since spectra of both neutron and proton deficient nuclei are of interest, this required the use of both nuclear reactors and accelerators. The facilities used for the production of source material for measurement are listed in Table II, which indicates the facility and types of reactions used. The major reactor facilities at the NRTS have fast pneumatic shuttle facilities as well as in-tank irradiation facilities for individual samples. Portable shuttle facilities were utilized at particle accelerators for production and measurement of short-lived nuclides. In the use of accelerators, particle energy was also employed as a variable to enhance the production of a single nuclide. In all cases, either spec-pure or mass-separated material was employed for irradiations. Following irradiation, the sample material was chemically purified and decay data utilized to identify all observed transitions with the nuclide in question.

TABLE II. FACILITIES USED TO PRODUCE ISOTOPIC SAMPLES

Reactor	Maximum Thermal Neutron Flux	Facilities Used
MTR	2 x 10^{14} n/cm^2/sec (thermal)	Fast pneumatic shuttle In-tank capsule
ETR	8 x 10^{14} n/cm^2/sec (thermal)	Pneumatic shuttle In-tank capsule Gas loop experiment
ATR	10^{15} n/cm^2/sec (thermal)	In-tank capsule
EBR-II fast	10^{14}n/cm^2/sec (fast)	In-tank capsule

Facility	Particle	Energy	Reactions
U. of Colorado cyclotron	proton, He^3	10-35 MeV protons	p,xn; He^3,xn; p,γ
GGA Linac	electron	20-30 MeV	γ,xn; γ,p
LRL Linac Livermore	electron	100 MeV	γ,xn; γ,p
SREL electron synchrocyclotron	proton	590 MeV	p,xn; spallation
ORNL production cyclotron	proton	20 MeV	p,xn

4. ACKNOWLEDGMENTS

The development of experimental techniques required and the experimental collection and analysis of data presented in the Spectrum Catalogue represents the combined efforts of many people on the laboratory staff. The preparation of this and previous editions of the Catalogue has been supported by both the Reactor Technology and Applied Technology Divisions of the U.S. Atomic Energy Commission. The experimental measurements were made principally by R. J. Gehrke, L. D. McIsaac, J. E. Cline, R. C. Greenwood, R. G. Helmer and the author. Development of computer data reduction techniques was largely due to the efforts of R. G. Helmer and Marie Putnam. On-line data system development was the responsibility of G. O. English, W. R. Myers, E. W. Killian, R. C. Davies, L. O. Johnson, E. E. Owen and R. A. Coates. Source preparation and chemistry was accomplished under the direction of L. D. McIsaac. Data file management was the responsibility of Evelyn Baston and the exacting and important task of data table preparation and checking was accomplished by Carol Ball.

In addition, a vote of thanks is extended to the technical staff at each of the experimental facilities which were used for source production. The production of a single quality spectrum, of necessity, involved the combined efforts of many talents and specialties.

It is the sincere desire of the laboratory staff that the results will be of general use to scientists in many disciplines.

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DISCUSSION

N. M. SPYROU: I am not certain whether you imply that the listed sensitivity factors for the single radionuclide do not change relatively in order when a practical case in activation analysis, i.e. within a matrix of several radionuclides, is encountered?

R. L. HEATH: The sensitivity index can only provide an indication of the most probable photon transitions which will be detectable in a complex spectrum. By restricting the energy search criteria first to criteria based upon detector response to single nuclides, the number of extraneous correlations in isotope identification will be reduced.

C. WEITKAMP: I would like to point out the tremendous difficulties encountered in the conversion of analysed peak areas into absolute intensities if small and inaccurately known sample impurities cause large peak perturbations. Dr. Heath has briefly touched upon the question; it might be interesting to know his approach to the problem.

R.L. HEATH: This matter is quite complex and it represents one of the most important considerations in the analysis of complex photon spectra. It is realized that the importance of isotopic purity and carefully measured intensities of photons emitted in the decay of a given nuclide must not be underestimated. In the preparation of spectra for the catalogue great care has been exercised to insure spectral purity. In the analysis of complex spectra a number of gamma rays from each nuclide must be used, if possible, in order to obtain the best results in the quantitative assay.

A.H. WAPSTRA (Chairman): Your quality identification number does not apply to measurements made with anti-Compton arrangements. Could indications applicable to such set-ups be added in future editions?

R.L. HEATH: The quantity "S", which we have termed the sensitivity index, is a measure of the magnitude of a given full-energy peak above the background in the experimental response of a spectrometer system to a given nuclide. The use of Compton suppression techniques would be expected to improve the response to less intense low-energy gamma rays to a typical spectrum. From a knowledge of peak-to-Compton ratios for a given system, it is possible to derive such values for any detector system. At present we plan to use Compton rejection spectrometers for gross fission product gamma-ray spectral assay. Examples of the relative response of such systems will be dealt with in the text of Volume I.

J.J. SCHMIDT: Are you analysing only your own measurements or are you acting as a kind of analysis centre for the measuring groups in the United States as well?

R.L. HEATH: The objective of this collection of experimental data is to present a consistent set of detector response data to individual nuclides, measured by the most recent techniques and equipment. This can be achieved only if the measurements are made with this objective in mind. Since data for some isotopes are extremely difficult to obtain, requiring much effort and special experimental equipment and facilities, collaborative efforts with other laboratories are required. However, so far, only data of our own group have been included. As the data collection is enlarged to include short-lived nuclides, it will become necessary that some data from other sources be included.

A COMPILATION OF MODERN NUCLEAR-DECAY DATA FOR HIGH-RESOLUTION GAMMA SPECTROSCOPY *

F.F. DYER, L.C. BATE
Analytical Chemistry Division,
Oak Ridge National Laboratory,
Oak Ridge, Tenn.,
United States of America

Abstract

A COMPILATION OF MODERN NUCLEAR-DECAY DATA FOR HIGH-RESOLUTION GAMMA SPECTROSCOPY.

Recently published radionuclide decay data necessary for interpreting and quantifying high-resolution gamma spectra were collected and tabulated. The data, consisting of gamma energies and intensities and other information, e.g. half-lives, fission yields, neutron cross-sections, etc., for over 500 radionuclides, were computer-processed to yield a tabular form highly useful to gamma-spectroscopists and activation analysts. The collection is unique in that many short-lived radionuclides, e.g. fission gases are included that are not found in similar data tables. Basically, two computer listings are produced. The first table lists all data for each radioisotope arranged according to increasing atomic number (with increasing mass number for isotopes of the same atomic number). The source of reference for each nuclide is documented. The second table consists of a list of sorted gamma energies arranged according to increasing energy. Opposite each gamma energy is the contributing radionuclide symbol, its mass number and half-life, a classification of nuclide type (e.g. fission product), several associated gamma energies and their intensities and a cross reference to the nuclide's entry in Table I of the paper. By careful attention to the elimination of non-prominent gamma rays, Table II of the paper is kept manageably small yet sufficiently complete to permit spectroscopists to rapidly identify the lines of gamma spectra and/or check the results of spectral-analysis codes. The data on magnetic tape along with a program to make listings are available to users on request to the ORNL Radiation Shielding Information Center. The organization of the tables, applications of the data to gamma-spectral studies of reactor components and activation analysis and additional data needs will be discussed.

1. Introduction

Large lithium-drifted germanium, Ge(Li) detectors have revolutionized the measurement of gamma-ray emitting radionuclides. These detectors can now be obtained with a counting efficiency of about one-fourth that of 3 in. x 3 in. sodium iodide detectors, and many laboratories are using Ge(Li) detectors with efficiencies greater than one-tenth that of NaI. These new larger detectors have resolutions as good as or better than the very small earlier ones. Thus one can have, for example, a detector with a relative counting efficiency[1] of 13% with a resolution of 2.1 KeV (FWHM) or better. Although these larger detectors are expensive, they have expanded the role of Ge(Li) from a research-only tool to one of analytical applications.

* Research sponsored by the US Atomic Energy Commission under contract with the Union Carbide Corporation.

[1] Relative counting efficiency is derived by counting a ^{60}Co source with both a Ge(Li) and a 3 in. x 3 in. NaI detector with the source placed 25 cm from both detectors. 100 times the ratio of that count rate of the 1332 KeV line obtained with Ge(Li) to the corresponding count rate for NaI is the relative counting efficiency.

To effectively use Ge(Li) detectors in a wide variety of applications one must be able both to collect pulse-height spectra of several thousand channels and resolve the spectra by computers. Many laboratories combine these two functions with computers that function as pulse height analyzers. Ge(Li) gamma spectra are "resolved" by measuring the net areas of full energy peaks (total area minus underlying base line), calculating peak centroids and their corresponding energies, and computing photon emission rates for each peak. These arrays of energies and photon emission rates are then compared to entries of energies and intensities in radionuclide tables to determine likely isotopes responsible for the peaks. When candidates are found, photon emission rates are divided by appropriate fractional absolute intensities to obtain disintegration rates. Thus we see the need for extensive accurate nuclear data.

We began our collection of data about three years ago during a study of fission product distribution in a molten salt nuclear reactor. Early in that study we began to collect gamma spectra with one to three hundred full energy peaks. Many of the radionuclides measured were rather short lived and because of the great number of peaks it was difficult to assign many of them to known radioisotopes. Except for the table by Gunnink et al [1] for some of the relatively long lived radionuclides, there were no lists of sorted accurate gamma energies that we could use to identify spectral lines. We thus began a systematic collection from the literature of fission product energy and intensity data and eventually processed about 1000 of these complex spectra [2]. Since that time we have expanded our data collection to all types of radionuclides and have included all data necessary for absolute neutron activation analysis. The collection now includes data for 513 radionuclides. The data are stored and edited on a master card file from which we generate listings and duplicate files on magnetic tape.

2. Criteria for Selection of Data

Where possible we have selected for inclusion in our data collection gamma energy and intensity data from critically evaluated publications such as the one by Martin and Blichert-Toft [3]. We have also relied heavily on the well founded experimental studies of Gunnink et al [1]. In the case of those radionuclides that have been studied by two or more investigators in the past few years, we normally select the most recent reference provided that the investigator: (1) has used the best modern experimental techniques, (2) was aware of the existing data for the radioisotope, and (3) gave adequate explanations for any large differences or unexpected results obtained (if any) compared to the existing data. In some instances we have been unable to choose between two or more references and have included separate entries for both references in our collection. Some radionuclides, of course, have been studied very little (for example, many short-lived fission products); and for these we tabulate whatever we can find. We do not, however, tabulate older values of gamma energies that have been determined by NaI spectroscopy and which may be uncertain by 10 KeV or more. We have used some of the older gamma intensity values where no recent study exists.

3. Data Editing Program

A computer program (NUCDAT) is used to read the data either from punched cards or magnetic tape, prepare new tapes from a master card file or copy existing tapes. NUCDAT makes listings of the data either from tape or cards as desired. The program will also generate radionuclide data libraries for both large and small computer programs that are used to process Ge(Li) gamma-ray spectra at Oak Ridge National Laboratory.

4. Content and Organization of Tables

Principally two tables are listed by NUCDAT. Table I consists of card images of the input data edited to be easily read. An example of a slightly abridged entry in Table 1 for the radionuclide ^{103}Ru is shown on page 4. One line (card) is used for all data such as half lives, cross sections, fission yield, etc. as shown in the example. Two lines are used for references and other notes; and the remainder of the entry consists of gamma energies and intensities. Energies are given in KeV, and are punched on cards with as many significant digits as given in the reference. Two digits after the decimal are printed; usually only one is significant. Intensities are expressed as photons emitted per 100 decays (absolute percentage, status A) if such values are available. In many instances only relative intensities are available (status R). We occasionally classify the intensity status as "D" when it appears, but we are not completely sure, that the intensities are expressed in absolute percentages.

To aid in identifying lines in gamma spectra, we classify radionuclides into categories according to the manner in which they are normally produced. As shown in the example of ^{103}Ru — class 0 denotes a neutron capture product, 1 a fission product, 2 a radionuclide formed both by fission and neutron capture, 3 a

TABLE I. EXAMPLE OF RADIONUCLIDE DATA ENTRY IN TABLE I OF NUCDAT

```
 A   B  C D E    F    G  H   I    J    K    L    M  N    O P  Q
210  RU103  44  39.6 D  B-  RU102 31.61 1.40  2  3.Ø  7 A  4.2
REF  M.J. MARTIN AND P.H. BLICHERT-TOFT RADIOACTIVE ATOMS,
REF  NUCLEAR DATA TABLES, SEC. A, VOL. 8 (1970).
ENERGY   % INT. ENERGY   % INT. ENERGY   % INT. ENERGY   % INT.
 39.55  Ø.056   53.11    Ø.34  294.88   Ø.21Ø  443.85   Ø.36Ø
497.90 89.Ø    556.90    Ø.8Ø  61Ø.2Ø   5.4Ø
```

WHERE THE HEADINGS DENOTE:

A=TABLE 1 ENTRY NUMBER
B=ISOTOPE SYMBOL
C=MASS NUMBER
D=STATE:BLANK=GROUND STATE,M=METASTABLE STATE
E=ATOMIC NUMBER
F=HALF LIFE
G=TIME UNIT OF HALF LIFE
H=MODE OF DECAY: EC=ELECTRON CAPTURE,B-=NEGATRON DECAY, B+=POSITRON DECAY
I=PRECURSOR SYMBOL FOR NEUTRON CAPTURE PRODUCT
J=PRECURSOR MASS NUMBER
K=NATURAL ABUNDANCE IN PERCENT OF PRECURSOR
L=2200 M/SEC NEUTRON CAPTURE CROSS SECTION OF PRECURSOR
M=CLASS OF RADIONUCLIDE:0=NEUTRON CAPTURE PRODUCT 1=FISSION PRODUCT,2=BOTH NEUTRON CAPTURE AND FISSION PRODUCT,3=NEUTRON DEFICIENT RADIONUCLIDE,4=NATURAL RADIONUCLIDE,5=SPECIAL RADIONUCLIDE CLASS,6=FISSION GAS.
N=FISSION YIELD IN PERCENT
O=NUMBER OF GAMMA RAY ENERGIES TABULATED
P=INTENSITY STATUS OF GAMMA RAYS:A=ABSOLUTE PERCENT (GAMMAS PER 100 DECAYS),R=RELATIVE,D=POSSIBLY ABSOLUTE
Q=NEUTRON CAPTURE RESONANCE INTEGRAL

TABLE II. EXAMPLE OF TYPICAL ENTRIES IN TABLE II OF NUCDAT, FOR GAMMA RAYS OF ^{56}Mn AND ^{134}I. The headings denote: Type = Radionuclide class as previously defined; NGT = Total number of gamma energies tabulated in Table I; ISC = Cross reference to Table I where data for radionuclides are tabulated.

Energy	Nuclide	Type	Half Life	NGT	ISC	Energy	Intensity	Energy	Intensity	Energy	Intensity	Energy	Intensity
846.60	Mn 56	0	2.57H	3	185	846.60	99.00	1811.20	30.00	2112.6	15.5		
847.03	I 134	1	53.00M	29	308	847.03	96.0	884.08	66.00	1072.53	14.30	595.40	11.20
						621.75	10.90	1136.12	9.15	540.80	8.60	677.34	8.20
						405.44	7.35	857.28	6.60	1806.90	5.60	974.63	4.93

neutron deficient radionuclide formed by charged particle bombardment, 4 a natural radionuclide, 5 a special "catch all" category, and 6 represents a fission gas. We have just recently classified fission gases in a separate class to cause our computer to reject identifications of fission gas daughters when we are interested only in the gases. More will be said later about the uses of a radionuclide classification system.

Table II, whose listing by NUCDAT is optional, consists of a sorted list of gamma energies, arranged by increasing energy, with several items of associated information given with each gamma energy. This table was designed for use by gamma spectroscopists and activation analysts to aid in identifying lines in gamma spectra. A spectroscopist may know the energies of the lines in a spectrum and wish to identify their radionuclide sources or more typically may wish to confirm (or deny) an identification made by a computer program. Because there are many thousands of gamma rays (many of which have nearly the same energies) emitted by radioisotopes, it is impossible to design a "fool proof" identification algorithm. Because computer programs frequently make mistakes in identifications it is necessary for spectroscopists to check their identifications.

Considerable effort was devoted to obtaining an arrangement of Table II that would permit the table to be used easily and efficiently. Typical entries for gamma rays of ^{56}Mn and ^{134}I are on page 6. For ease of reference, sorted energies are listed down the left margin. The items associated with each energy are listed across the page in what we have found to be an order of decreasing importance with respect to the identification of spectral lines. Normally the information of Table II is more than enough to identify all the radionuclides that produce a gamma spectrum. Adjacent to each energy is the radionuclide symbol and mass number followed by the radionuclide class, half life, the number of gamma energies tabulated in Table I and a cross reference to the radionuclide's entry in Table I. The class and half life are quantities that normally are the most important ones in rejecting invalid radionuclide candidates for spectral lines. The spectroscopist normally knows if a specimen that he is attempting to analyze can possibly contain fission products, neutron capture products, etc. He rejects candidates of the wrong class. He also rejects candidates whose half lives are incompatible with the specimen's decay time. For ease of application half lives are listed in their conventional units rather than a standardized unit. If, after these rejections, there still are two or more candidates from the sorted gamma energy list other associated energies may be examined. If they exist we list in Table II up to 12 associated gamma energies. These energies are again sorted and listed in order of decreasing intensity. Thus to make a spectral line identification it is normally only necessary to refer to the first two or three energies. Although normally two or three energies are sufficient to identify a radionuclide, we want to have the energies of the most intense gamma rays listed in Table II so that most of the lines in a spectrum can be easily identified. Listing too few would permit isotopes to go unidentified as well as cause a too frequent reference to Table I to get the "complete picture." Listing too many energies would make Table II too large to be manageable (if a radionuclide has 5 sorted energies it appears 5 times in Table II). We have found that the most useful version of Table II is produced if we sort either 12 gamma rays or up to that gamma ray whose intensity is one percent of the most intense one. Since most of our work does not involve neutron deficient radionuclides we find it useful to leave these radionuclides out of Table II. In this way we produce a compact but highly useful aid to our efforts in spectral line identifications.

The program NUCDAT will sort and list in Table II any one class or combination of classes desired. Thus it is possible for the user to produce any version of Table II that meets his needs.

5. Applications of this Data Collection

As previously mentioned we generate, from this collection of data, files that are used in computer programs to identify and measure peaks in gamma ray spectra. Currently most of our efforts are devoted to small computer application. We have two Nuclear Data 50/50 systems each consisting of a 4096-24-bit word hardwired analyzer and an 8192-18-bit word PDP-15 computer. Program MONSTR [4] is used to identify and measure peaks, calculate disintegration rates and parts per million for activation analysis problems. Identifications are made from a magnetic tape file of sorted gamma energies arranged with increasing energy. All of the energy entries of Table II are recorded in this file. Each energy has associated with it the mass and atomic numbers, the nuclide type, and the half-life in seconds. Candidates for the lines of a spectrum are selected by comparing spectrum and tape energies, nuclide types with a preset list of the nuclide types that may be present, and half-lives with the decay time. Candidates may also be entered manually in the candidate list.

During the search for candidates, a list of unidentified peaks is produced. Subsequently a radionuclide file is read with all information necessary to compute disintegration rates or elemental concentrations for activation analysis problems. Because the computer's memory is small, we read the radionuclide file one entry at a time until computations for all candidates were finished.

We are using these systems and this collection of data to carry out a wide range of work in absolute neutron activation analysis, and radionuclide measurements. We have just begun to test our scheme of absolute neutron activation analysis on standard samples whose trace element content has been well establishe and documented by comparative neutron activation analysis and other reliable methods. Using this approach, we hope to critically evaluate much of the data we have collected for neutron capture radionuclides.

We are also using high resolution gamma spectroscopy to study the release of fission products from the fuel of high-temperature gas cooled reactors and in-pile experiments. Although our reactor experiments are carried out to further our understanding of reactor components, they sometimes yield approximate values for absolute values of gamma-ray intensities. Recently, for example, an in-pile fission gas release study indicated that the absolute intensity of the 258 KeV gamma-ray of ^{138}X is 30 percent — a value closely approximating 32.5 percent as reported by Monnand [5].

6. Availability of Data to Users

Individuals or organizations who wish to obtain copies of these data can do so by sending a 1200 foot reel of industry compatible magnetic tape to the Radiation Shielding Information Center, Oak Ridge National Laboratory, and requesting data file DECAYGAM. A copy of the editing program NUCDAT written in FORTRAN IV and user's instructions will be supplied with the data.

7. Some Final Observations

Undoubtedly special problems will always arise in which radiochemical and/or comparative methods of radionuclide analysis will be required. Nevertheless it seems inevitable that the current trend in instrumental absolute methods of neutron activation analysis and gamma spectroscopy using Ge(Li) detectors supplemented by coincidence and anticoincidence techniques will continue. This trend is due not only to investigator's research interests but also to the simplicity and power of these methods as well as economic realities. For most organizations it has become economically infeasible to obtain by comparative

activation analysis the large number of analytical results required in such studies as trace element surveys of ecological and biological materials. In other areas of work such as gamma-ray scanning of nuclear fuel elements and other nuclear reactor components, the instrumental techniques are the only ones applicable.

In our current tabulation of data for 513 radionuclides, 164 have only relative gamma-ray intensities. Since the data for those radionuclides that are listed with absolute intensities were gathered from publications of reputable investigators, the data are presumably reliable. We, however, have tested the reliability of only a few. It is therefore evident then that much of the task remains in determining, evaluating and compiling a complete, reliable radionuclide data file for applied gamma spectroscopy.

There are also significant needs for data in other areas of applied nuclear technology. An example is the requirement of fission yields as a function of neutron energy for fission of ^{233}U, ^{239}Pu and other heavy nuclides to permit fission-product inventory calculations for the newer types of nuclear reactors such as the high temperature gas cooled reactor. It appears that much of this data has not been measured. Other examples of data needs can be found in the areas of shielding and dose-rate calculations.

At the present time there is no unified collection of nuclear data for applied nuclear technology. Although the applied nuclear scientists must use the variety of data that has been mentioned (radionuclide decay data, cross sections, etc), he must search for it in sources that are too diverse. We therefore believe that there is a place for specialists who would be responsible for evaluating and compiling complete nuclear data files that the user could apply easily and efficiently. The compiler should maintain communication with users to determine areas where additional data are needed. He should also assist in publisizing these needs. If such an effort were made with care, the extreme duplication of efforts now occurring in this endeavor would be reduced and the life of the applied nuclear scientist would be easier.

8. Acknowledgements

Although the sources of data are given for each radionuclide in our data file, we wish to acknowledge four specific sources. We especially thank the Nuclear Data Group of the Oak Ridge National Laboratory for the use of their splendid reference files which led us to much of the information collected. Our gratitude also goes to the Radiation Shielding Information Center of ORNL for their cooperation in making the data available to the scientific community. We wish to thank Ray Gunnink of the Lawrence Radiation Laboratory for his permission to include in our collection results from much of his experimental measurements [1]. Finally, we reference the papers of Adams and Dams [6,7] from which we have drawn heavily.

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DISCUSSION.

V. KRIVÁŇ: The starting point for the work described in your paper is absolute neutron activation analysis. (In your comments on my paper (IAEA-SM-170/60), too, you indicated that you perform activation analysis by this method). While I believe that activation analysis by the absolute method is more convenient than by the comparative method, I would be reluctant to make generalizations to this effect. There are two main difficulties in connection with the absolute method: the first has to do with the accuracy of the nuclear data needed and the second with the fact that the composition of the sample to be analysed is not well enough known and therefore one does not have sufficient certainty as to the accuracy of the analysis, particularly as regards interference reactions and self-shielding. In such cases even the best knowledge of the nuclear data will not be a guarantee of accuracy. I should therefore like to ask what, in actual practice, causes you most trouble in absolute activation analysis: is it problems relating to knowledge of nuclear data, to approximate knowledge of sample composition or to the variability of the neutron spectrum? In this connection it would be interesting to know whether, in this work, you irradiate only with thermal neutrons or with the entire fission spectrum. In the latter case the danger of interfering reactions would be greater.

F.F. DYER: In terms of the number of samples analysed and therefore of economics, ORNL uses mainly absolute methods in its thermal neutron activation analysis program. I am speaking of analysis that is carried on mainly as a service or with reference to applied problems, not for research. The types of samples involved are biological, environmental and the like. We are aware that certain elements in some of the samples cannot be measured by absolute methods because of such problems as neutron shadowing by atoms of the matrix or of a constituent or gamma-ray attenuation in the sample. However, for ORNL the need seems to be to have an efficient, inexpensive activation analysis method to meet the greatest need, leaving the more complex problems to be handled by special, more expensive methods. We have two NAA facilities with pneumatic tubes, one is in the Oak Ridge Research Reactor, with a thermal neutron flux of $\sim 4 \times 10^{13}$, and one in the High Flux Isotope Reactor, with a thermal neutron flux of $\sim 5 \times 10^{14}$. This latter facility has a very highly thermalized neutron flux.

J. BLACHOT: What criteria did you use for including "fission product" isotopes in your computations? Was it their half-life, their fission yield or was it something else.

F.F. DYER: Except in cases where we face limitations of time and money, we do not omit any radionuclides for which valid data are available. We try to pick the most recent "best" data.

J. BLACHOT: What method did you use for measuring the absolute intensity of the 258 keV line of ^{138}Xe?

F.F. DYER: A plot of the release rate to birth rate ratios versus decay constant for fission gases was made for an in-pile fuel study. The ^{138}Xe was not consistant with the other gases. The absolute intensity of the ^{138}Xe gamma ray was suspect. When it was assumed the curve was correct and the intensity was adjusted to make ^{138}Xe agree with the curve, the adjusted value agreed with that of Monnard.

Section XIV
VARIOUS APPLICATIONS

Chairman

A. T. G. FERGUSON (UK)

THE USE OF NUCLEAR DATA IN THE DESIGN OF RADIATION INSTRUMENTS FOR MINERAL EXPLORATION AND MINING

C.G. CLAYTON, L.G. SANDERS
Atomic Energy Research Establishment,
Harwell, United Kingdom

Presented by A.T.G. Ferguson

Abstract

THE USE OF NUCLEAR DATA IN THE DESIGN OF RADIATION INSTRUMENTS FOR MINERAL EXPLORATION AND MINING.

The increasing use of nuclear techniques for mineral analysis in a wide variety of environments associated with mineral prospecting and mining has emphasized the need for the presentation of nuclear data in a form which allows rapid assessment of the analytical problems. The present paper identifies a number of applications in the field of minerals and gives an example of the use of nuclear data for resolving a typical analytical problem and of the difficulties in applying existing compilations. A form of presentation of relevant nuclear data for use in this type of problem is proposed.

1. INTRODUCTION

Nuclear techniques are now gaining increasing importance as a means of providing a direct indication of the concentration of selected elements in rocks and minerals during geochemical explorations and in mining operations. Instruments based on these techniques have the advantage of ruggedness and a relatively low sensitivity to environmental conditions, and they offer the possibility of a direct measurement of elemental composition. Although some nuclear techniques have been used in oil field exploration for about the last twentyfive years, it is the recent introduction of Californium-252, the increased reliability and output of neutron tubes and the development of high resolution solid-state detectors coupled with a need for more early and less costly information on the elemental composition of rocks and minerals which has recently accented the need for these techniques for in-situ analysis. Easy access to selected nuclear data is essential if both rapid and detailed considerations are to be given to the choice of technique and to the design of equipment for use in the type of analytical problems which occur in geochemical prospecting and mining. The nuclear data which is available tends to reflect an early interest in reactor design.

The most important applications occur in logging boreholes drilled on land and into the sea bed and the possibility of using

nuclear techniques in vehicles towed on the sea bed is also currently being considered. The use of Radiation Instruments has become established for the control of underground mining operations and they are now being developed for ore grade evaluation in open-pit mining[1,2,3,4].

The applications of nuclear techniques in oil-well logging are well known and the problems of instrument design are relatively well understood, even if the solutions are not always evident. In general, the problems of interpretation arise from variations in borehole conditions, from the indirect nature of the required measurements of density, porosity, water content and oil/water interface location, as well as from variations in the lithology of the rocks surrounding the boreholes.

In geochemical prospecting and in metalliferous mining the problems are somewhat different. In these applications emphasis is on measuring the concentration of one or of several elements in the presence of other elements in the rock matrix which may interfere with the measurement. From the point of view of designing a suitable instrument, careful consideration has to be given to the most appropriate reaction, to the measuring technique and to any constraints imposed by the physical conditions which prevail in the measurement region.

This paper seeks to identify the most important reactions which can be used for in-situ mineral analysis and the type of data which is required. This data needs to be readily available and in a form which is relevant to the analytical problems to be considered. An example of the general route through a feasibility study is given which demonstrates the type and extent of the data needed for a design study. From this study conclusions are drawn to indicate the preferred compilation of the most suitable data for applications of this type. The problem chosen is one of analysis in borehole conditions, but the same general approach can be used for the analysis of minerals passing on a conveyor belt.

2. USEFUL INTERACTIONS

Elemental analyses based on photon, heavy ion and neutron interactions are currently being used. The most important tech-

niques based on photons as the primary radiations are X-ray fluorescence, γ-ray scatter [unselective and selective γ-γ logging, the (γ,γ') reaction and γ-ray resonance scattering] and photo-neutron activation analysis.

X-ray fluorescence and unselective and selective γ-γ logging are important techniques in mineral exploration and mining and they form the basis of a variety of instruments, but further development is not impeded by unavailability of suitable data so that these techniques will not be considered further here.

The only important application of a photoneutron reaction is in prospecting for beryllium, because of the low threshold (1.66 MeV) and the unique position (apart from deuterium) of this reaction is that it is the only photoneutron reaction which can be induced by using radioisotope sources. In general, the use of (γ,n) reactions is inhibited because of the relatively high energy (≈20 MeV) of the giant resonance, the fact that it varies only slightly over the Periodic Table, and the relatively low interaction cross-sections. Although attempts have been made to measure the activation products from (γ,n) reactions by using transportable accelerators in the field, these exercises have, as far as the authors are aware, found no useful applications in mineral prospecting and mining.

There is some interest at present in the (γ,γ') reaction[5] and in γ-ray resonance scattering for mineral analysis[6,7]. However, cross-sections are generally small and the number of elements which can be analysed by these methods is limited. In the case of the γ-ray resonance technique, the problems of developing a gaseous source of sufficiently high activity at high temperature and suitable for operation for long periods in a borehole have not yet been fully resolved. For these reasons these reactions are not considered further.

There is much current interest in heavy ion induced X-ray fluorescence techniques[8], but application to the analysis of natural materials is limited by the low penetration of the incident particle (especially important where inhomogeneity and particle-size effects occur), by the need to use high energy accelerators which are expensive and bulky for field operation, and by the restriction to the analysis of elements of atomic number below aluminium,

approximately. At higher atomic numbers, X-ray induced, non-dispersive X-ray fluorescence techniques are in common use.

Neutron interactions are of increasing importance but, although a considerable amount of nuclear data is available, access to a form which is suitable for rapid optimisation of an analytical technique is not always easy, as will be seen from the example given below.

When defining a neutron reaction for the present purpose, consideration must also be given to the origin of the radiation used to identify the reaction. Thus, the most important reactions are considered to be those associated with:-

(i) gamma radiation from neutron capture,

(ii) gamma radiation emitted during the radioactive decay of a nuclide formed by neutron capture,

(iii) gamma radiation from inelastic scattering of neutrons

and (iv) prompt γ-radiation following particle emission.

3. PRELIMINARY STUDIES

The initial study of an analytical problem of the type described below is conveniently undertaken in two stages. In the first stage, a rapid survey of the relevant data is made in order to see whether or not suitable reactions exist which give a reasonable probability of success in identifying the wanted element and to see whether or not significant interference can be expected from other elements present in the sample.

The viability of possible reactions are judged in relation to any constraints imposed by the type of source which is available, or can be tolerated, and by the particular application. In borehole logging, for example, except in special circumstances, the use of γ-rays from nuclides with a half-life exceeding 60 seconds approximately is excluded, because of the relatively long irradiation times which must then be used. However, if the half-life of the product nucleus is only a few seconds, a high induced radioactivity may be obtained and the half-life is still sufficient to equate with an acceptable separation of source and detector at the logging speeds which normally prevail. Under these circumstances interference from inelastic scatter γ-radiation and from

γ-rays from radiative neutron capture will be reduced. The merit of using inelastic scatter γ-rays is that complete independence of the measurement from the probe speed through the borehole can be achieved.

In the preliminary survey, consideration has also to be given to the environmental conditions; to the presence of drilling mud or water in the borehole, to the possible inclusion of a steel liner on the inside of the hole, to water pressure and temperature and to possible variations in hole diameter. Such considerations are likely to exclude a measurement based on low energy X-rays, or on β-particles emitted by a product nucleus, for example.

In general, the following procedure is adopted when carrying out a feasibility study:

(i) The nominal elemental composition of the rock and its mineral content is inspected,

(ii) for each wanted element and for each potential interfering element, the variation in reaction cross-sections with neutron energy is examined for emission of:-

(a) capture γ-rays,

(b) activation γ-rays,

(c) inelastic scatter γ-rays

and, (d) prompt γ-rays from particle reactions,

(iii) possible useful reactions are identified and the energies and relative intensities of the γ-rays associated with each reaction are considered,

(iv) tables, such as described in Section 3.1.1. are compiled so that an effective decision on the relative merits of the various reactions can be made.

From such a preliminary examination, a decision can be taken on the merits of a detailed theoretical approach to establish the design criteria for the system.

Assuming that a reaction can be identified, which gives promise of a useful analytical procedure, a more detailed study is carried out. Using the values of $M_{p\gamma}$ (see Section 3.3.1.) which relate to unit thermal neutron flux, an appropriate next step is to calculate the actual spatial distribution of thermal neutron flux in the medium around the source. This involves a knowledge of the neutron cross-sections from the maximum neutron energy of the source to

TABLE I. MAJOR ELEMENTAL CONSTITUENTS OF COAL AND TYPICAL ABUNDANCES

Element	Atomic weight	Weight % of natural element in sulphur-free coal	Mass number	% abundance in natural element	Atom number density in coal* ($10^{23}/cm^3$)
C	12.011	91	12 13	98.89 1.11	0.6317 7.09×10^{-3}
H	1.008	4.0	1 2	99.985 0.015	0.3346 5.02×10^{-5}
O	15.999	3.0	16 17 18	99.759 0.037 0.204	1.577×10^{-2} 5.85×10^{-6} 3.23×10^{-5}
N	14.007	2.0	14 15	99.63 0.37	1.20×10^{-2} 4.45×10^{-5}
S	32.064	Additional weight % sulphur≠ 4.6	32 33 34 36	95.0 0.76 4.22 0.014	1.144×10^{-2} 9.15×10^{-5} 5.08×10^{-4} 1.69×10^{-6}

* Assumes $\rho_{coal} = 1.4\ g/cm^3$.

≠ Assumes equal atom number densities for N and S.

thermal energies for all elements in the sample. The detail of this calculation depends on the particular problem and it may be sufficient to consider a simple two-group diffusion theory or it may be necessary to employ a fully developed multi-group code.

When this has been done, the volume source of γ-rays can be derived and the γ-ray intensity at the detector for the selected energy can then be calculated.

3.1 Example: measurement of the sulphur content of coal in borehole conditions

The application dictates the largest diameter to which the probe can be made and the depth and condition of the borehole. No consideration is given at this stage to the possible presence of water in the borehole but it is assumed that the coal is free of moisture, heavy mineral content (ash) and organic debris. It is also assumed that the Rank (related to carbon/hydrogen ratio) is constant and this is generally true for coal deposits in a particular geographical region. With these limitations Table I is compiled which gives the relevant properties of the major elements in a typical coal.

The relative merits of each possible reaction are now considered in turn.

3.1.1 Radiative neutron capture

This reaction is convenient in as much as it 'fits' into a borehole logging system: the high energy γ-rays emitted are compatible with the environmental conditions.

Table II gives the particle and macroscopic cross-sections of the major elements we wish to consider and an examination of this data gives the first indication as to whether or not a suitably large reaction cross-section exists and the possible origin of an interfering reaction.

However, before adequate judgement can be made, it is necessary to know also the energies and emission probabilities of the γ-rays emitted at neutron capture, and these are tabulated, as far as possible, in Table III. It can be seen that a technique based on measuring the 5.42 MeV γ-ray from sulphur-33 appears promising

TABLE II. REACTION CROSS-SECTIONS FOR THERMAL NEUTRON RADIATIVE CAPTURE FOR MOST IMPORTANT ELEMENTS IN COAL

Element	Mass number	Reaction cross-section (10^{-24} cm^2)	Macroscopic cross-section* (cm^{-1})
C	12	3.4×10^{-3}	2.15×10^{-4}
	13	9.0×10^{-4}	•
H	1	0.332	1.11×10^{-2}
	2	5.7×10^{-4}	•
O	16	1.8×10^{-4}	2.84×10^{-7}
	17	2.4×10^{-5}	•
	18	2.0×10^{-4}	•
N	14	7.5×10^{-2}	9.0×10^{-5}
	15	2.4×10^{-5}	•
S	32	0.51	5.83×10^{-4}
	33	2.3×10^{-3}	•
	34	0.27	1.37×10^{-5}
	36	0.14	•

* Relates to atom number density for particular element in coal.

• Indicates negligible macroscopic cross-section.

since it has a relatively high value of I_γ and the energy is above the energy of any of the γ-rays emitted by carbon, hydrogen and oxygen. From Table II it can also be seen that the reaction $^{32}S(n,\gamma)^{33}S$ has the highest particle cross-section of the isotopes considered.

Further examination of Table III indicates that if nitrogen is present in the sample, it could provide a major source of interference since several γ-rays are emitted from nitrogen-15 which have energies close to the favoured γ-ray energy from sulphur-33.

A comparison of the significance of interfering γ-rays can be obtained by combining the absorption data of Table II with the emission data of Table III, and if we consider a volume element of

TABLE III. ENERGIES AND INTENSITIES OF GAMMA RAYS FOLLOWING THERMAL NEUTRON RADIATIVE CAPTURE BY MAJOR ELEMENTS IN COAL

Element	Product isotope	E_γ (MeV)	I_γ*
C	13	4.945 3.684 1.261	67.0 31.8 29.2
H	2	2.223	$\geqslant$ 97
O	17	3.271 2.180 1.088	≠ ≠ ≠
N	15	10.828 8.309 7.299 6.321 5.560 5.532 5.297 5.267 4.508 3.675 3.531	15.0 4.22 8.36 16.65 9.05 17.79 18.58 25.41 15.81 15.52 9.58
S	33	8.641 7.800 5.420 5.047 4.870 4.639 4.431 3.723 3.370 3.221	1.91 2.81 42.44 2.26 8.24 1.49 3.16 2.03 3.78 19.46

* I_γ = number of gamma rays/100 neutrons captured.

≠ Data not available.

TABLE IV. VALUES OF $M_{p\gamma}$, THE NUMBER OF γ-RAYS OF A GIVEN ENERGY EMITTED PER SECOND PER cm^3 OF SAMPLE PER UNIT THERMAL NEUTRON FLUX

Product nucleus ^{15}N		Product nucleus ^{33}S	
E_γ	$M_{p\gamma}$ $10^{-5}/cm^3$ sec.	E_γ	$M_{p\gamma}$ $10^{-5}/cm^3$ sec.
10.826	1.35	8.641	1.11
8.309	0.38	7.800	1.64
7.299	0.75	5.420	24.74
6.321	1.50	5.047	1.32
5.560	0.81	4.870	4.80
5.532	1.60	4.639	0.87
5.297	1.67	4.431	1.84
5.267	2.29	3.723	1.18
4.508	1.42	3.370	2.20
3.675	1.40	3.221	11.35
3.531	0.86		

coal in a uniform neutron flux, a useful parameter $M_{p\gamma}$ can be defined such that,

$M_{p\gamma}$ = the number of prompt γ-rays of a given energy emitted per second per unit volume of sample (1 cm^3) per unit thermal neutron flux (1n/cm^2 sec.).

$$= \Sigma \cdot I_\gamma \times 10^{-2} \ \gamma/cm^3 \text{ sec.}$$

where Σ is the macroscopic cross-section for the reaction and

I_γ is the number of γ-rays emitted per 100 neutrons captured.

Values of $M_{p\gamma}$ for the product nuclei sulphur-33 and nitrogen-15 are shown in Table IV. Where necessary, it is a simple matter to provide a similar table for all isotopes present in the sample. Examination of Table IV shows that the 5.42 MeV γ-ray from sulphur-33 should dominate the spectrum for equal numbers of nitrogen and sulphur atoms.

3.1.2 Thermal neutron activation

It is important to consider the production of γ-rays from any radioactive nuclides which can result from thermal neutron capture since they may present a possible method for elemental analysis, or a possible source of interference, or a means of compensating for interference. Apart from this, it is necessary to consider the possibility of induced radioactivity producing a radiological hazard.

It is convenient to compile four tables similar to Table II but including the half-lives of the product nuclides for (n,γ), (n,p), (n,α) and (n,2n) reactions with non-negligible macroscopic cross-sections and yielding unstable, γ-emitting product nuclei.

Considering (n,γ) reactions only, the product nuclides from from the high-abundance isotopes, ^{13}C, ^{2}H, ^{17}O, ^{15}N and ^{33}S are all stable and it is only necessary to consider the reaction $^{34}S(n,\gamma)^{35}S$, as shown in Table V. The half-life of sulphur-35 is so long that no appreciable radioactivity will accrue and, in addition, only β-particles are emitted.

TABLE V. THERMAL NEUTRON ACTIVATION IN COAL

Element	Product isotope mass number	Macroscopic cross-section of target nuclide cm^{-1}	Half-life days
S	35	1.37×10^{-5}	86.73

In the present example it can be seen from similar considerations that (n,p), (n,α) and (n,2n) reactions can also be ignored for one of the following reasons:-

(i) the cross-section is negligible,

(ii) the half-life of the product nuclide is very small,

(iii) the half-life of the product nuclide is very large, or it is stable,

and (iv) the product nuclide is, or is almost, a pure β-emitter.

For sulphur, for example, we have:-

$^{32}S(n,p)^{32}P$: ^{32}P is a pure β-emitter.

$^{32}S(n,\alpha)^{29}Si$: ^{29}Si is stable.

$^{34}S(n,p)^{34}P$: The macroscopic cross-section is very small.

$^{34}S(n,\alpha)^{31}Si$: ^{31}Si has a long half-life (2.64 hrs) and γ-rays are emitted in only 0.7% of the disintegrations.

If any activation reactions are important, then a table analogous to Table IV can be compiled for each reaction. The parameter of comparison is then defined as $M_{a\gamma}$ where

$M_{a\gamma}$ = the number of activation γ-rays of a given energy emitted per second per unit volume of sample (1 cm^3) per unit thermal neutron flux (1n/cm^2 sec.) after irradiation periods of, say, 10 secs, 100 secs and 5 x $T_{\frac{1}{2}}$ (for $T_{\frac{1}{2}}$ > 100 secs).

3.1.3 Fast neutron inelastic scattering

In general, information on inelastic scatter cross-sections is not readily available, but by ignoring elements in coal other than those given in Table I, the most significant γ-rays, referring to the first excited states above ground level, can be written down as in Table VI. The variations in the inelastic scatter cross-section with neutron energy for excitation of the 1st level of sulphur-32 are shown in Table VII. Excitation could be obtained using an $^{241}Am/B$ source which has a maximum neutron intensity at 3 MeV, approximately, the neutron emission rate being reduced by a factor 5, approximately, at 5 MeV.

It is likely that the γ-rays from carbon, in spite of the higher atomic concentration of carbon in coal, would probably not swamp the γ-rays from sulphur-32. Carbon-12 has a cross-section which rises to 200-400 mb, but even the double escape peak (3.42 MeV) is well separated from the sulphur-32 inelastic scatter γ-ray. The high energy threshold of oxygen (6.13 MeV) and the low cross-section of nitrogen (6 mb at 4 MeV) suggest that these reactions would not interfere significantly. A complete study would require the cross-sections to be properly averaged over all neutron energies in the sample.

TABLE VI. ENERGIES OF THE 1st EXCITED STATES OF THE MAJOR ELEMENTS PRESENT IN COAL

Element	Energy of 1st excited state MeV
^{12}C	4.44
^{2}H	-
^{16}O	6.05* 6.13
^{14}N	2.31
^{32}S	2.24

* Not useful since de-excitation by pair emission only is allowed.

TABLE VII. VARIATION OF NEUTRON INELASTIC SCATTER CROSS-SECTIONS FOR 1st EXCITED LEVEL OF SULPHUR-32

Neutron energy MeV	Inelastic scatter cross-sections mb
3.5	330
3.85	440
4.2	380
4.5	340
4.8	290

As above, a comparison parameter $M_{i\gamma}$ can be used defined such that $M_{i\gamma}$ = the number of prompt γ-rays of a given energy emitted per second, per unit sample volume (1 cm^3) per unit neutron flux (1n/cm^2 sec.), arising from inelastic neutron scattering.

3.1.4 Fast neutron particle reactions

A reliable estimate of the value of an analysis based on this reaction is difficult to make since the energy/intensity spectra of the fast neutron flux should be determined before the reaction rates can be deduced from the differential cross-sections. However, a useful first-step can be made by examining the cross-sections as a function of neutron energy in order to decide whether a specific reaction is worth further consideration as a basis of an analytical technique, or whether it may constitute a potential source of interference to some other preferred reaction.

An examination is based on the neutron sources available and, for field applications, these are restricted to the (α,n) isotopic neutron sources, of which the most important are ^{227}Ac/Be, ^{241}Am/Be, ^{242}Cm/Be, ^{228}Th/Be, ^{241}Am/B,Li, the spontaneous fission source ^{252}Cf and the accelerator neutron source $^{3}H(d,n)^{4}He$. In currently

TABLE VIII. FAST NEUTRON PARTICLE REACTIONS FOR SULPHUR

Reaction	Comments
$^{32}S(n,p)^{32}P$	Cross-section is 200-400 mb over neutron energies 4-16 MeV. But ^{32}P is a pure β-emitter.
$^{32}S(n,t)^{30}P$	Cross-section is 500 mb at 16 MeV, 16 mb at 18 MeV but negligible below 15 MeV.
$^{32}S(n,\alpha)^{29}Si$	Product nucleus is stable.
$^{32}S(n,2n)^{31}S$	Reaction Q = -15.08 MeV.
$^{34}S(n,p)^{34}P$	Cross-section is 75 mb at 14 MeV yielding 2.13 MeV γ-rays (25%) but combined with product half-life of 12.4 sec. and isotopic abundance of 4.22%, overall yield is poor.
$^{34}S(n,\alpha)^{31}Si$	^{31}Si is almost a pure β-emitter.

available neutron tubes, deuteron energies are approximately 100 keV so that the reaction yields neutrons with energies of about 13 to 14 MeV.

The possible particle reactions for sulphur do not appear promising, as can be seen from Table VIII.

3.1.5 COMMENT

From the above considerations, it is clear that a measurement of the 5.42 MeV γ-ray from radiative neutron capture appears to offer the possibility of a measurement of the sulphur content and is worthy of further study. Although γ-radiation of adjacent energies occur in nitrogen-15, the expected intensities are considerably lower than from sulphur-33 for the 4% sulphur concentration assumed. At lower sulphur/nitrogen concentrations, the dominance of the 5.42 MeV γ-ray from sulphur over the γ-rays from nitrogen will not be so great.

However, so far a coal model containing only five elements relating mainly to the carboniferous content of coal has been considered. In reality, natural coal also contains mineral matter at concentrations which vary from a few per cent to 30-40%, depending on the origin of the coal. The composition of the mineral content also varies according to locality, but generally contains significant content of alumina, silica and iron pyrites. A typical analysis of the mineral content of a coal in the United Kingdom is given in Table IX. The considerations which have been given to the 5-element model now need to be extended to include the possible effects of any other elements which may be present.

The practical situation is, in fact, not nearly so bad as may appear since the reason for measuring the sulphur content of coal in a borehole usually excludes an application to coals of high (>10% approx.) mineral content. At the lower concentration, however, the same distribution of elements is likely to occur.

4. OTHER ANALYSES

Although consideration has been restricted to the measurement of sulphur in coal, the problem of identifying a suitable reaction

TABLE IX. TYPICAL DISTRIBUTION OF CONCENTRATIONS OF ELEMENTS IN THE MINERAL MATTER IN COAL

Chemical form	Approximate concentration range % ash*
SiO_2	2 - 53
Al_2O_3	2 - 40
Fe_2O_3	1 - 66
CaO	1 - 33
MgO	1 - 11
TiO_2	1 - 2
Na_2O	1 - 8
K_2O	1 - 5
Mn_3O_4	0 - 2
P_2O_5	0 - 8

* Data on mineral matter is only available from data on the ash residue from coal after burning. Ash represents about 85% of the mineral matter from which it is derived. Sulphur in the mineral matter is lost to the ash by conversion of FeS_2 to Fe_2O_3.

to form the basis of an instrument for use in other logging applications is the same. Apart from variations in the environmental conditions governing the application it is only the wanted and interfering elements which vary. To design a probe, for example, for use in one iron field, measurements of iron (58% to 68%), phosphorus (0.05% to 0.5%), silicon (0.5% to 8%), aluminium (0.25% to 5%) and manganese (0.2% to 3%) are required in a matrix also containing varying concentrations of titanium dioxide, calcium oxide, magnesium oxide and water.

The design of a probe to measure fluorite concentration directly (>30%) in the United Kingdom requires consideration to be given to the presence of other minerals such as barite, calcite (mainly) as well as possible small concentrations of galena, sphalerite, pyrite and quartz.

5. DISCUSSION AND RECOMMENDATIONS

5.1 Present status of the preferred nuclear data

Much of the data required in the applications discussed here is available. However, it is not easily retrievable because the sources are, to an appreciable extent, spread through a number of publications and irrelevant data is mixed with that required. In addition, relevant data is often not evaluated and presented as a "best set".

For example, the cross-section data listed in the current CINDA bibliography[9] must be sought out and collated from the references listed. Data, ready assembled, can be obtained from the information given in the CINDU catalogue[10] but the great majority is raw in the sense that a best fit for all the results given is not available. The same comment applies to the plots of cross-sections for a variety of neutron reactions collected and presented in Nuclear Data by Jessen, et al.[11] (a form directly comparable with BNL 325 and Supplements[12]).

Information on thermal neutron capture cross-sections and associated gamma rays are tabulated by Bartholomew, Groshev, et al. in Nuclear Data[13]. Although this data is very valuable for applications of the type referred to, the user is often confronted with the necessity of making an assessment to determine his own recommended value. These publications do contain values of the half-lives of product nuclei although, from the point of view of "activation" analysis they are restricted to (n,γ) reactions.

The "Handbook of Nuclear Data for Neutron Activation Analysis", by Aliev, et al.[14] is a most useful compilation of values of cross-sections, half-lives and radiations from product nuclei. It contains data both for thermal neutrons and for neutrons with energies in the region of 14 MeV, and includes (n,particle) as well as (n,γ) reactions. However, it does not contain data for

intermediate energies, nor information relating to the energy and emission intensities of prompt gamma rays following neutron capture.

In general, cross-sections for inelastic neutron scattering can be obtained from, say, BNL-325[12] or from the references quoted in the Nuclear Physics compilations of Energy Levels of Light Nuclei by Ajzenberg-Selove and Lauritsen[15]. For these applications, it is important that the cross-sections for particular excited levels are given, and this is in contrast to presently available data which generally identifies the total scatter cross-sections.

5.2 Preferred presentation

The compilation and presentation of data in a form most suitable for applications of the type described above would embody all the information necessary to make both a quick assessment and a detailed study of the most promising method. The information should have been evaluated to yield recommended values and it should not only be in a form which allows an experienced person to scan the data and make the first selections of promising reactions but it should also allow any data which is irrelevant to the problem to be discarded rapidly.

A system of data tabulation which meets the above requirements regarding content and form is recommended as follows.

Each nuclide would be associated with a data set containing information which identifies the nuclide and provides relevant characteristics for reactions with respect to thermal neutrons and to neutrons in the energy range from thermal to 14 MeV. Each data set would consist of the following sub-sets.

(i) Nuclide characterisation

1. Nuclide symbol and atomic number.
2. Mass whole number.
3. Natural abundance of each isotope.

(ii) Thermal neutron data

4. List of possible neutron reactions for each isotope.
5. Half-life of product nucleus or isomer following each reaction.

6. Average cross-section for each reaction (natural isotopic mixture).

7. Average cross-section for each reaction with respect to each isotope.

8. Energies of prompt γ-rays and their relative intensities for each reaction.

9. Energies of γ-rays and their relative intensities for each product nucleus or isomer.

(iii) Data for neutron energies between thermal and 14 MeV

10. List of threshold energies for all possible reactions.

11. Half-life of product nucleus or isomer following each reaction.

12. Cross-section for each reaction with respect to each isotope as a function of energy[1].

13. Energies of prompt γ-rays and their relative intensities for each reaction.

14. Energies of γ-rays and their relative intensities for each product nucleus or isomer.

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[1] For rapid assessments, this data should be available in a graphical form, but for detailed calculations it should be computer compatible and is probably most conveniently stored (as in some present data sets) as pairs of values of energy and cross-section which define the best curve through the available data.

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DISCUSSION

J.A. CZUBEK: What kind of boundary conditions do the authors use for theoretical calculations when applying neutron methods to dry boreholes, which is a frequent case in solid mineral exploration?

A.G.T. FERGUSON: In general, the calculations are based on a Monte-Carlo code for cylindrical systems, so that the problem of boundary conditions does not apply.

J.A. CZUBEK: Is the spectrometry of gamma radiation from the radiative capture of thermal neutrons used for the exploration of certain minerals? If so, could you tell me too for which minerals and up to what grade, because in nuclear geophysics there are some differences with respect to conventional laboratory methods. In nuclear geophysics the final signal is proportional to the ratio $(\Sigma_{()}^{i}/\Sigma_{\alpha}$, where $\Sigma_{()}^{i}$ is the cross-section for a given i-th isotope for a given reaction, whereas Σ_{α} is the total absorption cross-section of the rock. For such a situation, where the ore grade is high, the calibration curve is no longer linear.

A.T.G. FERGUSON: The intention is to use the spectrometry of gamma rays from thermal neutron capture in exploration but as yet neither the range of element nor the concentrations at which operations are to be carried out has been established.

J.A. CZUBEK: Do you have some special neutron unit for standardizing your measurements (so as to be able to compare results obtained in the same region but with different kinds of probes) or do you use a curve calibrated in units cps versus concentration of individual element in the ore?

A.T.G. FERGUSON: The design of a facility for use in calibrating each instrument as and when it is developed is now under consideration.

NUCLEAR DATA FOR BREMSSTRAHLUNG ACTIVATION ANALYSIS OF VALUABLE TRACE ELEMENTS IN THE COPPER ORE

W. RATYŃSKI, A. STEGNER, Z. SUJKOWSKI
Institute of Nuclear Research,
Department of Nuclear Physics,
Świerk near Otwock, Poland

Abstract

NUCLEAR DATA FOR BREMSSTRAHLUNG ACTIVATION ANALYSIS OF VALUABLE TRACE ELEMENTS IN THE COPPER ORE.

Bremsstrahlung activation analysis with the help of a betatron has recently been applied successfully to continuously controlling the copper-ore mining process. In this paper, the possibility of applying the same method to the analysis of valuable elements contained in the waste material left after copper processing is discussed. The available nuclear data relevant to the analysis are collected. A user-oriented form of presentation of these data is proposed. Further data needs are pointed out. The data presented concern the elements sought as well as those elements which contribute to the background. Detailed data analysis is given for the elements Mo, Ti and Rh. Some auxiliary yield measurements have been carried out and the results are presented in tabular and graphical form. Complementary methods of activation and fluorescence analysis are discussed in connection with the copper-mine or copper-works waste material problem.

1. INTRODUCTION

Photonuclear reactions have been used as a tool for activation analysis in many practical applications. The feasibility of the method and the accuracies obtainable vary greatly and have to be considered individually for each particular case. The range of possible applications has increased very significantly in the last few days, along with rapid progress in high-resolution gamma-ray spectroscopy. This has also brought about requirements for accurate nuclear data concerning the production cross-sections and the decay schemes of the products. The fact that such data are often either not available or scattered in the literature and difficult to retrieve, constitutes a considerable hindrance in developing various practical applications.

An investment in a photonuclear activation facility – e.g. a betatron – in an industrial plant is usually made only if it serves the purpose of controlling either some important stage in the production process or the quality of the main product. Other uses of the facility, which are only potentially possible and which cannot be outlined in detail before the investment is approved, must be ignored in the initial cost analysis. Industrial laboratories having a research budget have more margin for action and may be satisfied with only an approximate economic analysis. In any case, some kind of cost analysis has to precede the installation. An inclusion of various by-products in this analysis may sometimes be a decisive factor in the consideration. This is yet another reason for requiring good relevant nuclear data and nuclear data compilations.

TABLE I. PHOTOACTIVATION PROCESSES IN COPPER AND IN THE MAIN ELEMENTS PRESENT IN THE COPPER ORE AND GIVING RISE TO THE COMMON BACKGROUND

Target nuclide	Isotopic abundance	Reaction	Threshold	Maximum cross-section and corresponding energy	Reaction product	Half-life T	Decay mode	Main γ-lines and their intensities (% per disintegration)	References
1	2	3	4	5	6	7	8	9	10
^{27}Al	100%	(γ, n)	13.3 MeV	13 mb (20 MeV)	^{26}Al	6.4 s	β^+	511 keV (200%)	10
^{16}O	99.8%	(γ, n)	15.6 MeV	7 mb (22.2 MeV)	^{15}O	124 s	β^+	511 keV (200%)	8, 5
		(γ, 2n)	29 MeV	≲2.5 mb	^{14}O	71 s	β^+	511 keV (199%) 2311 keV (100%)	9, 5
^{12}C	98.9%	(γ, n)	18.7 MeV	7 mb (22.5 MeV)	^{11}C	20.3 m	β^+	511 keV (200%)	8, 5
		(γ, n)	32 MeV	1.5 mb (35 MeV)	^{10}C	19.4 s	β^+	511 keV (200%) 717 keV (98.3%) 1030 keV (1.7%)	8, 5
^{28}Si	92.2%	(γ, n)	17.2 MeV	10 mb (19.8 MeV	^{27}Si	4.2 s	β^+	511 keV (200%)	9, 5, 10
^{29}Si	4.7%	(γ, p)	12.3 MeV	33 mb (20.5 MeV)	^{28}Al	2.3 m	β^-	1780 keV (100%)	11, 5

1	2	3	4	5	6	7	8	9	10
^{30}Si	3.1%	(γ, p)	13.7 MeV	31 mb (21.5 MeV)	^{29}Al	6.6 m	β^-	1280 keV (94%) 2430 keV (6%)	11, 5
		(γ, np)	22.9 MeV		^{28}Al	2.3 m	β^-	1780 keV (100%)	11, 5
^{24}Mg	78.6%	(γ, n)	16.5 MeV	6.7 mb (19 MeV)	^{23}Mg	12 s	β^+	439 keV (9%) 511 keV (200%)	11, 5, 12
^{25}Mg	10.2%	(γ, p)	12.1 MeV	27 mb (20.5 MeV)	^{24}Na	15 h	β^-	1369 keV (99%) 2754 keV (99%)	11, 5
		(γ, 2n)	23.9 MeV		^{23}Mg	12 s	β^+	439 keV (9%) 511 keV (200%)	11, 5
^{26}Mg	11.2%	(γ, p)	14.3 MeV	25 mb (22 MeV)	^{25}Na	60 s	β^-	400 keV (14.2%) 580 keV (14.2%) 980 keV (14.8%) 1610 keV (6%)	11, 5
		(γ, np)	23.2 MeV		^{24}Na	15 h	β^-	1369 keV (99%) 2754 keV (99%)	11, 5
^{32}S	95.1%	(γ, n)	15.1 MeV	12 mb (20 MeV)	^{31}S	2.7 s	β^+	511 keV (200%) 1270 keV (1.1%)	5, 10
		(γ, np)	21.2 MeV		^{30}P	2.5 m	β^+	511 keV (200%) 2230 keV (0.5%)	5, 15
^{33}S	0.7%	(γ, p)	9.6 MeV		^{32}P	14.3 d	β^-		5, 15
^{34}S	4.2%	(γ, p)	10.9 MeV		^{33}P	25 d	β^-		5, 15

TABLE I. (cont.)

Target nuclide	Isotopic abundance	Reaction	Threshold	Maximum cross-section and corresponding energy	Reaction product	Half-life T	Decay mode	Main γ-lines and their intensities (% per disintegration)	References
1	2	3	4	5	6	7	8	9	10
^{40}Ca	97%	(γ, n)	15.7 MeV	16 mb (19.5 MeV)	^{39}Ca	0.87 s	β^+	511 keV (200%)	5, 13, 14
		(γ, 2n)	~28 MeV	~3 mb (~48 MeV)	^{38}Ca → ^{38m}K	0.66 s 0.95 s	β^+ β^+	511 keV (200%) 511 keV (200%)	5, 14
		(γ, np)	24.3 MeV	~2.5 mb (≳28 MeV)	^{38m}K ^{38}K	0.95 s 7.7 m	β^+ β^+	511 keV (200%) 511 keV (198%) 2170 keV (99%)	5, 14
^{44}Ca	2.1%	(γ, p)	12.2 MeV		^{43}K	22.4 h	β^-	220 keV (4%) 338 keV (8%) 374 keV (99%) 394 keV (2%) 594 keV (16%) 619 keV (99%)	5, 15
^{43}Ca	0.15%	(γ, p)	10.7 MeV		^{42}K	12.4 h	β^-	1524 keV (18%)	5, 15

1	2	3	4	5	6	7	8	9	10
		(γ, p)			^{47}K	17.5 s	β^-	2010 keV (84%) 2600 keV (15%)	5
^{48}Ca	0.18%	(γ, n)	10.1 MeV		^{47}Ca	4.5 d	β^-	490 keV (7%) 810 keV (7%) 1330 keV (76%)	
					^{47}Sc	3.4 d	β^-	160 keV (73%)	
		(γ, n)	10.8 MeV	70 mb (16.5 MeV)	^{62}Cu	9.8 m	β^+	511 keV (185%) 880 keV (0.3%) 1130 keV (0.1%) 1172 keV (0.3%)	7, 5
^{63}Cu	68.9%	(γ, 2n)	19 MeV	15 mb (24 MeV)	^{61}Cu	3.3 h	β^+, EC	67 keV (2.3%) 284 keV (24%) 380 keV (2.5%) 511 keV (195%) 580 keV (1.5%) 665 keV (11%) 940 keV (1.4%) 1150 keV (0.5%) 1190 keV (4.7%)	7, 5
^{65}Cu	31.1%	(γ, n)	9.9 MeV	78 mb (17 MeV)	^{64}Cu	12.8 h	β^+, EC, β^-	511 keV (38%) 1340 keV (0.5%)	7, 5

This work has been undertaken to collect the main nuclear data which may be of interest for extending the applications of the betatrons newly installed at the copper ore mines from controlling the mining process to analysing the processed ore samples and the contents of the waste material. Several valuable elements in trace amounts are known to be present in the wastes and recovery of some of them may prove to be economically advantageous. Numerous inadequacies of the existing data became apparent in course of the work. The purpose of the present paper is twofold: a) to discuss the problem of user-oriented data presentation, i. e. the kind of data needed and the form of presentation, and b) to present partial results of data analysis and of auxiliary measurements for selected elements encountered in the copper ores.

To the authors' knowledge there are neither user-oriented nor purely scientific up-to data compilations on photoactivation processes. Useful sources of general information are the relatively recent bibliography of the subject [1] and a review article [2]. A search through the original literature is, however, necessary for each particular element to be analysed.

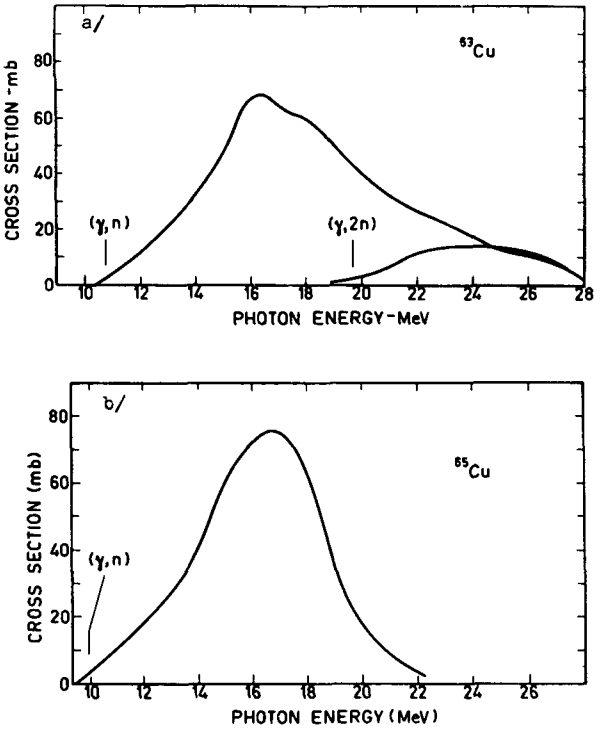

FIG. 1a, b. Cross-sections for (γ, n) and (γ, 2n) reactions on Cu-isotopes as a function of photon energy according to Ref. [7]. A monochromatic photon beam from annihilation in flight was used in the original work.

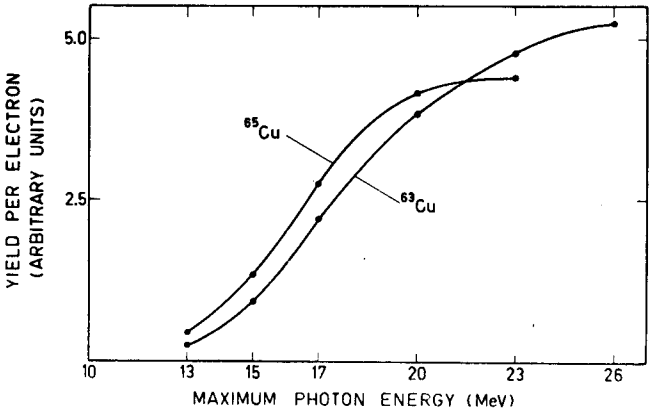

FIG. 2. Photoactivation yield curves per electron obtained from Fig. 1.

2. GENERAL DATA

In a concise form, Table I presents the main characteristics for the photoactivation process for copper and for elements present in the copper ore and contributing to the background. The entries can be divided into two groups: reaction characteristics (type of reaction, threshold, maximum cross-section and corresponding energy) and reaction product characteristics (half-life, type of decay and energies and relative intensities of main gamma transitions). It is seen that except for the copper isotopes, all the β^+-active species produced are either very short-lived or have a high-energy reaction threshold. This allows a sensitive detection technique based on measuring in coincidence the annihilation radiation from the 9.8 min ^{63}Cu isotope to be developed.

3. DETAILED DATA: COPPER

Figures 1a, b show the differential cross-sections for the three reactions of interest: ^{63}Cu(γ, n), ^{63}Cu$(\gamma, 2n)$ and ^{65}Cu(γ, n) (see Ref. [7]). Figure 2 shows the corresponding integrated cross-sections (yield curves).

4. DETAILED DATA: MOLYBDENUM

4.1. Compiled data

Table II presents the main data for Mo isotopes. Figures 3 and 4 show the yield curves for reactions leading to radioactive products and the corresponding differential cross-sections, respectively (see Ref. [3]).

TABLE II. PHOTOACTIVATION PROCESSES IN NATURAL Mo

Target nuclide	Isotopic abundance	Reaction	Maximum cross-section and corresponding energy	Reaction	Half-life T	Decay mode	Main γ-lines and their intensities (% per disintegration)
1	2	3	4	5	6	7	8
^{100}Mo	9.6%	(γ, p) (γ, n)	178 mb (15 MeV)	^{99}Nb → ^{99}Mo → ^{99m}Tc	10 s; 2.4 m 67 h 6 h	β⁻ β⁻ γ	181 keV (9.5%), 740 keV (12%) 141 keV (88%)
^{98}Mo	23.8%	(γ, p)	19.4 mb (20 MeV)	^{97}Nb	72 m	β⁻	659 keV (98%)
^{98}Mo ^{97}Mo	23.8% 9.5%	(γ, np) (γ, p)	7.8 mb (28 MeV) 26 mb (20 MeV)	^{96}Nb	23.4 h	β⁻	459 keV (26%), 569 keV (54%), 778 keV (98%), 1092 keV (48%), 1200 keV (20%)
^{92}Mo	15.8%	(γ, n)	76 mb 49 mb (17 MeV)	^{91m}Mo → ^{91}Mo	66 s 15.5 m	γ, EC, β⁺ EC, β⁺	511 keV (76%), 652 keV (57%), 1210 keV (16%), 1530 keV (22%), 511 keV (188%)
^{92}Mo ^{92}Mo	15.5% 15.8%	(γ, 2n) (γ, np)	28 mb (29 MeV)	^{90}Mo → ^{90}Nb	5.7 h 14.6 h	EC, β⁺ EC, β⁺	122 keV (100%), 163 keV (9.4%), 257 keV (90%), 511 keV (50%) 141 keV (100%), 511 keV (108%), 1140 keV (97%), 2320 keV (91%)

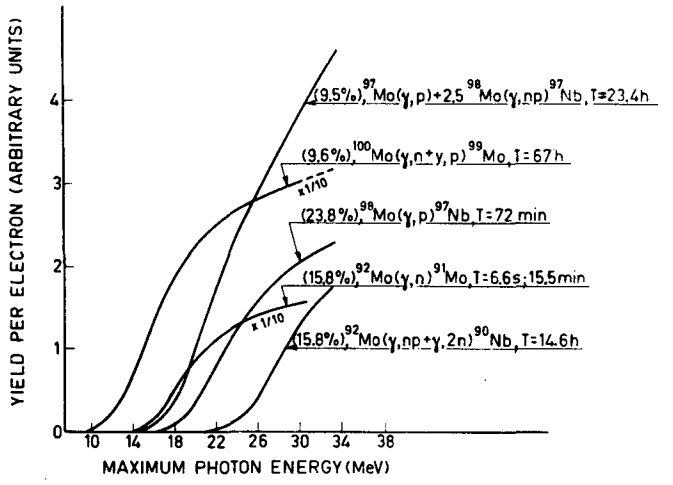

FIG. 3. Photoactivation yield curves for Mo-isotopes according to Ref. [3].

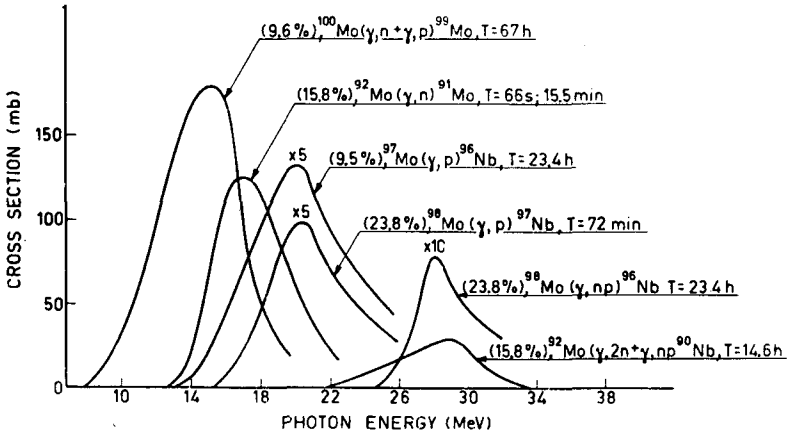

FIG. 4. Cross-sections for photoreactions on Mo-isotopes as a function of photon energy [3].

4.2. Measurements

The following reaction products obtained from bremsstrahlung irradiation of natural Mo may be considered particularly useful for the activation analysis by means of standard Ge(Li) spectrometry: ^{91m}Mo, ^{99}Mo and ^{97}Nb. Several experiments using the 30-MeV betatron of the INR in Świerk were done to check this conclusion.

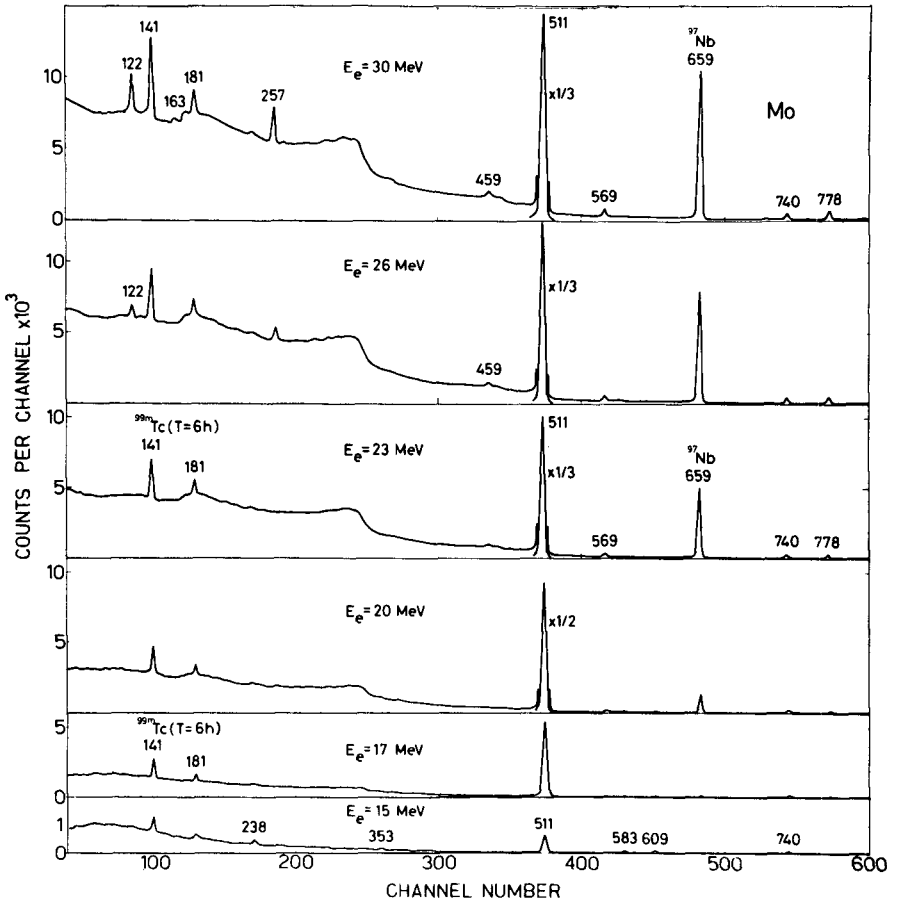

FIG. 5. Gamma-ray spectra of long-lived products of natural Mo irradiated with bremsstrahlung of various maximum energies.

Short-lived products may be advantageous to study in some analyses. The 652-keV γ-ray de-exciting the 66-s ^{91m}Mo isomer was, therefore, measured for the maximum bremsstrahlung energies varying between 15 and 26 MeV. Samples of about 5 g natural Mo were irradiated for 2 minutes together with a Cu sample serving as a flux monitor (the reaction ^{63}Cu$(\gamma, n)^{62}$Cu). The gamma-ray spectra were then measured in standard conditions using a 20-cm^3 Ge(Li) spectrometer placed in a lead shielding. The minimum detectable amount of natural Mo was found to be 0.100 g for a total photon flux of 5.7×10^8 cm$^{-2} \cdot$ s^{-1}. In the real conditions of the Mo-containing wastes there are a number of other short-lived activities contributing to the background, and the sensitivity of the method is still much lower. The method must thus be considered impractical.

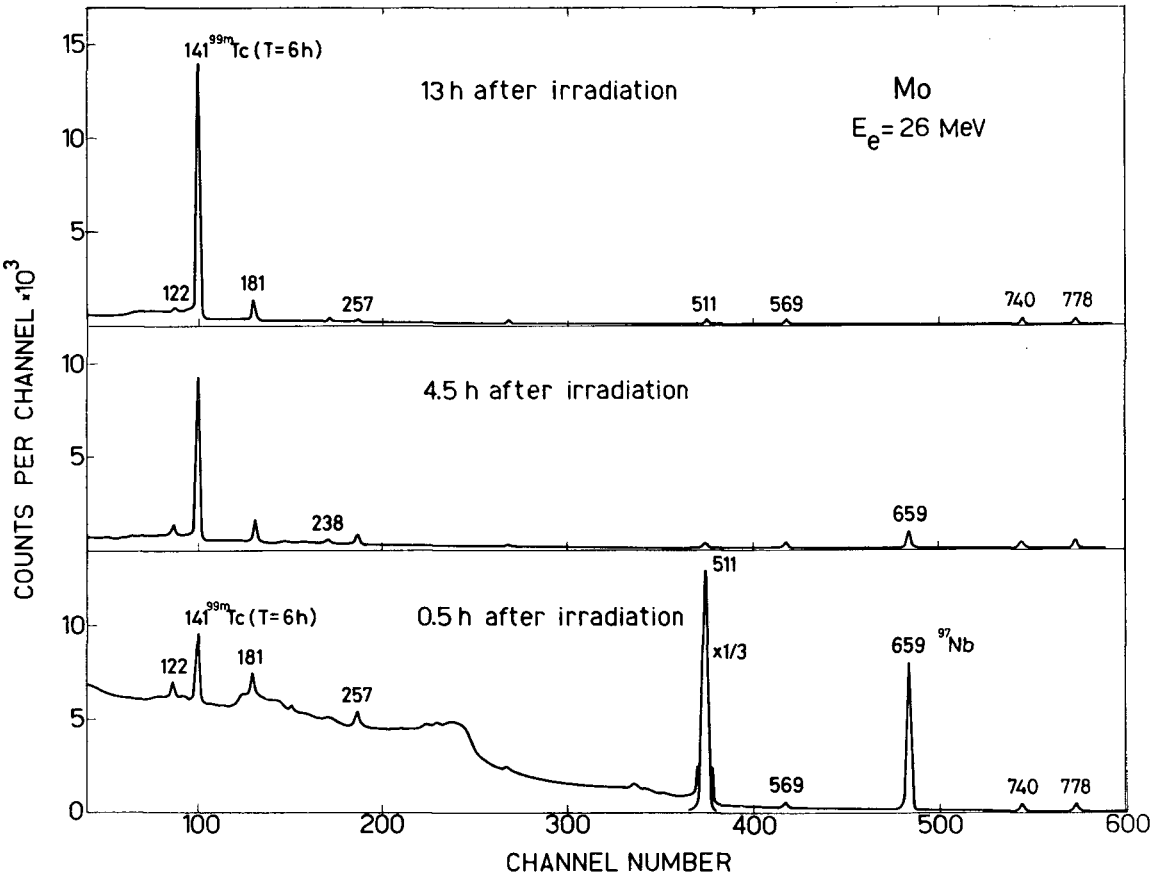

FIG. 6. Gamma-ray spectra of Mo + γ reaction products illustrating the growth in time of the 141-keV gamma line.

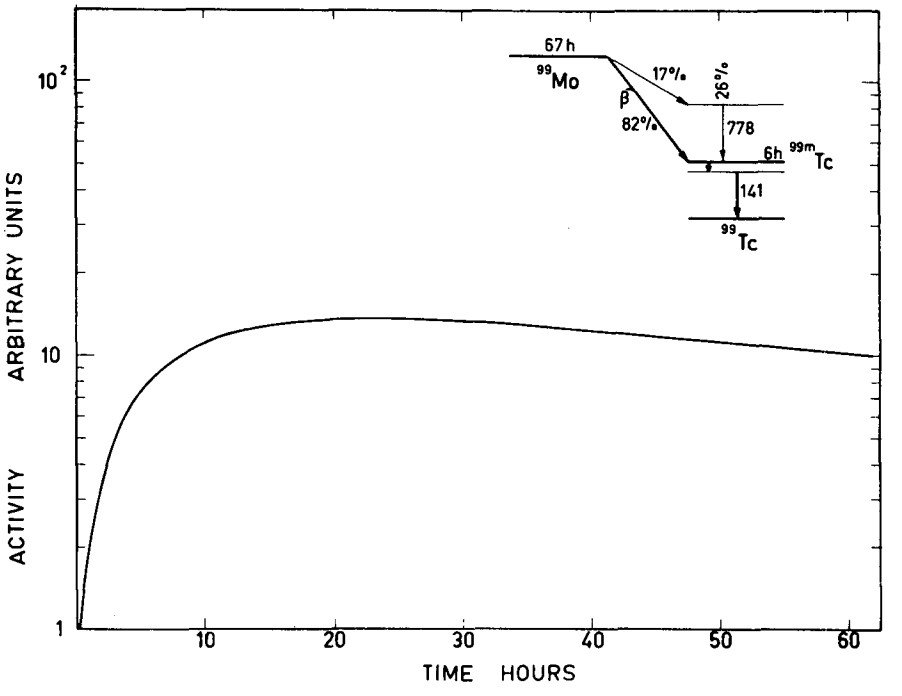

FIG. 7. Time dependence of the intensity of the 141-keV gamma line. Inset shows the decay scheme of ^{99}Mo leading to formation of the ^{99m}To-isomer.

Figure 5 shows the gamma-ray spectra of the long-lived products of Mo, irradiated with bremsstrahlung of E_{max} varying between 15 and 30 MeV. The irradiations lasted 72 minutes each, the spectra were measured during 60 minutes runs starting 30 minutes after the irradiations. The ^{65}Cu(γ, n) reaction was used for the flux monitoring. Several strong, well-separated lines are seen in the spectra. Detection of the 659-keV γ-line from the 72-min ^{97}Nb activity yields the minimum detectable amount of natural Mo of 0.026 g (in the experimental conditions quoted and for E_{max} = 23 MeV and a photon flux of 5.7×10^8 cm$^{-2} \cdot$ s^{-1}; the background was assumed equal to that contributed by a 74.6-g copper ore sample of 0.04% Cu content, irradiated and measured in the same experimental conditions). An order-of-magnitude improvement is easily attainable when a larger detector and stronger flux are used.

The 141-keV gamma-line is of interest. This line de-excites the 6-h ^{99m}Tc isomer produced in the 67-h β^--decay of ^{99}Mo. The line shows, therefore, a characteristic growth in time during the first 20 hours after the irradiation (see Figs 6 and 7). This fact, plus the rather high cross-section and high detector efficiency at 141 keV gamma-ray energy, enable a sensitive quantitative analysis of the Mo content to be made. In the present conditions, a 1-h measurement starting 13 hours after an

TABLE III. PHOTOACTIVATION PROCESSES IN NATURAL Ti

Target nuclide	Isotopic abundance	Reaction	Maximum cross-section and corresponding energy	Reaction product	Half-life T	Decay mode	Main γ-lines and their intensities (% per disintegration)
1	2	3	4	5	6	7	8
^{46}Ti	7.9%	(γ, np)	2.5 mb (28.3 MeV)	^{44}Sc	3.9 h	β^+, EC	511 keV (19.8%), 1159 keV (99%)
^{48}Ti	73.9%	(γ, p)	28.5 mb (22.5 MeV)	^{47}Sc	3.4 d	β^-	160 keV (73%)
^{49}Ti	5.5%	(γ, np)					
^{49}Ti	5.5% 1	(γ, p)	11 mb (21.7 MeV)	^{48}Sc	44 h	β^-	178 keV (6%), 983 keV (100%)
^{50}Ti	5.3%	(γ, np)					1040 keV (100%), 1313 keV (100%)
^{47}Ti	7.3%	(γ, p)	24 mb (21 MeV)	^{46m}Sc ↓ ^{46}Sc	20 s	γ	142 keV (100%)
^{48}Ti	73.9%	(γ, np)			83.9 d	β^-	889 keV (100%), 1120 keV (100%)
^{46}Ti	7.9%	(γ, n)	31.5 mb (20.5 MeV)	^{45}Ti	3.09 h	β^+, EC	511 keV (168%)
^{47}Ti	7.3%	(γ, 2n)					
^{103}Rh	100%	(γ, n)	200 mb (16 MeV)	^{102}Rh	206 d	β^+, EC, β^-	475 keV (55%), 628 keV (4%)
					2.9 y	EC	418 keV (13%), 475 keV (95%)
							628 keV (7.5%), 632 keV (47%)
							698 keV (41%), 768 keV (30%)
							1047 keV (40%), 1113 keV (17%)
		(γ, 2n)	44 mb (20 MeV)	^{101m}Rh ↓ ^{101m}Rh	4.5 d	EC, γ	157 keV (10%), 307 keV (82%)
					3 yr	EC	

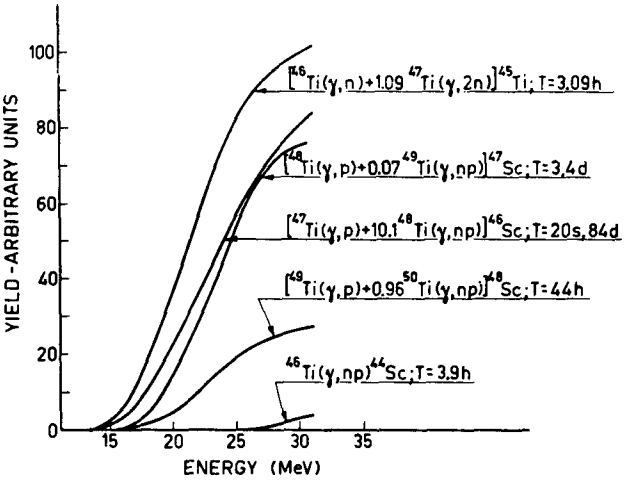

FIG. 8. Photoactivation yield curves for Ti-isotopes according to Ref. [4].

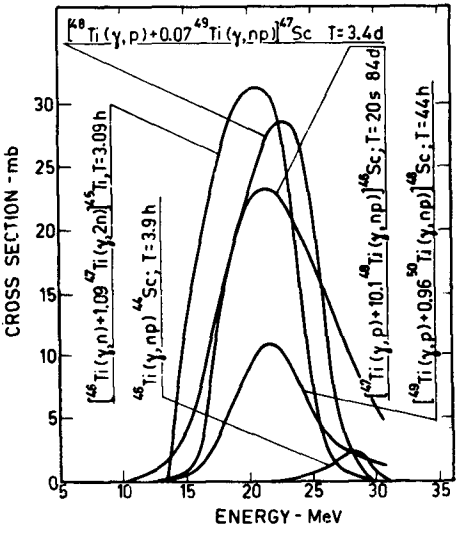

FIG. 9. Cross-sections for photoreactions on Ti-isotopes as a function of photon energy [4].

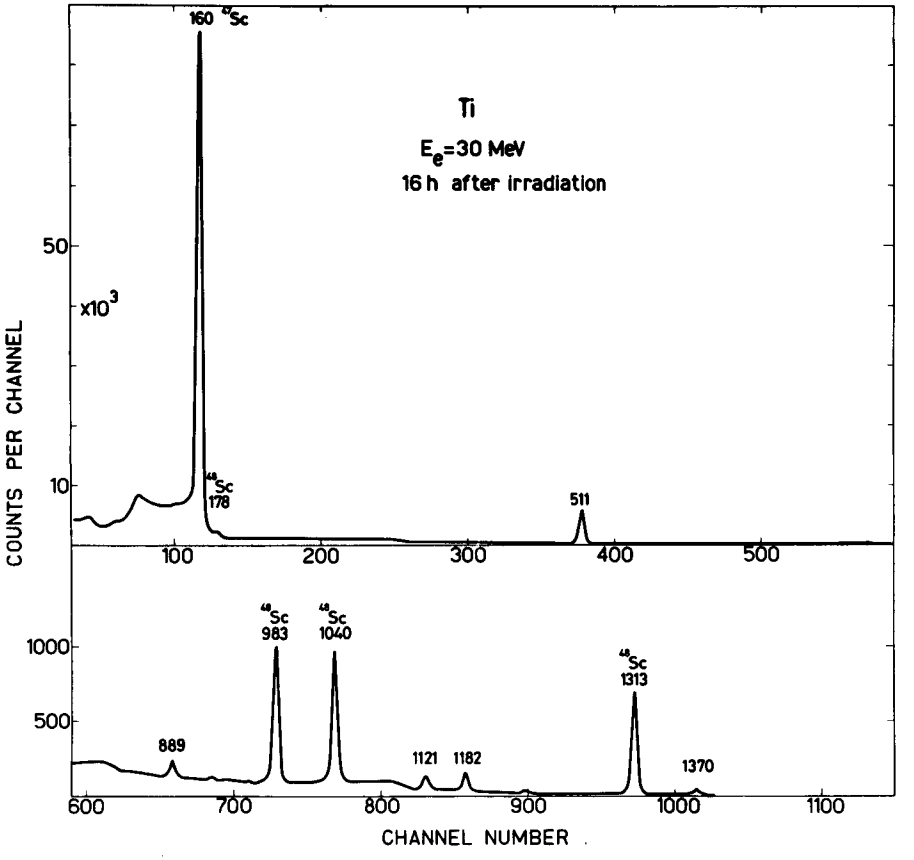

FIG. 10. Example of gamma-ray spectra of Ti + γ reaction products.

irradiation of about 1 h permits detectability to reach down to 0.034 g of natural Mo. In a more elaborate experimental set-up and making use of the growth of activity (i.e. doing two experiments, e.g. at about 5 h and 24 h after irradiation) an improvement of two orders of magnitude seems to be attainable relatively easily.

5. DETAILED DATA: Ti AND Rh

Table III presents the main data for Ti and Rh isotopes. Figures 8 and 9 show the yield curves for bremsstrahlung activation and the cross-sections for reactions on natural Ti leading to radioactive products [4].

The 3.4-d ^{47}Sc and 1.8-d ^{48}Sc product activities are most promising for the standard activation analysis. Figure 10 shows the γ-ray spectrum measured 16 h after 2.5 h of irradiation. The 160-keV transition from

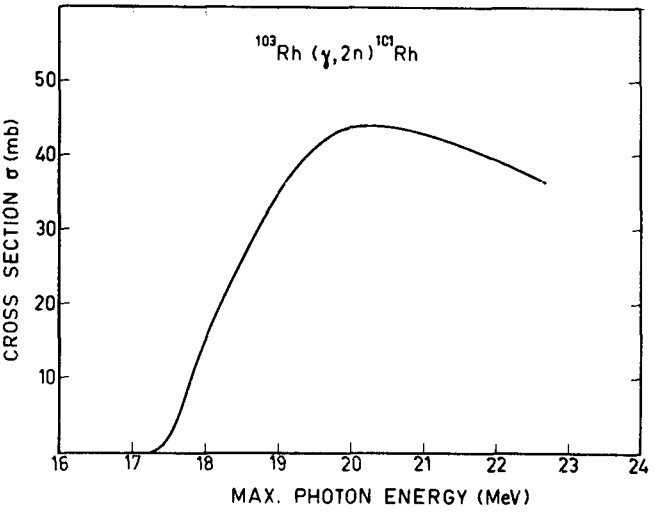

FIG. 11. Cross-section for (γ, 2n) reaction on Rh.

^{47}Sc dominates the spectrum. Using this line, the detectability of Ti can be determined to be 0.040 g. The corresponding figure based on the high-energy lines of ^{48}Sc is 0.200 g.

Only the ^{103}Rh (γ, 2n)^{101}Rh reaction may practically be used for the activation analysis of rhodium content. Figure 11 shows the reaction cross-section according to Ref. [16]. It may be pointed out that the high-energy threshold is rather unfavourable for the activation analysis because of the relatively high background.

Another possibility is to detect the traces of Rh using the ^{103}Rh(n, γ)^{104m} Rh reaction with thermal neutrons (see below).

6. COMPLEMENTARY METHODS

The bremsstrahlung activation facility may also be used as a neutron source. The thermalized neutron flux from an ordinary betatron is not very high, but often sufficient for activation of isotopes with high production yields. This was checked using the INR betatron. Activation of, e.g., ^{52}V (3.8 min, E = 1434 keV) in the (n, γ) reaction was found to be feasible. It may, therefore, be of interest for the betatron user to have a listing of possible (n, γ) activations for the same elements as are considered for the photoactivation analysis. Detailed consideration of the (n, γ) activation method is beyond the scope of this paper.

A very powerful and accurate quantitative trace analysis can be done by using the characteristic X-rays following bombardment of a sample with charged particles or photons of suitable energy (fluorescence analysis). This method is limited to rather thin surface layers. It may, therefore, be applied to samples of good homogeneity. The method is most advantageous when high-Z traces are searched for in the bulk of low-Z material. Such

TABLE IV. BASIC DATA FOR BREMSSTRAHLUNG ACTIVATION ANALYSIS [a]

Target element	Reaction product	Half-life	Activation			Measurement (Ge (Li) detector)		Detectability (mg)
			Maximum energy (MeV)	Irradiation time	Waiting time	Measuring time	Main γ-lines E (MeV) (Relative intensity per decay (%))	
Mo	^{97}Nb	72 min	23	60 min	30 min	60 min	0.659 (98)	20
Mo	^{99}Mo	67 h	26	60 min	10 h	60 min	0.141 (88)	30
	^{99m}Tc	6h		60 min	5 h and 24 h	60 min 60 min	0.141 (88)	2

[a] Flux assumed: 6×10^8 photons/cm^2·s

situations are encountered in copper-ore processing. An installation of a fluorescence analysis set-up as a complementary facility in an industrial betatron laboratory may, therefore, be considered to be a relatively cheap and useful extension of the analysing possibilities.

7. CONCLUSIONS

A user-oriented compilation on photoactivation processes is clearly needed. The extent of such a compilation, the form of data presentation, the choice of entries, etc. are important items if the compilation is really to serve its purpose. In our opinion, such a compilation should consist of two parts: Part I should contain a concise table with information giving a kind of brief recipe to be followed for finding optimum conditions of detecting each element. This could be illustrated – but would not have to – by typical γ-ray spectra. Comments on, e. g. the detectabilities in standard conditions, would be optional. An example of such a table is given below (Table IV) with data selected for the few elements discussed earlier.

Part II would present the data for a more advanced user. Cross-sections, yield curves and selected decay schemes would be in place here as well as comments on particular methods to be used, provided that they are not too specialized (e. g. coincidence techniques, etc.). Data for some very special techniques, e. g. making use of short-lived isomeric states in the milli- and micro-second range for the activation analysis purposes (with beam pulsing, etc.) should not be presented in such a general-purpose compilation, but reference to those techniques should be given whenever applicable.

As mentioned earlier, the compilation should be amended with selected (n, γ) data, having in mind that a bremsstrahlung facility may also be used as a neutron source.

We wish to acknowledge the co-operation of J. Janiczek, and the betatron group. We also express our gratitude to J. Mordalski for several stimulating discussions.

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DISCUSSION

G.A. BARTHOLOMEW: Do the mines have available specially designed betatrons for this application or do they use a general-purpose model?

Z. SUJKOWSKI: The betatrons have been made especially for the mines but changes and adaptations compared with a general purpose betatron are minor. Care was taken that the industrial version should be simple to operate and allow a high degree of automatization.

D. BERÉNYI: What are the time requirements for the method, including sample preparation?

Z. SUJKOWSKI: I take it that you are referring to the copper ore samples. Pulverized samples are packed into standard plastic containers, transported to the betatron, irradiated for a few minutes and then transported to the detection set-up. All this is done automatically and does not take much time. The transport time is less than a minute. The measuring time is 5 - 10 min. Only the sample changing at the detectors and one button pushing per sample are manual. Three samples are usually irradiated at the same time in one betatron and then measured in three coincidence arrangements. If you wanted to obtain equivalent information using the conventional methods of wet chemistry it would require several (I believe about 10) hours per sample and considerably more man-power.

I.A. KONDUROV: The paper presented by Dr. Sujkowski suggests that the best way to satisfy hard-to-please users is to give them not a restricted paper compilation but a special program and an input data file for calculating the content of the isotopes of interest from the γ-yield for a given primary radiation energy, time of irradiation, etc.

ЭНЕРГИИ И ИНТЕНСИВНОСТИ γ-ЛУЧЕЙ ДЛЯ КАЛИБРОВКИ Ge (Li) ДЕТЕКТОРОВ В ДИАПАЗОНЕ 3-10 МэВ

Л.Л.СОКОЛОВСКИЙ, А.Е.ИГНАТОЧКИН
Институт атомной энергии им.И.В.Курчатова,
Москва,
Союз Советских Социалистических Республик

Представлен Ф.Е.Чукреевым

Abstract–Аннотация

GAMMA RAY ENERGIES AND INTENSITIES FOR CALIBRATING Ge(Li) DETECTORS IN THE RANGE 3-10 MeV.

Ge(Li) detectors are being used more for measuring gamma ray spectra over a wide energy range. The high resolution of such detectors entails certain requirements as the accuracy of the calibration curves, so that it is essential to establish an appropriate set of standards for the energy and intensity calibration of Ge(Li) detectors at energies up to 10 MeV. At energies up to 3 MeV, Ge(Li) detectors are calibrated using the gamma rays from sources whose energies and intensities are well known from recent precision measurements performed with magnetic and crystal spectrometers. Howerer, the range of gamma energies investigated in nuclear reactions extends to 10 MeV, and as there are very few calibration curves for these higher energies it becomes much more difficult to work with Ge(Li) detectors. The author discusses the present situation as regards the calibration of Ge(Li) detectors in the range 3-10 MeV and the standards used by various workers in determining the energy of a particular radiation and measuring the relative efficiency of Ge(Li) spectrometers. Revised tables are presented of gamma transition energies and intensities in the reaction (n, γ) for the elements most frequently used as standards.

ЭНЕРГИИ И ИНТЕНСИВНОСТИ γ-ЛУЧЕЙ ДЛЯ КАЛИБРОВКИ Ge(Li) ДЕТЕКТОРОВ В ДИАПАЗОНЕ 3-10 МэВ.

Ge(Li) детекторы находят в настоящее время все большее применение при измерении спектров γ-лучей в широкой области энергий. Высокое разрешение этих детекторов накладывает определенные требования на точность калибровочных линий, и поэтому необходимо установить подходящий набор стандартов для калибровки Ge(Li) детекторов по энергии и интенсивности в диапазоне энергий вплоть до 10 МэВ. В области до 3 МэВ калибровка Ge(Li) детекторов проводится при помощи γ-лучей радиоактивных источников, энергии и интенсивности которых хорошо известны из недавних прецизионных измерений, выполненных на магнитных и кристаллических спектрометрах. Однако диапазон γ-лучей, исследуемых в ядерных реакциях, простирается до 10 МэВ и здесь калибровочные линии чрезвычайно редки, что существенно усложняет работу с Ge(Li) детекторами в этом диапазоне энергии. В настоящей работе рассматривается современное состояние по калибровке Ge(Li) детекторов в диапазоне 3-10 МэВ. Проводится анализ используемых различными авторами стандартов для определения энергии исследуемого излучения, а также измерения относительной эффективности Ge(Li) спектрометров. Приведены пересмотренные таблицы энергий и интенсивностей γ-переходов в реакции (n, γ) для элементов, наиболее часто используемых в качестве стандартов.

В течение последних нескольких лет германиевые детекторы находят все большее применение при измерении гамма-спектров в широкой области энергий. Высокое разрешение этих детекторов накладывает определенные требования на точность калибровочных линий, и поэтому необходимо установить подходящий набор стандартов для калибровки Ge(Li) детекторов по энергии и интенсивности в диапазоне энергий вплоть до 10 МэВ.

Анализ существующего положения показывает, что в различных энергетических диапазонах состояние с калибровкой германиевых детекторов существенно отличается. В области энергий до 1 МэВ мы обычно имеем резко ограниченный из-за подходящего периода полураспада набор стандартных радиоактивных источников с хорошо известными значениями энергий гамма-переходов, многократно измеренными с высокой точностью на дифракционных и магнитных спектрометрах [1]. Получение кривой эффективности в этой области энергий также не вызывает особенных затруднений, поскольку для этого необходимо лишь использовать источники с хорошо известной интенсивностью. В области энергий от 1 до 3 МэВ количество обычно используемых стандартных радиоактивных источников существенно меньше. Наиболее подходящим является ^{56}Co [1], так как этот источник обладает достаточно равномерно распределенным по энергетическому диапазону набором интенсивных гамма-переходов с хорошо известными значениями энергий и относительных интенсивностей. Что касается диапазона 3-10 МэВ, то здесь, к сожалению, экспериментаторы не пришли еще к единому мнению о выборе определенного стандарта при калибровке германиевых детекторов. Только анализ работ по радиационному захвату тепловых нейтронов, представленных на международной конференции в Швеции (Студсвик, 1969), показывает, что авторы использовали для калибровки своих спектрометров весьма разнообразный набор элементов-мишеней: Ti, Ni, Co, Cl, Hg, Dy, Fe, N, C, Si, H. Если рассматривать более широкий круг работ, то этот набор еще более увеличится. Причем следует отметить, что в некоторых случаях выбор ядра-мишени в реакции (n, γ), используемой для калибровки германиевого спектрометра, зачастую бывает необоснованным, что приводит к снижению качества полученных результатов.

Однако в последнее время наблюдается некоторая тенденция стандартизовать калибровку германиевых детекторов в диапазоне 3-10 МэВ, используя для этой цели реакцию $^{14}N(n, \gamma)^{15}N$ [1,2]. Подобный набор является достаточно обоснованным, так как эта реакция весьма легко осуществляется на реакторах, а при работе с внутренней мишенью она даже является источником фона. Кроме этого значения энергии гамма-лучей, сопровождающих радиационный захват тепловых нейтронов ядрами азота, равномерно покрывают интервал энергий от 10 до 1,7 МэВ интенсивными линиями. Схема гамма-переходов азота-15 изучена весьма полно, что играет существенную роль при построении кривой эффективности германиевого спектрометра, поскольку возможно использовать двухступенчатые каскады. Эта реакция неоднократно исследовалась, и поэтому имеются надежные данные, полученные с помощью германиевых детекторов, по энергии и интенсивности гамма-переходов в ^{15}N.

В некоторых случаях, когда по ряду причин использование реакции $^{14}N(n, \gamma)^{15}N$ невозможно, следует использовать реакции $^{28}Si(n, \gamma)^{29}Si$ и $^{12}C(n, \gamma)^{13}C$. Эти реакции обладают практически теми же перечисленными выше преимуществами, что и реакция с азотом.

Введение подобного стандарта при калибровке германиевых детекторов существенно улучшит возможность сопоставления результатов различных авторов и позволит более объективно оценивать качество полученных результатов.

Ниже в таблицах I, II и III приведены значения энергий и интенсивностей гамма-переходов из реакций $^{14}N(n, \gamma)^{15}N$, $^{28}Si(n, \gamma)^{29}Si$,

ТАБЛИЦА I. УСРЕДНЕННЫЕ ЗНАЧЕНИЯ ЭНЕРГИИ И ИНТЕНСИВНОСТЕЙ ГАММА-ПЕРЕХОДОВ В РЕАКЦИИ $^{14}N(n,\gamma)^{15}N$ [a)]

E_γ, кэВ	I_γ, %	E_γ, кэВ	I_γ, %
10829,40 ± 0,16	13,79 ± 0,05	3855,13 ± 0,16	0,50 ± 0,22
9920,0 ± 1,9	0,11 ± 0,03	3677,80 ± 0,24	16,12 ± 0,24
9150,8 ± 1,2	1,61 ± 0,03	3532,18 ± 0,18	10,17 ± 0,15
9048,74 ± 0,12	0,23 ± 0,04	3013,78 ± 0,22	0,78 ± 0,16
8569,00 ± 0,64	0,18 ± 0,04	2831,04 ± 0,17	2,06 ± 0,14
8310,47 ±0,28	3,66 ± 0,19	2520,11 ± 0,98	5,96 ± 0,13
7299,01 ± 0,16	9,31 ± 0,27	1999,59 ± 0,17	4,39 ± 0,27
6322,26 ± 0,22	18,18 ± 0,21	1988,92 ± 0,35	1,1 ± 0,5
5562,06 ± 0,38	10,32 ± 0,04	1884,80 ± 0,04	20,43 ± 0,32
5533,07 ± 0,37	18,86 ± 0,11	1853,9 ± 1,2	0,70 ± 0,22
5298,21 ± 0,32	22,06 ± 0,30	1681,48 ± 0,18	1,56 ± 0,10
5269,42 ± 0,68	31,12 ± 0,24	1678,23 ± 0,17	7,83 ±0,54
4508,81 ± 0,12	15,64 ± 0,56		
3883,60 ± 0,70	0,82 ± 0,04		

а) Использовались данные работ [2,3,9-16].

ТАБЛИЦА II. УСРЕДНЕННЫЕ ЭНЕРГИИ И ИНТЕНСИВНОСТИ ГАММА-ЛУЧЕЙ ИЗ РЕАКЦИИ $^{28}Si(n,\gamma)^{29}Si$ [d)]

E_γ, кэВ	I_γ, %	E_γ, кэВ	I_γ, %
8472,21 ± 0,49	2,61 ± 0,14	3954,42 ± 0,04	1,95 ± 0,20
7199,62 ± 0,15	9,0 ± 1,0	3660,93 ± 0,15	3,80 ± 0,20
6444,4 ± 1,4	0,16 ± 0,05	3539,00 ± 0,09	66,5 ± 2,5
6380,01 ± 0,36	13,2 ± 1,3	2426,05 ± 0,38	2,76 ± 0,16
6046,25 ± 0,16	0,66 ± 0,08	2092,85 ± 0,37	20,5 ± 0,50
5107,03 ± 0,57	4,12 ± 0,29	2028,8 ± 1,4	-
4933,85 ± 0,11	63,8 ± 1,8	1273,28 ± 0,12	19,0 ± 1,0

а) Использовались данные работ [3,4,5].

ТАБЛИЦА III. УСРЕДНЕННЫЕ ЭНЕРГИИ И ИНТЕНСИВНОСТИ ГАММА-ЛУЧЕЙ ИЗ РЕАКЦИИ $^{12}C(n,\gamma)^{13}C$[a)]

E_γ, кэВ	I_γ, кэВ
4945,33 ± 0,16	67,0 ± 0,1
3683,93 ± 0,26	31,8 ± 0,1
1261,75 ± 0,23	29,3 ± 0,4

a) Использовались данные работ [6-10].

$^{12}C(n,\gamma)^{13}C$. Приведенные в таблицах значения представляют собой взвешенные средние значения, определяемые формулой

$$\bar{X} = \frac{\sum p_i x_i}{\sum p_i}$$

где x_i – значение энергии или интенсивности гамма-перехода, полученное в i-й работе, а $p_i = 1/\sigma i^2$ – вес измерения. Взвешенное среднее квадратическое отклонение определяется формулой

$$S = \sqrt{\frac{\sum p_i (x_i - \bar{x})^2}{\sum p_i}}$$

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DISCUSSION

J.A. CZUBEK: The paper gives the whole set of gamma lines (which are known with varying degrees of accuracy) for calibration of the energy scale of the solid state detector. In my opinion, however, users are more interested in obtaining an answer to the question of what accuracy can be obtained in the determination of an unknown energy by using the proposed set of energies. From the statistical point of view, this is a rather complicated problem because it involves using the calibration curve in the opposite direction, and the calibration curve is itself defined by the gamma-ray energy set in which each energy is known to a different accuracy.

F.E. CHUKREEV: I think that the regressive analysis procedure would provide an answer to this question, but the result would be dependent on the specific features of each experiment.

G.A. BARTHOLOMEW: Dr. Sokolovsky obtains high precision on his intensities by using weighted averages of the results of various workers. His results seem to conflict somewhat with the points I was making in the presentation of my paper (IAEA-SM-170/37) about low accuracies in absolute intensities, which seem to be more the rule in heavy elements. I would like to make two comments: first, these are light elements with simple spectra and can in fact be used to check the spectrometer efficiency functions. Nevertheless I am still somewhat worried about possible inbreeding of information. Second, some of the problems encountered in heavy elements do not occur in these light elements and this makes it easier to get good agreement in intensities. I would still be inclined to suspect intensity precisions better than about 5%.

F.E. CHUKREEV: I would merely draw attention to the fact that Sokolovsky's results are consistent with the views of Professor Bartholomew. The nucleus ^{15}N belongs to the nuclei which transit to the ground state by emitted simple spectra.

C. WEITKAMP: Speaking from a practitioner's point of view, I would like to stress the importance of using more than one element or isotope for both intensity and energy calibrations when ultimate accuracy of analysis is being aimed at. The main reason for this is the inadequate coverage of the whole energy range, and also mutual perturbation of peaks in many calibration spectra, such as that from the reaction $^{14}N(n,\gamma)^{15}N$.

D.J. HOREN: Have all the transitions been fitted into a decay scheme to check the internal consistency of the intensities?

F.E. CHUKREEV: Yes, of course.

CALCULATION OF SPALLATION YIELDS OF PRODUCTS INDUCED BY THE INTERACTION OF HIGH-ENERGY PROTONS WITH COMPLEX NUCLEI

E. ROSS, K. BÄCHMANN
Fachbereich Anorganische Chemie und Kernchemie,
Technische Hochschule Darmstadt, Darmstadt,
Federal Republic of Germany

Abstract

CALCULATION OF SPALLATION YIELDS OF PRODUCTS INDUCED BY THE INTERACTION OF HIGH-ENERGY PROTONS WITH COMPLEX NUCLEI.

A number of cross-sections for nuclides produced by the interaction of high-energy protons (300 MeV, 580 MeV, 19 GeV) with tantalum and (580 MeV) with gold has been determined. The Rudstam formula, which is not valid for elements with atomic number $Z > 50$ and proton energies > 500 MeV, has been improved. The parameter P is given as a function of E_p and A_T (mass number of the target nucleus). The parameter T (being constant in the Rudstam formula) is given as function of the mass difference between A_T and A_p (mass number of the product). It is demonstrated that the improved formula fits the experimental data over a large range of proton energies and target nuclei very well (± 20%). Comparison between the formula of Rudstam, Silberberg and Tsao, and our formula has been carried out. The improved equations are suitable for calculating yields of nuclides produced in accelerator materials. Cross-sections for products induced by cosmic rays can be calculated.

The number of investigations dealing with the interaction of complex nuclei with charged particles of middle or high energy has increased considerably during the last fifteen years. This work has confirmed the validity of Serber's two-step model [1] for proton energies up to ≈ 1 GeV. According to this model, particles are emitted in a first fast step ($t \approx 10^{-22}$ s) by direct interaction of the projectile with the individual nucleons in the target nucleus ("fast cascade"). After distribution of the deposition energy over the residual nucleus, fission and particle evaporation are in competition, thus leading to the products found experimentally. In this paper, the reaction path "fast cascade plus particle evaporation" will be characterized as "spallation".

The extensive experimental work contains a large amount of information on cross-sections, particularly with protons as projectiles. Compilations of these values have been published by Bruninx [2] and Silberberg and Tsao [3]. It turns out, however, that the present data are still incomplete because cross-sections are functions of three variables (particle energy, target nucleus, product). To obtain further unknown data by experiments is possible but rather time-consuming.

For this reason, it seemed to be reasonable to develop equations for the calculation of cross-sections. Such equations should not only be based on theoretical considerations (such as the Monte-Carlo calculations which are not in agreement with experimental results) but should also take into

account the known experimental data. Rudstam [4] has developed such an equation for the calculation of spallation cross-sections:

$$\sigma(Z,A) = \frac{144\, f_1 f_2 P A_t^{0.367}}{1 - \frac{0.3}{PA_t} - \left(0.7 - \frac{0.3}{PA_t}\, e^{-PA\ t}\right)} \exp(-P\Delta A) \exp\left(-R\left|Z - SA + TA^2\right|\right)^{3/2} \qquad (1)$$

$$= \sigma_0 \exp(-P\Delta A) \exp\left(-R\left|Z - SA + TA^2\right|\right)^{3/2}$$

where Z, A are the atomic and the mass number of the product, respectively, A_t is the mass number of the target nucleus, and $\Delta A = A_t - A$; P, R, S, T, f_1, f_2 are parameters.

A detailed explanation of the parameters is given in Ref. [4]. Equation (1) can be divided into two terms: the first term ($\sigma_0 \exp(-P\Delta A)$ indicates an exponential decrease of the yield-versus-mass curve with increasing mass difference ΔA. The second, Gaussian-like, term describes the charge dispersion on an isobar. Figure 1 shows an example for curves calculated by using the Rudstam formula [5].

The numerical values of the parameters used in Eq. (1) have been determined from experimental data by Rudstam. Most of these experiments have been carried out with targets of $A_t \lesssim 100$. In this mass range, the Rudstam formula fits the experimental results well whereas in the mass range $A_t \gtrsim 100$, particularly at high energies, the fit becomes increasingly poorer. For this reason, some authors have tried to modify the Rudstam formula in such a way that a satisfying fit is obtained also for heavier target nuclei. Such studies have been carried out by Schwarz and Oeschger [6] as well as by Silberberg and Tsao [3]. Both groups have modified the parameter P because Rudstam [4] has already indicated that the value of P might decrease with increasing A_t. Because of the lack of experimental data, Rudstam could not make more exact statements.

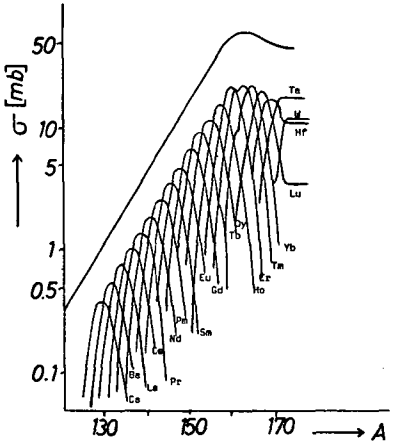

FIG. 1. Charge-dispersion curves and yield-versus-mass curve calculated for the spallation of tantalum with 660 MeV-protons using Rudstam's formula [5].

TABLE I. EQUATIONS FOR THE CALCULATIONS OF PARAMETER P

Author	Formula	Limits of validity	Remarks
RUDSTAM [4]	$P = 20 \cdot E_p^{-0,77}$	for $E_p < E_o$	E_p in MeV
	$P = 0.056$	for $E_p \geqslant E_o$	
SCHWARZ and OESCHGER [6]	$P = 0.237 \cdot E_p^{-0.667} \cdot \frac{1}{1+0.02 \cdot A_t}$	for $E_p < E_o$	E_p in GeV
	$P = \text{const.}$	for $E_p \geqslant E_o$	
SILBERBERG and TSAO [3]	$P = 20 \cdot E_p^{-0.77} \cdot C_p$	for $E_p < E_o$	E_p in MeV
	$C_p = 1-1.5 \cdot 10^{-5}(A_t-100) \frac{E_o+150}{E_p+150}$		
	$E_o = 69 \cdot A_t^{0.867}$		
	$P = 0.77 \cdot A_t^{-2/3}$	for $E_p \geqslant E_o$	

TABLE I (cont.)

Author	Formula	Limits of Validity	Remarks
ROSS and BÄCHMANN	$P = 20 \cdot E_p^{-0.77} \cdot C_p$	for $E_p < E_o$	E_p in MeV
	$C_p = 1 - 1.5 \cdot 10^{-5} (A_t - 100)^x \frac{E_o + 150}{E_p + 150}$		
	$x = \frac{1 + \frac{E_o}{10^4}}{1 + \frac{E_o}{E_p^2}}$		
	$E_o = 69 \cdot A_t^{0.867}$	for $E_p \geqslant E_o$	
	$P = 0.77 - A_t^{-2/3}$		

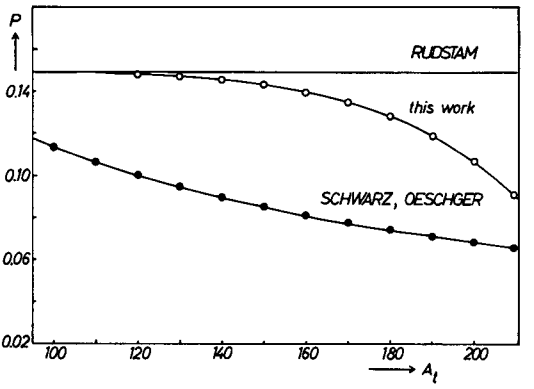

FIG. 2. Parameter P versus target mass number at E_p = 0.58 GeV, calculated with different formulas.

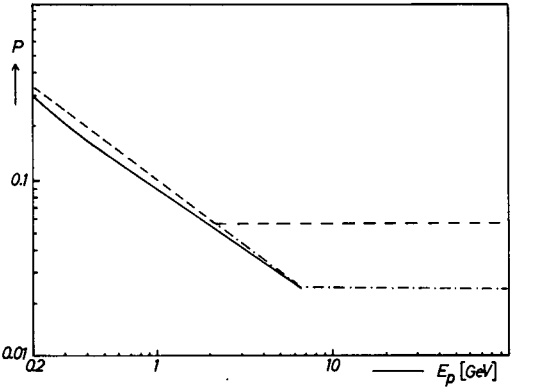

FIG. 3. Parameter P versus proton energy E_p for tantalum as target, calculated with different formulas. – – – – Rudstam [4], –·–·– Silberberg and Tsao [3], ——— this work.

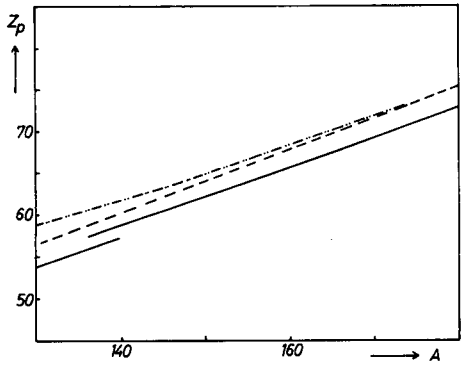

FIG. 4. Calculated maximum of the charge-dispersion curve Z_p versus product mass number (tantalum target). ——— beta-stability line taken from Coryell [17], – – – Z_p-curve calculated according to Rudstam [4], –··–·· Z_p-curve calculated in this work.

TABLE II. EQUATIONS FOR THE CALCULATION OF THE PARAMETER T

Author	Equation
RUDSTAM [4]	$T = 0.00038 \pm 0.00002$
SILBERBERG and TSAO [3]	$T = 0.00038 \pm 0.00002$
ROSS and BÄCHMANN	$T = 0.00038 - 6.087 \cdot 10^{-10} \cdot (\Delta A)^{3.115}$

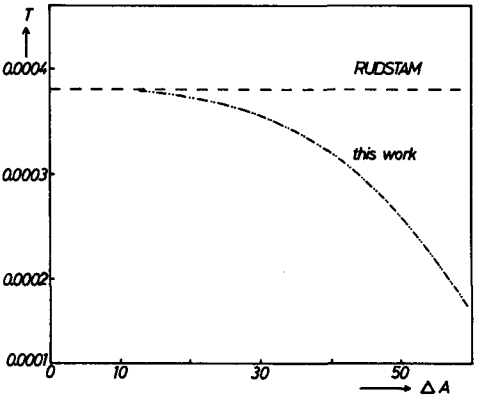

FIG. 5. Parameter T versus mass difference $\Delta A(\Delta A = A_t - A)$.

A comparison of the calculated cross-sections — using the formulas for P given by the authors cited above — with experimental data reported by Neidhart and Bächmann [7, 8] (Ta + p; E_p = 0.58; 19 GeV) and Grover [9] (Ta + p; E_p = 5.7 GeV), respectively, gives rather poor agreement. At 0.58 GeV, the slope of the yield-versus-mass curve calculated according to Rudstam and Silberberg, respectively, is too steep, compared with the experiment whereas the slope calculated according to Schwarz is too flat. At the higher proton energies, the Silberberg formula fits the experimental slope correctly while Rudstam's equation gives values too high.

Our formula for P is based on the assumption that at high proton energies (> 1 GeV) the Silberberg formula is correct while at lower energies the value of P has to be reduced compared with Rudstam and Silberberg,

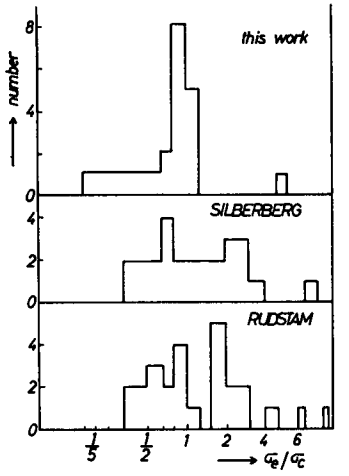

FIG. 6. Histogram of the ratios of experimental and calculated cross-sections. Experimental values taken from Ref. [7] (E_p = 0.58 GeV; Ta).

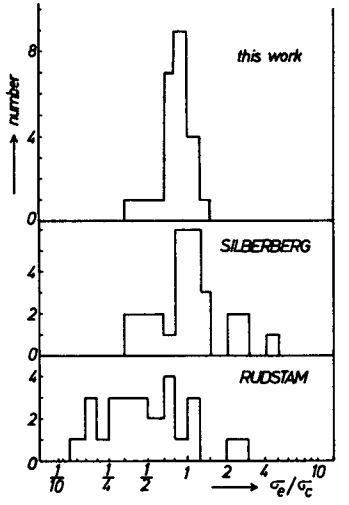

FIG. 7. Histogram of the ratios of experimental and calculated cross-sections. Experimental values taken from Ref. [9] (E_p = 5.7 GeV; Ta).

respectively. Table I contains the formulations made for P by several authors. In all cases, except Rudstam, P is given as a function of the proton energy E_p and the target mass number A_t. E_p is kept constant; for different target mass numbers the curves shown in Fig. 2 are obtained. The curve which has been calculated according to our formula decreases steeply with increasing values of A_t. The shape of the curve is confirmed by the results of Schramm [10] who has reported P-values of about 0.02 in

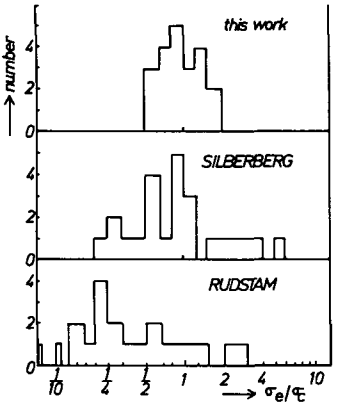

FIG. 8. Histogram of the ratios of experimental and calculated cross-sections. Experimental values taken from Ref. [8] (E_p = 19 GeV; Ta).

the case of $A_t \geq 232$. The formula of Schwarz and Oeschger seems to be useful at low values of A_t ($Z_t \leq Z_{Cu}$) [6] but not for heavier targets, despite a rather good correspondence with experimental values at very high target mass numbers. Figure 3 shows P as a function of E_p for tantalum as target nuclide. The differences between the different formulas are mainly based on the values of E_0, the proton energy above which P is constant.

A comparison between experimental and calculated cross-sections showed that satisfying agreement cannot be obtained by modifying P alone. The second, Gaussian-like term of Eq. (1) has to be corrected, too, e. g. by variation of the parameter T. So far, all authors have used a constant value of T. In this case, the curve connecting the peak of the charge-dispersion curve in different isobars has a more or less constant distance to the beta-stability line (Fig. 4). The modification of the formula has been carried out using cross-sections reported by Neidhart and Bächmann [7]. In contrast to the other authors [3, 4], the value of T depends on the mass difference ΔA (see Table II). Therefore, the distance between the beta-stability line and the maximum of the charge-dispersion curve grows with increasing ΔA (see Fig. 4). In Fig. 5, T has been plotted versus ΔA.

To check the quality of the fit, the ratios between experimental and calculated cross-sections were determined. The histograms (Figs 6 to 8) show the distribution of these ratios around the optimum value of unity. These histograms make evident that the best fit is obtained with our formula. The average ratio between experimental and calculated cross-sections and its standard deviation, calculated for each experiment, confirms this result as can be seen from Table III. This improvement is mainly based on the new formula for the parameter T.

We have tried to make an extension of our formula to describe the maximum of the excitation function (^{149}Tb from gold) at $E_p \approx 2$ GeV which has been reported by several authors [11-14]. So far, these attempts have not been very successful. It seems to be necessary to introduce more parameters. The equation for P, however, seems to be correct because the slope of the experimental and of the calculated yield-versus-mass

TABLE III. AVERAGE RATIO OF EXPERIMENTAL AND CALCULATED CROSS-SECTIONS FOR THE REACTION OF TANTALUM WITH PROTONS OF DIFFERENT ENERGIES

Proton energy (GeV): 0.58 [7]

Formula of	Average ratio	Standard deviation (absol. scale)	Standard deviation (%)
RUDSTAM	2.054	2.675	130
SILBERBERG	2.021	2.372	117
this work	1.070	0.649	61

Proton energy (GeV): 5,7 [9]

Formula of	Average ratio	Standard deviation (absol. scale)	Standard deviation (%)
RUDSTAM	0.615	0.559	91
SILBERBERG	1.228	0.940	77
this work	0.891	0,225	25

Proton energy (GeV): 19 [8]

Formula of	Average ratio	Standard deviation (absol. scale)	Standard deviation (%)
RUDSTAM	0.538	0.642	119
SILBERBERG	1.220	1.195	98
this work	1.029	0.322	31

curve is the same [15]. It is thought that the decrease of T with increasing ΔA (Fig. 5) is too steep, thus leading to a shift of the calculated charge-dispersion curves into the region of neutron-deficient nuclides at high values of ΔA. This effect may originate in the use of experimental cross-sections [7] of nuclides adjacent to magic numbers. Silberberg and Tsao [3] have reported that the charge-dispersion curves of isobars adjacent to magic numbers are shifted towards the next magic number. So far, this effect could not be calculated because of the lack of experimental data and thus has not been taken into account in the formulation of our equation for T.

A basic problem of these semi-empirical equations is the reliability of the experimental cross-sections. It has been found [16] that the same cross-sections determined simultaneously in different laboratories showed differences of about 50%. The sources of error are experimental uncertainties such as activity measurements and the chemical analyses, as well as uncertainties concerning nuclear data such as abundances of radiation, cross-sections of monitor reactions, etc. It is therefore extremely important to examine the values used in literature for the determination of cross-sections and, perhaps, to re-calculate the reported cross-sections. Unfortunately, in many cases it is not indicated which nuclear data have been used.

From the above discussion, it turns out that it is possible, by the aid of our formula, to calculate spallation cross-sections in the mass range $A_t \geq 209$ with an accuracy of 25-30%. A further improvement seems to be possible by using more experimental data for the calculation.

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DISCUSSION

A.T.G. FERGUSON: Did this work have any motivation other than scientific interest in interpolating between existing cross-section data?

E. ROSS: I see applications at present in the calculation of the activities which are formed in high-energy particle accelerators by nuclear reactions between the projectiles and the accelerator materials (shielding, beam tubes, etc.). Reference may be made in this connection to paper IAEA-SM-170/35, which was presented by Dr. Thorson. Another field of application is problems connected with the space sciences, for example, the activation of spacecraft by high-energy protons and similar particles which are present in space.

Z. SUJKOWSKI: Can you get the yields of light elements produced by spallation or fragmentation of heavy targets?

E. ROSS: This is not possible.

Z. SUJKOWSKI: Do you include spallation-fission competition in your treatment or do you ignore the fission altogether?

E. ROSS: In principle, we can separate a spallation and a fission contribution to the cross-sections observed experimentally. The problem is, however, that fission does not play an important role when targets such as gold or tantalum are used and since the estimations have an error of 20 - 30%, the fission contribution lies within these limits.

A SURVEY OF NUCLEAR DATA USE IN APPLIED FIELDS*

M. LEDERER, J. M. HOLLANDER
Lawrence Berkeley Laboratory,
University of California,
Berkeley, Calif., United States of America

Abstract

A SURVEY OF NUCLEAR DATA USE IN APPLIED FIELDS.

In connection with compilation activities for the Table of Isotopes, we are conducting a survey of nuclear data needs outside the field of basic nuclear science, principally among users of radioisotopes. Questionnaires are being mailed to people in such diverse fields as medicine, environmental studies, and oceanography. The questions are intended to ascertain what sources of data are used, what specific types of data are required, to what extent existing compilations fill these needs, and what changes are needed or desired. Several questions at the end of the list concern specifically the use of the Table of Isotopes. A preliminary summary of the response to date to the survey is discussed.

To plan compilation work for the Table of Isotopes and to co-ordinate our activities with those of other groups, we are undertaking a survey of the requirements for nuclear data of data users outside the field of basic nuclear physics. Since the spirit of our survey is very close to the purpose of this entire symposium, I hope our presumption in reporting on our modest effort will be excused. It is certainly not our intent to preempt the work of the symposium! We should simply like to describe the questionnaire that we are distributing, and to provide some information on the preliminary response we have received.

Figure 1 shows the actual questionnaire. Questions 1 - 3 cover general data on the respondent's type of work and his reasons for using nuclear data. Question 4 defines the specific data that he uses. Question 5 concerns the user's preference for best values or measured data, and the number of such values he wants to see. Question 6 deals with the frequency of use of different data sources. Questions 7 and 8 are an attempt to determine the extent to which the user is satisfied with these sources, and the reasons for his dissatisfaction, if any. Question 9 is intended to enlarge on the answers to the previous questions, and to explore the possibility of additional interest in non-printed forms of the data – magnetic tapes or direct, on-line retrieval. In addition to these questions, we have enclosed a sample format for the 7th edition of the Table of Isotopes and invited the respondent to comment on it.

We have at this time a partial response to the first mailing of the questionnaire, and we wish to report on it here. Part of the purpose of the first mailing was to determine whether the questions are adequate to obtain the desired information, so I shall comment on the problems we have encountered in interpreting the answers as I describe the results. For any

* Work performed under the auspices of the US Atomic Energy Commission and the US National Bureau of Standards.

statisticians I should set forth my qualifications in this field: I have read the book How to Lie with Statistics, by Huff and Geis [1].

The preliminary mailing was to 66 members of the Society of Nuclear Medicine. The names were selected from the entire membership list by a manufacturer and distributor of radioisotopes, and are presumed to represent an active and sophisticated group of data users in the medical field.

To date, we have received 34 responses. A majority (23) of the respondents work in universities. All of them use radioisotopes, and a large number also use accelerators (17) and reactors (12). They are heavy users of nuclear data: 24 claim to use data frequently, 10 occasionally.

The range of types of data required was broader than we had anticipated, even for a select group of medical users. Figure 2 summarizes the responses to question 4. It is noteworthy that a significant number of respondents expressed a need for such specialized data as detailed neutron and charged-particle cross-sections.

The response to question 5 (Fig.3) was mixed. Since a number of people indicated a need for both the best reported values and an adopted best value, while some specifically indicated that either was satisfactory, we have tabulated these responses separately from the request for only one of these. Obviously, the question needs to be stated more precisely. The present results do, however, indicate a divided opinion about the desirability of giving best values versus measured values.

The sources of data that the respondents claim to use are summarized in Fig.4. It is noteworthy that five people indicated frequent use of the MIRD (Medical Internal Radiation Dose) pamphlets.

The response to questions 7 and 8 was most illuminating, and also most difficult to interpret. Most of the respondents (26) indicated that the data fulfil their needs "quite well", 2 people said "completely", and 6 indicated "only partially". More important, the reasons for dissatisfaction indicate a general malaise concerning the difficulty of interpreting the available data, often on the part of people who also indicated a need for very specialized data, and who even claimed that the existing compilations did not cover enough categories of data.

In some such cases, there appears to be a need for more highly reduced or evaluated data, such as electron energies and absolute intensities; this need is also reflected in a frequent use of the MIRD pamphlets.

One further comment concerning the degree of satisfaction with and complaints about the data: a number of respondents appeared to direct their responses to these questions specifically at the Table of Isotopes, rather than at all the data compilations they use, which was not the intent of the question. This question will be re-phrased for our next mailing.

Aside from cold statistics, the responses contained some intelligent comments, and a few that were highly enlightening. In such cases, a follow-up by direct contact with the respondent should be profitable. We were pleased with the response, and believe that the results of the survey will be quite useful. We plan to continue the survey, and we shall make a more extensive report on responses to it at a later date.

1. Personal Data

Name:_____(optional)

Occupation:_____

Name of Employer:_____(optional)

Employer is a : ☐ Private Corporation
☐ Government operated installation
☐ Government contract installation
☐ University_____(name of dept. or college)
☐ Other_____

2. (a) Do you use or encounter radioisotopes, nuclear reactors, or charged-particle accelerators, or deal with nuclear properties in your work?

☐ radioisotopes ☐ reactors
☐ accelerators ☐ nuclear properties

(b) For what purpose? (Type of application, e.g.: tracers in chemical studies, medical diagnostics, abrasion studies, monitoring radioactive pollution, applications in cosmology, etc.)

3. How often do you use sources of nuclear data in your work (compilations, wall charts, original publications, journals, etc.)?

☐ Frequently ☐ Occasionally ☐ Rarely ☐ Never

(IF THE ANSWER TO QUESTION 3 IS "NEVER", IT IS NOT NECESSARY TO ANSWER THE FOLLOWING QUESTIONS.)

4. What types of data do you require? (check any which you use).

Type of data

☐ (a) Half-lives of radioactive substances

☐ (b) Isotopic abundances

☐ (c) Nuclear masses

☐ (d) Nuclear spins and moments

☐ (e) Neutron capture or fission cross-sections

☐ (f) Nuclear decay modes and genetic (parent-daughter) relationships

☐ (g) Means of producing radioisotopes

(h) Energies and intensities of radiations:

☐ gamma rays

☐ X-rays

☐ beta particles

☐ conversion electrons

☐ alpha particles

☐ other radiations (specify)

_____

_____

_____

_____

FIG. 1. Survey Questionnaire, as sent in first mailing. One additional page (not shown) requested comments on the proposed content and format of the 7th edition of the Table of Isotopes.

☐ (i) Detailed neutron reaction data (energy-dependent cross sections, resonance parameters)

☐ (j) Neutron capture gamma spectra

(k) Charged-particle or photo-induced reaction data:

- ☐ Gross cross sections (Excitation functions)
- ☐ Cross-sections for specific levels
- ☐ Angular distributions
- ☐ Gamma-ray spectra

☐ (l) Nuclear level schemes

☐ (m) Other types of data (specify)

_____

_____

_____

5. How complete a listing of a given quantity do you require? (Check one or more.)

- ☐ All reported values
- ☐ A few of the best reported values
- ☐ The one most precise or accurate reported value
- ☐ A weighted average or adopted "best" value
- ☐ Other (specify)

_____

_____

_____

6. What sources of nuclear data do you consult and how frequently do you use them? (Check appropriate boxes for any sources that you use.)

Compilation	Amount of Use: Frequent	Occasional	Rare
(a) Table of Isotopes, 6th edition (1967)	☐	☐	☐
(b) Table of Isotopes, earlier editions	☐	☐	☐
(c) Nuclear Data Sheets (the journal Nuclear Data B and earlier loose-leaf sheets)	☐	☐	☐
(d) Nuclide "wall" charts (GE, Batelle N.W., or other versions)	☐	☐	☐
(e) BNL-325 (the "Barn Book")	☐	☐	☐
(f) "Light-element" compilations (by F. Ajzenberg-Selove or Endt and van der Leun, published in the journal Nuclear Physics, or other (specify)	☐	☐	☐
(g) Any "specialized" tables of specific properties, such as nuclear moments, nuclear masses, fundamental constants, electron binding energies, etc. (specify)			
_____	☐	☐	☐
_____	☐	☐	☐

FIG. 1 (continued)

_____	☐	☐	☐
_____	☐	☐	☐
(h) Data supplied by manufacturer or distributor of radioisotopes.	☐	☐	☐
(i) Other (specify)			
_____	☐	☐	☐
_____	☐	☐	☐
_____	☐	☐	☐
_____	☐	☐	☐

7. How well do the sources of data you use fulfill your requirements?

☐ Completely ☐ Quite well ☐ Only partially ☐ Poorly

8. In what ways do you find them inadequate? (Check any answers which are applicable.)

☐ Do not cover some of the required categories of data
☐ Cover too many categories of data
☐ Not enough detail
☐ Too much detail
☐ Too difficult to interpret
☐ Too far out of date
☐ Other (specify)

_____

_____

9. Would you like to have the data in different forms, such as:

☐ Another type of printed compilation (specify)

_____

☐ A magnetic tape

☐ Retrievable on-line at a computer terminal (such as a teletype connected to a computer data bank via telephone lines)?

☐ Other (specify)

_____

_____

FIG. 1 (continued)

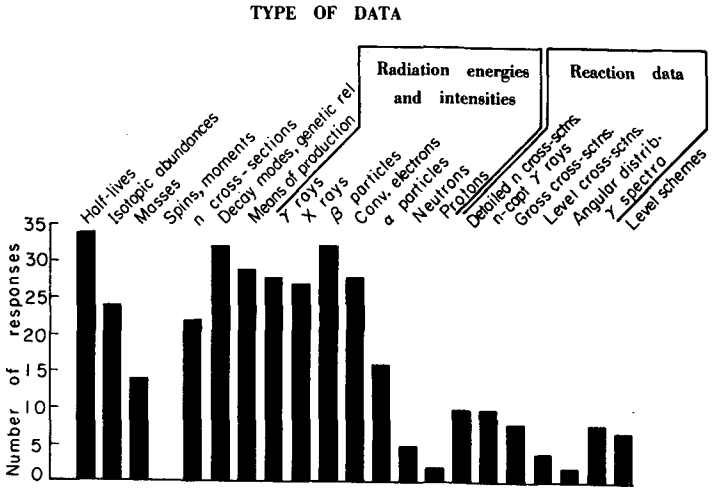

FIG. 2 Responses to question 4 ("What type of data do you require?").

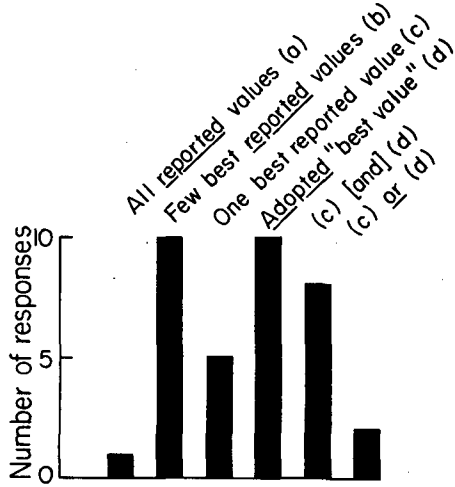

FIG. 3 Responses to question 5 ("How complete a listing of a given quantity do you require?"). Respondents who checked both "The one most precise..... reported value" and "A weighted average or adopted best value" are represented by the next to last bar; respondents who inserted "or" between these two choices are represented by the last bar.

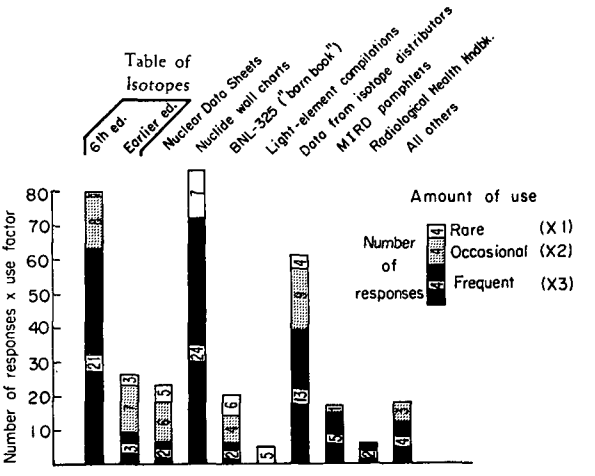

FIG. 4 Responses to question 8 ("What source of nuclear data do you consult.....?"). Actual numbers of responses in each of the usage categories "frequent", "occasional", and "rare" are given in the bars. The height of the bar segments are proportional to 3, 2, and 1 times the number of responses for frequent, occasional, or rare use, respectively. Write-in responses are included, except for a few data sources cited only once.

REFERENCES

[1] HUFF, D., GEIS, I., How to Lie with Statistics, W.W. Norton and Co., New York (1954).

DISCUSSION

N.M. SPYROU: If your survey eventually shows that the majority of users require specific and separate nuclear data, would you consider preparing separate "cookbooks"?

M. LEDERER: We would consider doing whatever was wanted! However, the impression I get from this Symposium is that there are already many excellent "cookbook" compilations (in existence or being written) and that what is most needed for these is good basic data evaluations.

H. MÜNZEL: We have made a similar survey of the need for nuclear data in activation analysis. The results of this survey are available as report from the Nuclear Research Centre at Karlsruhe.

J.J. SCHMIDT: When extending your very useful enquiry do you intend to "internationalize" it, i.e. to call on people outside the United States as well?

M. LEDERER: Yes, we do. The difficulty is, of course, in obtaining satisfactory mailing lists. Our next mailing will be to authors of papers listed in Nuclear Science Abstracts, and should include some non-U.S. researchers.

I.A. KONDUROV: Could you give us some statistics concerning replies to the 9th question?

M. LEDERER: There was very little response. One person indicated that he already had data (neutron cross-sections) on tape. I believe one or

two people expressed interest in tape and/or on-line retrieval. There were scattered requests for different types of compilations, most of which weren't very enlightening.

A.T.G. FERGUSON: Dr. Rose and I have made a pilot study of needs for nuclear data in a number of fields by interviewing in some depth selected groups of users. One significant point that emerged was that the majority of users were unaware of the wide range of data compilations available. They were very happy to receive the preliminary list of Compilations prepared at the first meeting of the IWGNSRD-INDC(NDS)-46/U+W).

D.J. HOREN: I would agree with Dr. Ferguson's comment and I think some of the comments made earlier at this Symposium seem to confirm this, for instance, as regards knowledge of the Martin and Blichert-Toft compilation.

Section XV

ACTIVATION ANALYSIS: CHARGED PARTICLES AND PHOTONS

Chairman

H. MÜNZEL (Federal Republic of Germany)

IMPORTANCE ET ROLE DES CONSTANTES NUCLEAIRES EN ANALYSE PAR ACTIVATION AUX PARTICULES CHARGEES OU PHOTONS γ

Ch. ENGELMANN
CEA, Centre d'études nucléaires
de Saclay, France

Abstract–Résumé

IMPORTANCE AND ROLE OF NUCLEAR DATA IN ACTIVATION ANALYSIS USING CHARGED PARTICLES OR PHOTONS.

The importance and the role of nuclear data in activation analysis using charged particles or photons are discussed; after a general survey of activation analysis, different standardization methods are presented, the influence of competitive nuclear reactions is discussed and the importance of compilations of certain nuclear data and of special compilations directly aimed at activation analysis is shown.

IMPORTANCE ET ROLE DES CONSTANTES NUCLEAIRES EN ANALYSE PAR ACTIVATION AUX PARTICULES CHARGEES OU PHOTONS γ.

L'auteur discute l'importance et le rôle des constantes nucléaires en analyse par activation aux particules chargées ou photons γ; après s'être occupé des généralités sur l'analyse par activation, il présente les différentes méthodes d'étalonnage, discute l'influence des réactions nucléaires compétitives, et montre l'intérêt qu'ont les compilations de certaines constantes nucléaires et les compilations particulières destinées directement à l'analyse par activation.

INTRODUCTION

L'utilisation des particules chargées et des photons γ en analyse par activation s'est avant tout imposée à cause des possibilités remarquables qu'ils offrent pour déterminer les éléments légers, notamment le bore, le carbone, l'azote, l'oxygène et le fluor.

Ces moyens d'irradiation conduisent, pour ces éléments, à des limites de détection situées entre 10^{-2} et 10^{-4} $\mu g \cdot g^{-1}$.

Les sections efficaces des réactions nucléaires par lesquelles on les dose sont de quelques dizaines de millibarns pour les photons γ. Elles sont 10 à 100 fois plus élevées avec les particules chargées.

Ce sont donc ces dernières qui permettent de réaliser les analyses les plus sensibles. Mais leur emploi est beaucoup plus délicat, de sorte qu'en définitive, on préfère toujours se servir d'abord des photons γ et n'utiliser les particules chargées que là où ils ne peuvent être mis en œuvre, en particulier lorsque la sensibilité est insuffisante.

Ainsi, ces deux moyens d'irradiation sont complémentaires et s'avèrent aussi indispensables l'un que l'autre.

Déjà maintenant, les degrés de pureté de nombreux matériaux sont tels que seules ces nouvelles méthodes analytiques rendent possible la détermination exacte de certains éléments légers.

Par exemple, on sait élaborer des métaux alcalins, du sodium notamment dans lequel les concentrations en carbone et oxygène sont inférieures à 1 $\mu g \cdot g^{-1}$. L'activation par photons γ est le procédé unique qui permette de réaliser une telle analyse. De la même façon l'azote,

qui semble une impureté très importante dans les métaux réfractaires, se trouve dans ces derniers à des teneurs de l'ordre de 1 $\mu g \cdot g^{-1}$. Là encore les photons γ sont sans concurrence.

On pourrait citer de nombreux autres exemples qui, comme les précédents, mettent en évidence les immenses possibilités de l'analyse par activation aux particules chargées ou photons γ pour résoudre les problèmes analytiques particulièrement délicats voire impossibles avec les méthodes conventionnelles.

L'apparition d'accélérateurs compacts, beaucoup moins coûteux que les grandes machines construites pour des expériences de physique nucléaire, notamment de cyclotrons pour les particules chargées et de microtrons pour la production de flux très importants de photons γ de freinage, entraînera certainement une très large diffusion de ces techniques dans les prochaines années.

Il semble donc souhaitable que des recueils contenant toutes les données nucléaires et d'autres informations intéressant directement l'analyse par activation aux particules chargées et photons γ soient constitués, afin que les utilisateurs actuels et futurs de ces méthodes puissent, d'une manière simple et sans ambiguïté, d'une part savoir quelles sont les réactions nucléaires les plus spécifiques pour doser tel élément et l'énergie que les projectiles incidents doivent posséder, et, d'autre part, connaître les sensibilités de détection possibles ainsi que l'importance relative des interférences éventuelles, dues à des réactions nucléaires compétitives sur d'autres éléments contenus dans l'échantillon irradié.

1. GENERALITES SUR L'ANALYSE PAR ACTIVATION AUX PARTICULES CHARGEES ET PHOTONS γ

1.1. Analyse par activation aux photons γ

La source la plus intense de photons γ de grande énergie que l'on sache produire est constituée par le rayonnement de freinage d'électrons accélérés bombardant une cible métallique de numéro atomique élevé, tungstène, platine, or .... Le spectre est continu, l'énergie maximale est égale à l'énergie cinétique des électrons incidents. Le faisceau de photons γ est très directif. Les activations varient d'une façon relativement importante dès que l'on s'éloigne de la cible [1]. La position des échantillons pendant l'irradiation derrière celle-ci doit être précise et reproductible. Ils sont amenés par l'intermédiaire d'un circuit pneumatique dans des cartouches qui sont envoyées depuis le laboratoire.

La relation de base de l'analyse par activation aux photons γ est la suivante:

$$A_s = \int_{E_s}^{E_{max}} dE \int_0^{\Omega} \phi \; n \; N \; \sigma_{rad} \; \sigma_{(\gamma,x)} \; \Psi_{\Omega} \; d\Omega \qquad (1)$$

A_s = nombre de noyaux atomiques subissant par unité de temps la réaction d'activation (γ, x), encore égal à l'activité à saturation du radioélément produit

E_{max} = énergie cinétique des électrons incidents
E_s = énergie seuil de la réaction nucléaire d'activation (γ, x)
Ω = angle solide sous-tendu par l'échantillon
ϕ = flux du faisceau d'électrons
N = nombre de noyaux par cm^2 de la cible de conversion
n = nombre de noyaux par cm^2 de l'élément activé par réaction (γ, x)
σ_{rad} = section efficace de production du rayonnement de freinage
$\sigma_{(\gamma,x)}$ = section efficace de la réaction nucléaire d'activation
Ψ_Ω = fonction de distribution angulaire du rayonnement de freinage, dans laquelle il est tenu compte de l'absorption des photons γ à la fois dans la cible et dans l'échantillon.

Supposons que pendant l'irradiation ϕ soit invariable. N est une constante de la cible. D'autre part, si l'élément activé est distribué d'une façon homogène dans l'échantillon irradié, on pourra considérer n comme constant également. De sorte que l'expression précédente devient

$$A_s = \phi N n \int_{E_s}^{E_{max}} dE \int_0^{\Omega} \sigma_{rad}\ \sigma_{(\gamma,x)}\ \Psi_\Omega\ d\Omega \qquad (2)$$

Même si l'on peut exprimer les différents termes entrant sous le signe somme de cette relation sous une forme analytique, il faut reconnaître que le calcul de A_s est complexe.

Posons

$$k_E = N \int_{E_s}^{E_{max}} dE \int_0^{\Omega} \sigma_{rad}\ \sigma_{(\gamma,x)}\ \Psi_\Omega\ d\Omega \qquad (3)$$

Si l'on pouvait déterminer ces coefficients k_E pour une réaction nucléaire donnée, en fonction de l'énergie maximale des électrons E_{max}, on aurait une formule facilement utilisable du type

$$A_s = k_E \phi n \qquad (4)$$

qui permettrait de prévoir les performances de la méthode en fonction de l'énergie des électrons incidents.

Pour rendre l'emploi de la relation (4) plus général on pourrait convenir d'un arrangement géométrique cible-échantillon donné et fixer les dimensions de ces derniers à des valeurs standards telles que l'on puisse, quel que soit le matériau irradié, négliger l'influence de l'absorption des photons γ.

Si de telles tables des coefficients d'étalonnage k_E existaient, il suffirait d'irradier un échantillon, de mesurer l'intensité du faisceau d'électrons ϕ d'une part, l'activité induite d'autre part, pour pouvoir déterminer immédiatement la concentration n.

1.2. Analyse par activation aux particules chargées

Les particules chargées subissant un freinage très important en pénétrant dans les milieux matériels, l'échantillon irradié n'est pas activé uniformément. En particulier, si son épaisseur est supérieure au parcours total, la zone effectivement activée est limitée, d'une part par le diamètre du faisceau, et d'autre part par le parcours efficace le long duquel l'énergie des particules incidentes est supérieure au seuil E_s de la réaction nucléaire d'activation considérée.

Dans un échantillon, l'énergie des particules chargées dans les tranches successives est fonction de l'épaisseur traversée x. A cette dernière correspond donc une valeur parfaitement définie de la section efficace de la réaction nucléaire utilisée, soit σ_x.

Ainsi, l'activation dans les tranches successives d'un échantillon épais est en corrélation directe avec la fonction d'excitation σ_E.

Il est donc indispensable, sous peine de commettre des erreurs analytiques non négligeables, que l'élément à doser soit homogènement réparti dans l'échantillon irradié, à moins que l'épaisseur de ce dernier soit faible et telle que la section efficace dans le domaine d'énergie correspondant soit constante. Dans ce cas, en effet, la relation donnant l'activité à saturation est particulièrement simple:

$$A_s = \phi \, n \, \sigma_0 \, x \tag{5}$$

A_s = activité à saturation en désintégrations par seconde
ϕ = nombre de particules chargées par seconde atteignant l'échantillon
σ_0 = section efficace de la réaction nucléaire utilisée, pour l'énergie incidente E_0
n = nombre d'atomes de l'élément à doser par gramme d'échantillon
x = épaisseur de l'échantillon en $g \cdot cm^{-2}$.

Dans le cas général, le calcul de l'activité induite est plus compliqué. La relation de base est la suivante:

$$A_s = \int_0^R \phi \, n \, \sigma_x \, dx \tag{6}$$

R = parcours total des particules chargées dans le matériau irradié
σ_x = section efficace de la réaction nucléaire d'activation en fonction de la profondeur de pénétration x.

On suppose naturellement que l'épaisseur de l'échantillon est supérieure au parcours efficace.

La diminution de ϕ dans la zone activée étant le plus souvent négligeable, on peut considérer ce terme comme invariable.

Si de plus, l'élément à doser est réparti d'une façon homogène dans l'échantillon, n est constant, et la relation (6) devient:

$$A_s = \phi \, n \int_0^R \sigma_x \, dx \tag{7}$$

Par un changement de variable, cette expression pèut se mettre sous une autre forme:

$$A_s = \phi\, n \int_{E_0}^{0} \sigma_E \left(\frac{dE}{dx}\right)_E^{-1} dE \qquad (8)$$

E_0 = énergie incidente des particules chargées
σ_E = fonction d'excitation de la réaction nucléaire
$(dE/dx)_E$ = pouvoir de ralentissement du milieu constituant l'échantillon irradié, pour les particules chargées utilisées.

2. LES DIFFERENTES METHODES D'ETALONNAGE

2.1. Cas de l'activation par photons γ

D'après ce que l'on a vu plus haut, l'idéal serait de disposer d'une table donnant les coefficients d'étalonnage k_E pour chaque réaction nucléaire présentant un intérêt sur le plan analytique en fonction de l'énergie du faisceau d'électrons E_{max}. En mesurant avec précision l'intensité de ce dernier, ce qui semble possible, on pourrait calculer la concentration n en se servant de la relation (4).

Une telle table n'existant pas, on procède de la façon suivante: avec l'échantillon à analyser on irradie simultanément un étalon de même forme géométrique, dans lequel l'élément à doser se trouve à une teneur parfaitement connue.

C'est une méthode comparative qui donne d'excellents résultats. Cet étalon primaire peut à la rigueur être remplacé par un moniteur de flux constitué d'un autre élément que celui à doser, éventuellement la matrice elle-même; on utilise alors la technique de l'étalonnage interne.

Il est fort probable que, pour effectuer des dosages précis, ces modes opératoires seront toujours préférés à celui reposant sur l'utilisation de coefficients k_E prédéterminés. Le calcul exact de ces derniers semble en effet délicat. En revanche, leur connaissance, même approchée, présenterait un intérêt évident pour évaluer a priori les performances de l'activation par photons γ.

Il faut d'ailleurs signaler que l'on peut trouver dans la littérature des courbes d'activation déterminées expérimentalement qui représentent la variation de l'activité induite (qui est directement proportionnelle à k_E) dans une masse unitaire de l'élément considéré par la réaction nucléaire qui l'active, en fonction de l'énergie et pour une intensité de faisceau donnée [2-4].

2.2. Cas de l'activation par particules chargées

On distingue essentiellement les cinq méthodes suivantes:
- méthode absolue
- méthode de la section efficace moyenne
- méthode de l'épaisseur équivalente
- méthode comparative
- méthode de l'étalon interne.

La première repose sur l'utilisation d'une des relations (5) à (8), suivant les cas. Sa mise en œuvre nécessite donc la connaissance précise de la fonction d'excitation de la réaction nucléaire utilisée et la possibilité de pouvoir mesurer avec exactitude l'intensité du faisceau.

La seconde fait intervenir la notion de section efficace moyenne

$$\bar{\sigma} = \frac{\int_0^R \sigma_x \, dx}{\int_0^R dx} \tag{9}$$

Soit encore, en changeant de variable

$$\bar{\sigma} = \frac{\int_{E_0}^0 \sigma_E \left(\frac{dE}{dx}\right)_E^{-1} dE}{\int_{E_0}^0 \left(\frac{dE}{dx}\right)_E^{-1} dE} \tag{10}$$

Elle a été proposée par Ricci et Hahn en 1965 [5]. Ces auteurs ont démontré que cette section efficace moyenne est pratiquement indépendante du milieu dans lequel se trouve l'élément activé.

Ils ont en effet établi la relation

$$\bar{\sigma} = \frac{\int_{E_0}^0 \sigma_E \, E \, dE}{\int_{E_0}^0 E \, dE} \tag{11}$$

qui met en évidence que la section efficace moyenne ne dépend que de la fonction d'excitation σ_E et de l'énergie E_0 des particules chargées utilisées.

Il est donc possible d'établir des tables donnant en fonction de l'énergie E_0 les valeurs de la section efficace moyenne. Cela pour toutes les réactions nucléaires utilisables en analyse par activation.

Connaissant l'activité à saturation, l'intensité du faisceau et le parcours total R, il sera facile de calculer la concentration à partir de l'expression

$$A_s = \phi \, n \, \bar{\sigma} \, R \tag{12}$$

L'intérêt d'une compilation des sections efficaces moyennes en fonction de l'énergie des réactions nucléaires potentiellement utilisables pour l'analyse par activation est évident. Elle est indispensable pour calculer les teneurs si l'on a recours à ce procédé d'étalonnage, mais surtout, elle permettrait de prévoir les sensibilités de détection, donc les performances intrinsèques des diverses réactions nucléaires en fonction de l'énergie. Ceci permet notamment de définir a priori les conditions optimales d'activation, en particulier la réaction nucléaire à

utiliser et l'énergie des particules incidentes pour effectuer une analyse donnée.

La méthode de l'épaisseur équivalente est antérieure à la précédente. Elle a été proposée par l'auteur en 1964 [6]. C'est une méthode entièrement expérimentale.

L'épaisseur équivalente est définie comme étant la profondeur e d'une zone fictive dans laquelle l'activation de l'élément considéré, supposé distribué d'une manière homogène, serait constante.

En d'autres termes, on aurait

$$A_s = \phi \, n \, \sigma_0 \, e \tag{13}$$

En tenant compte de la relation (7) on a donc par définition

$$e = \frac{\int_0^R \sigma_x \, dx}{\sigma_0} \tag{14}$$

En plaçant devant l'échantillon un étalon très mince d'épaisseur Δx telle que l'on peut y considérer la section efficace comme pratiquement constante et égale à σ_0, on a

$$A_s' = \phi \, n' \sigma_0 \, \Delta x \tag{15}$$

n' étant la concentration de l'élément à doser dans l'étalon mince. A partir des relations (13) et (15) on établit la formule suivante:

$$n = \frac{A_s}{A_s'} \cdot \frac{\Delta x}{e} \cdot n' \tag{16}$$

Pour mettre en œuvre cette méthode, la connaissance de la fonction d'excitation de la réaction nucléaire utilisée n'est donc pas indispensable.

Cependant, on doit irradier simultanément avec l'échantillon un étalon mince à teneur parfaitement connue de l'élément à doser. Or la fabrication de ce dernier pose des problèmes. Elle s'avère délicate, voire impossible, pour de nombreux éléments, de sorte que l'on doit avoir recours à des étalons secondaires jouant un rôle de moniteur de flux ou se servir de la relation (13) en mesurant avec précision l'intensité du faisceau.

La connaissance de la valeur exacte de la section efficace σ_0 pour l'énergie incidente des particules chargées est donc nécessaire. D'où l'importance de l'établissement de la fonction d'excitation de la réaction nucléaire utilisée.

Dans les deux autres méthodes on se sert d'étalons épais. On ne les développe pas ici. Pour plus de détails les concernant on pourra, par exemple, consulter la référence [2]. Leur emploi n'implique en effet pas la connaissance de la section efficace ou d'une autre constante nucléaire.

3. INFLUENCE DES REACTIONS NUCLEAIRES COMPETITIVES

Le principe de l'analyse par activation reposant sur la mesure de l'activité d'un radioélément Y produit par réaction nucléaire à partir de

l'élément X à doser, il y a interférence si Y peut être obtenu à partir d'autres éléments. On appelle réactions nucléaires compétitives celles qui conduisent à cette sorte de perturbation.

Par exemple, pour doser le carbone par activation aux photons γ, on se sert de la réaction

$$^{12}C\ (\gamma, n)^{11}C \text{ de seuil } E_s \simeq 18,7 \text{ MeV}$$

Or, le radioélément carbone-11 peut également être produit à partir de l'azote et de l'oxygène par les réactions suivantes:

$$^{14}N\ (\gamma, t)^{11}C \text{ de seuil } E_s = 22,7 \text{ MeV}$$
$$^{16}O\ (\gamma, \alpha n)^{11}C \text{ de seuil } E_s = 26 \text{ MeV}$$

De telles réactions nucléaires compétitives existent dans presque tous les cas. En ce qui concerne la détermination des éléments légers par activation aux photons γ ou particules chargées on pourra trouver des informations relatives à l'importance des erreurs analytiques que certaines pourraient introduire dans les références [2 à 4] et [7].

Les sources d'erreur sont très nombreuses. On peut les éliminer dans certains cas lorsque les énergies seuils sont très différentes ou minimiser leur influence par le choix de l'énergie maximale d'irradiation.

Il est donc très important, d'une part, de savoir a priori quels sont les éléments qui pourraient donner lieu par le moyen d'activation choisi à la formation du radioélément sur lequel repose le dosage de l'impureté recherchée, d'autre part, de connaître la grandeur de l'interférence que ces éléments perturbateurs pourraient entraîner.

En d'autres termes il s'agit d'avoir une vue d'ensemble de toutes les réactions nucléaires potentiellement possibles en se limitant naturellement aux énergies utilisées en analyse par activation et qui pourraient conduire à la production du même radioélément que celui par lequel on dose l'élément à déterminer, et de connaître leur seuil et surtout leur fonction d'excitation, ce qui permet d'évaluer à l'avance l'importance de l'erreur analytique qu'elles pourraient occasionner.

4. INTERET D'UNE COMPILATION DE CERTAINES CONSTANTES NUCLEAIRES

De ce qui précède il ressort que les constantes nucléaires qui interviennent constamment en analyse par activation sont avant tout les sections efficaces des réactions nucléaires utilisées et de celles qui pourraient donner lieu à des interférences.

Il nous semble par conséquent qu'il serait particulièrement intéressant si l'on pouvait disposer d'une compilation regroupant, en se limitant au domaine d'énergie propre à l'analyse par activation, les réactions nucléaires pouvant conduire à la formation des radioéléments dont on se sert dans les procédés analytiques, leur énergie seuil et les fonctions d'excitation correspondantes.

L'intérêt d'une telle compilation est évident. Elle permettrait d'un coup d'œil:

- d'avoir une idée précise des réactions nucléaires utilisables a priori pour déterminer un élément donné, compte tenu des possibilités d'irradiation dont on dispose
- de choisir les conditions d'irradiation optimales pour avoir le maximum de sensibilité avec le minimum d'interférences.

En analyse par activation aux particules chargées ou photons γ, principalement en ce qui concerne la détermination des éléments légers, il est impossible d'établir des règles générales liées uniquement à l'élément qu'il s'agit de doser. Dans presque tous les cas on doit tenir compte de l'environnement dans lequel ce dernier se trouve. Et, souvent, c'est lui qui impose la nature et l'énergie des particules convenant le mieux pour résoudre le problème posé.

De telles compilations évitent ainsi de perdre beaucoup de temps dans des essais préliminaires et surtout de commettre des erreurs analytiques par suite d'un mauvais choix des conditions expérimentales.

A propos des domaines d'énergie dans lesquels se situe l'analyse par activation, il est difficile de fixer d'une façon définitive des frontières. Il semble, en se référant aux travaux publiés par les principaux laboratoires utilisant de telles techniques, que les énergies maximales des photons et des particules chargées pourraient être limitées aux valeurs suivantes:

Photons γ	50 à 60 MeV
p	15 à 20 MeV
d	20 à 25 MeV
t	20 à 25 MeV
^{3}He	25 à 30 MeV
α	45 à 50 MeV

D'autre part, une limitation de ces compilations vers les basses énergies n'est pas souhaitable. Au contraire, il est toujours intéressant de connaître une fonction d'excitation jusqu'à l'énergie seuil.

Notamment, dans les méthodes d'analyse de couches minces par activation aux particules chargées, on utilise ces dernières à des énergies très voisines des seuils afin de limiter la zone activée à une très faible épaisseur. Il est ainsi intéressant de connaître, voire avec beaucoup de précision, la forme de la fonction d'excitation dans le voisinage de l'énergie seuil.

En ce qui concerne les dosages de traces d'éléments légers, pour lesquels les particules chargées et les photons γ sont les plus compétitifs, les radioéléments qui interviennent le plus souvent sont ceux indiqués dans le tableau I, dans lequel on trouve également les éléments qui sont déterminables par leur intermédiaire et les moyens d'activation correspondants les plus intéressants au point de vue de la sensibilité de détection et des interférences.

On peut ajouter à ces radioéléments quelques autres d'usage moins fréquent mais présentant néanmoins de l'intérêt pour doser certains éléments, comme par exemple le lithium [8] ou le soufre [9]. Ils sont rassemblés dans le tableau II.

En fait, la liste des radioéléments par lesquels des éléments, d'une façon générale et non restrictive, sont dosables par activation aux particules

TABLEAU I. PRINCIPAUX RADIOELEMENTS PAR LESQUELS LES ELEMENTS LEGERS Be, B, C, N, O et F SONT DETERMINES PAR ACTIVATION AUX PARTICULES CHARGEES ET PHOTONS γ

Radioélément	Eléments pouvant être dosés par son intermédiaire	Moyen d'activation à utiliser
^{11}C	Be	^{3}He ou α
$T_{\frac{1}{2}}$ = 20,3 min	B	p ou d
	C	γ, ^{3}He, α
	N	p
^{13}N	B	^{3}He ou α
$T_{\frac{1}{2}}$ = 9,96 min	C	p ou d
	N	γ, ^{3}He, α
	O	p
^{15}O	C	α
$T_{\frac{1}{2}}$ = 2,03 min	N	d
	O	γ
^{17}F	N	α
$T_{\frac{1}{2}}$ = 66 s	O	d
^{18}F	O	p, t, ^{3}He, α
$T_{\frac{1}{2}}$ = 109,7 min	F	γ, p, d, α

chargées ou photons γ est très longue. Cependant, le plus souvent, on se sert de ces derniers pour doser les éléments légers consignés dans les tableaux I et II.

Dans ces deux tableaux, l'utilisation des tritons comme moyen d'activation n'apparaît que pour le dosage de l'oxygène. Or il est fort probable que ce type de particules conviendra pour déterminer de nombreux autres éléments. Mais actuellement, peu d'accélérateurs sont conçus pour fournir des faisceaux intenses d'énergie supérieure à 5 MeV. Cependant, il serait intéressant dans le cadre d'une compilation de constantes nucléaires pour l'analyse par activation de prendre en considération les tritons dans la gamme d'énergie indiquée. En effet, on peut prévoir que dans l'avenir, certaines machines soient spécialement adaptées pour accélérer ces particules, d'autant plus que, sur le plan de l'analyse par activation, leurs possibilités semblent très prometteuses.

On peut trouver dans la littérature de nombreuses compilations de constantes nucléaires dont certaines rendent de très grands services aux spécialistes de l'analyse par activation.

Notamment, les tables des énergies seuils publiées par Howerton et coll. [10] se rapportant à des réactions de type (x, n), (x, 2n), (x, p), (x, np), (x, d), (x, nd), (x, t), (x, nt), (x, ^{3}He), (x, n^{3}He), (x, α), (x, nα) induites par des photons γ, des protons, des deutons, des tritons, des hélions-3 et des particules α.

TABLEAU II. AUTRES RADIOELEMENTS POUVANT PRESENTER UN INTERET ANALYTIQUE, NOTAMMENT POUR DOSER Li, Be, B, ^{18}O, F ET S

Radioélément	Eléments pouvant être dosés par son intermédiaire	Moyen d'activation à utiliser
^{6}He $T_{\frac{1}{2}} = 0,802$ s	Li	γ
^{8}Li $T_{\frac{1}{2}} = 0,884$ s	Be	γ
^{9}Li $T_{\frac{1}{2}} = 0,176$ s	B	γ
^{7}Be $T_{\frac{1}{2}} = 53,4$ j	Li B	p ou d p ou d
^{17}N $T_{\frac{1}{2}} = 4,2$ s	^{18}O F	γ γ
^{34}Cl $T_{\frac{1}{2}} = 32$ min	S	p ou d

En ce qui concerne les réactions photonucléaires, Hunt et coll. [11] ont réuni dans un rapport les fonctions d'excitation publiées dans la littérature. Bien que relativement ancien et incomplet, ce document s'avère cependant souvent utile.

On peut citer également la table des parcours et des pouvoirs de ralentissement des particules chargées de Williamson et coll. [12] dont on se sert couramment.

Il existe de nombreuses autres compilations, mais le plus souvent, les informations qu'elles contiennent sont d'une utilisation moins immédiate que les précédentes.

Par exemple, on peut mentionner celles de Fuller et coll. [13] pour les réactions photonucléaires, McGowan et coll. [14] pour les particules chargées, dans lesquelles on trouve les références relatives aux réactions nucléaires déjà étudiées. Ces documents sont très complets et fort intéressants, notamment lors de l'étude et de la mise au point de nouvelles méthodes. En effet, connaissant a priori les réactions nucléaires dont il faut tenir compte, elles permettent de retrouver les résultats expérimentaux déjà publiés à leur sujet.

5. COMPILATIONS PARTICULIERES DESTINEES DIRECTEMENT A L'ANALYSE PAR ACTIVATION

Elles devraient constituer le document de base des spécialistes de l'analyse par activation ou des analystes en général, dans lequel doivent

figurer en clair toutes les informations indispensables à connaître lorsque l'on se propose de déterminer un élément dans un milieu donné.

En particulier, à notre avis, elles devraient comporter pour chaque élément:

a) Les moyens d'activation utilisables pour le doser: les réactions nucléaires les plus intéressantes, leur énergie seuil et les fonctions d'excitation correspondantes.

b) Les limites de détection en fonction de l'énergie des particules choisies, pour une intensité de faisceau de référence: par exemple, 10 μA pour les particules chargées et 100 μA moyen pour le faisceau d'électrons générateurs de photons γ de freinage. Avec les particules chargées, la limite de détection dépendant du milieu analysé on pourra adopter comme matériau de référence l'aluminium, comme on l'a déjà proposé par ailleurs [2, 15]. Les limites de détection pour les autres matériaux s'en déduisent d'une manière très simple en tenant compte des parcours efficaces. Pour les photons γ il sera important de préciser la nature de la cible de conversion utilisée, son épaisseur, le diamètre du faisceau d'électrons, celui de l'échantillon et surtout la distance de ce dernier à la cible. En ce qui concerne ces paramètres on pourrait définir des valeurs standards.

c) Les éléments pouvant interférer par réactions nucléaires compétitives: leur énergie seuil, leur fonction d'excitation et, en fonction de l'énergie des projectiles incidents, l'importance relative de l'interférence qu'elles entraînent.

Il s'agirait là, naturellement, d'une compilation hautement spécialisée, mais qui, compte tenu du développement que connaît actuellement l'analyse par activation aux particules chargée et photons γ, nous semble justifiée.

Pour déterminer les limites de détection comme l'importance relative des réactions nucléaires compétitives, on peut procéder de plusieurs façons:

a) Si les fonctions d'excitation des réactions nucléaires considérées sont connues, on peut calculer les sections efficaces moyennes $\bar{\sigma}$ dans le cas des particules chargées et les coefficients k_E pour les photons γ. Même si l'on ne peut connaître qu'une valeur approchée de ces deux grandeurs, cela serait déjà intéressant. En se servant respectivement des relations (4) et (12) on obtient les limites de détection en définissant l'activité minimale correspondante et on peut estimer l'importance des interférences.

b) Un moyen plus direct consiste à déterminer les courbes d'activation des éléments auxquels on s'intéresse. Elles représentent en fonction de l'énergie et pour une intensité de faisceau donnée, l'activité induite dans ces derniers. Dans le cas de l'activation par photons γ on la rapporte à l'unité de masse d'élément. Pour les particules chargées il est commode de la relier à l'unité de teneur massique, par exemple 1 $\mu g \cdot g^{-1}$, de l'élément dans un milieu de référence donné.

En fait, ce sont de telles courbes d'activation qui sont le plus directement exploitables par les analystes. Elles permettent d'atteindre d'une façon immédiate les sensibilités de détection ainsi que l'importance relative des interférences, en fonction de l'énergie.

En ce qui concerne plus spécialement les éléments légers Be, B, C, N, O et F on pourra trouver des compilations de courbes d'activation, de courbes représentant la variation de l'importance relative des interférences ainsi que des tableaux donnant les sensibilités de détection d'une part, les quantités ou les teneurs des éléments qui par réactions nucléaires

compétitives entraînent une erreur analytique par excès de 100%, d'autre part, dans [2, 7, 15, 16] pour les particules chargées p, d, ^{3}He, α, et dans [2-4] pour les photons γ de freinage.

Krasnov et coll. [17] ont également déterminé un très grand nombre de courbes d'activation relatives aux éléments à partir desquels on peut obtenir les radioéléments ^{11}C, ^{13}N, ^{18}F, par activation aux p, d, ^{3}He, et α.

Depuis quelques années on voit apparaître de plus en plus, dans la littérature spécialisée, des publications avec de telles courbes qui s'avèrent d'une utilisation particulièrement simple et agréable.

CONCLUSION

Sur le plan de l'analyse par activation aux particules chargées ou photons γ, il est clair que la compilation idéale serait constituée d'une part par les courbes d'activation des éléments, telles que nous les avons définies, en y ajoutant les réactions nucléaires ainsi que leurs énergies seuils, et d'autre part par des tableaux contenant les sensibilités de détection, pour une intensité de faisceau donnée, ainsi que les éléments pouvant interférer et l'importance relative de l'erreur analytique qu'ils introduisent, en fonction de l'énergie.

On pourra adopter, pour les particules chargées, un milieu de référence arbitraire; on avait proposé l'aluminium [2, 15], mais tout autre matériau pourrait convenir. Pour les photons γ, des conditions expérimentales standards peuvent être parfaitement bien définiinies, de sorte que les informations les concernant soient effectivement exploitables quel que soit l'accélérateur employé, et cela sans ambiguïté.

A défaut d'une telle compilation à caractère très pratique, un recueil contenant les réactions nucléaires utilisables et celles dont il faut tenir compte par suite des interférences qu'elles pourraient engendrer, ainsi que leurs énergies seuils et les fonctions correspondantes, rendrait déjà de très grands services. Des tableaux d'excitation donnant les valeurs des sections efficaces moyennes $\bar{\sigma}$ des réactions nucléaires par particules chargées ainsi que les coefficients d'étalonnage k_E pour les photons γ, en fonction de l'énergie, compléteraient très utilement les données précédentes. Le calcul des coefficients k_E semblant particulièrement complexe, ceux-ci pourraient à la rigueur être remplacés, bien que pratiquement elles soient beaucoup moins intéressantes, par les sections efficaces intégrées $\Sigma = \int \sigma_E \, dE$ des réactions photonucléaires, qui permettent en effet d'apprécier d'une manière approximative l'importance de l'activation d'un élément, notamment par comparaison avec un autre.

Comme on l'a déjà indiqué, de telles compilations sont largement justifiées compte tenu du développement que connaissent actuellement les méthodes d'analyse par activation aux particules chargées ou photons γ.

Elles pourraient constituer le document de travail indispensable aux analystes, au même titre que les catalogues de spectres γ, de sections efficaces des réactions nucléaires par neutrons thermiques, etc.

Dans tous les cas, elles faciliteraient la tâche en évitant des pertes de temps souvent considérables par suite des tâtonnements préliminaires fréquemment imposés par l'absence d'informations précises, et surtout les erreurs analytiques consécutives à un mauvais choix des conditions expérimentales d'activation.

On s'est surtout préoccupé des éléments légers. Mais les particules chargées ou les photons γ s'avèrent également des moyens d'activation très intéressants pour doser d'autres éléments. Les compilations précédentes ne doivent donc pas être limitées aux seuls éléments légers mais se rapporter à tous ceux pour lesquels ces méthodes analytiques sont effectivement utilisables.

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DISCUSSION

W.B. LEWIS: Would not your techniques also be very appropriate for analysing for the minor isotopes among the light elements, for example, ^{13}C?

Ch. ENGELMANN: They would, of course. In fact, they can sometimes be used for this purpose by taking advantage of the specific features of the reaction.

Z. SUJKOWSKI: You mention in your paper that it is easy to define standard conditions for photoactivation and you propose quoting the electron current. I wonder, however, whether this will give you a unique determination of the irradiation conditions. The photon flux, which you are ultimately interested in, will in general depend on the geometry used, electron target, etc. Perhaps it would be better to quote the photon flux at the irradiated target and the maximum photon energy. I have one other comment: it would be very useful if standards (standard reactions) for measuring photon flux were worked out, as has been done in the case, for example, of (n, γ) activation.

Ch. ENGELMANN: I agree with you but unfortunately there is a difficulty, namely, the fact that the photon spectrum is continuous. In my opinion, accurate measurements of a continuous photon spectrum for use as analysis standards is not a very convenient solution. For example, if there is a variation in your spectral distribution, you may have a very considerable variation in your activation and there is no certainty whatever that your equipment for measuring gamma photon flux will be able to include this or make you aware of it. It is for these reasons that we prefer to measure the intensity and energy of the electron beams, which we consider to be more tangible parameters. Moreover, it is these parameters which govern the shape of the spectrum and the intensity of gamma flux in the region of interest.

NUCLEAR REACTIONS INDUCED BY FAST PARTICLES
Accuracy of the calculated activities

U. JÄGER, H. MÜNZEL
Nuclear Chemistry Group,
Institute for Radiochemistry,
Nuclear Research Centre, Karlsruhe,
Federal Republic of Germany

Abstract

NUCLEAR REACTIONS INDUCED BY FAST PARTICLES: ACCURACY OF THE CALCULATED ACTIVITIES.

Many applications and calculations in activation analysis are using the excitation functions for the nuclear reactions published in the literature or obtained from the systematics. In general, it is very difficult to judge the accuracy of the individual value obtained in such calculations. Therefore, an estimation was made of the accuracy that can be expected. This was done by calculating the activity produced in thick targets using the excitation functions for a given nuclear reaction published by different authors. The results are discussed with regard to the errors of excitation functions stated in the publications.

In those cases where no experimental excitation functions are available one has to resort to the estimated values based on the systematics. To gain an impression of the errors expected for the activities obtained with the aid of such values several calculations were made and compared with available experimental data.

These calculations were done for nuclear reactions induced by charged particles and fast neutrons.

For many applications, such as the calculation of the sensitivities or detection limits of activation analysis, excitation functions for nuclear reactions are necessary. These excitation functions were normally obtained from the literature or from systematics. In general, it is very difficult to judge their accuracy. Therefore, the expected uncertainty of the excitation functions was estimated.

For charged particles, this estimation was done by calculating the activity which would be produced in a thick target, by using the excitation functions for a given nuclear reaction published by different authors. The mean value was calculated and compared with the individual values.

The drawback of this procedure is, of course, that the mean value is treated as the true one. But, in our opinion, this is the only method of obtaining at least an approximation for the accuracy of the excitation functions given.

As examples, the (α, xn) and (p, xn) reactions were chosen. All reactions of this type were used, where two or even more excitation functions are listed in the extensive compilations of excitation functions for charged-particle reactions [1].

The distribution of the deviations from the mean values is shown in the histogram of Fig.1. To gain an impression of the probability that the deviation is less than a given value, the histogram has been integrated. The upper part of Fig.1 contains the results. It is, for example, shown that in 70% of the cases considered the deviations are less than 20%.

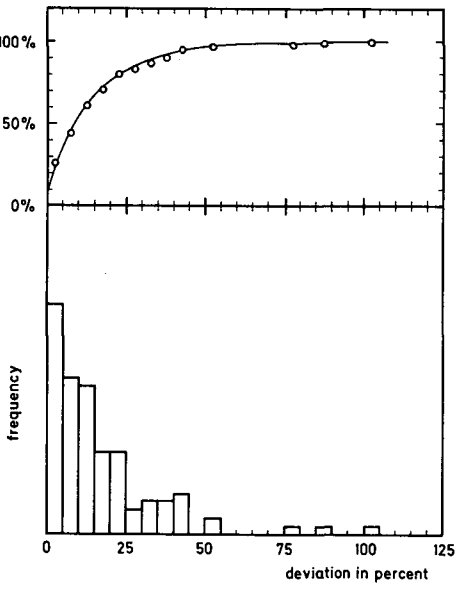

FIG. 1. Frequency distribution of deviations of the thick-target yields from the mean value for charged-particle reactions. The upper part shows the integration of the histogram.

In this evaluation of the data, one has to note that in the cases where excitation functions were determined twice or even more times, the authors normally did know the result of the foregoing work and their own values were probably checked carefully if deviations occurred. Therefore, in one or another case where only a single excitation function is known, the accuracy is possibly not so good as could be expected from Fig.1.

Furthermore, it is interesting to know whether the deviations are within the limits of error of the experimental data. Unfortunately, no errors are given explicitly for excitation functions. But as an approximation, the errors stated for the cross-sections can be used for this comparison.

The ratio of the percentage deviation from the mean value to the errors stated in the publications was calculated. The distribution of these values is shown in the lower part of Fig.2. The integration of the histogram in the upper part of the same figure shows that only 60% of the deviations obtained are smaller than the deviations expected from the errors given in the publications.

In the cases where no experimental excitation functions are available, one has to resort to estimated values based on the systematics. To get an impression of the uncertainty of these estimated excitation functions, the thick target yields were calculated and compared with the mean value of the experimental data used before.

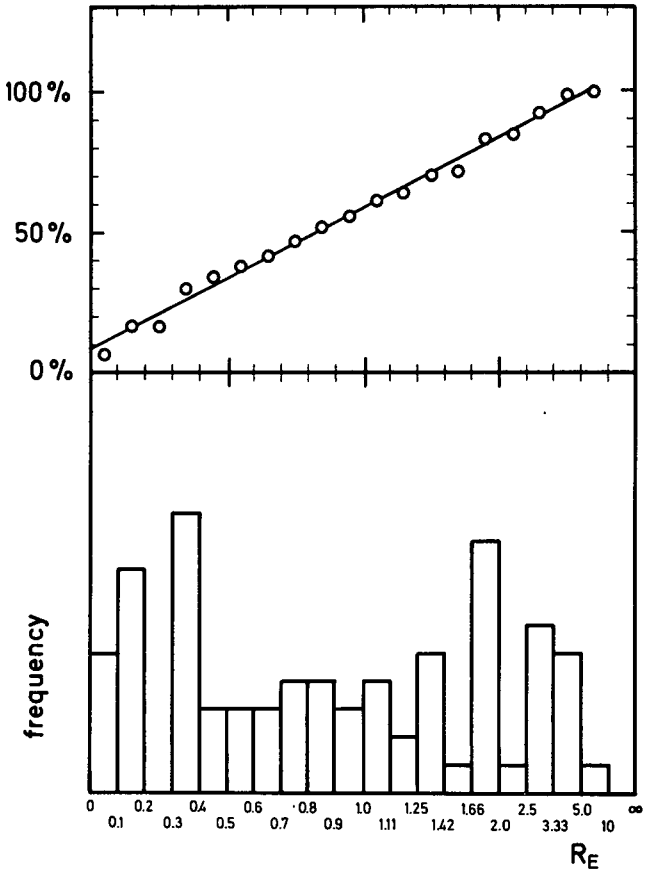

FIG. 2. Frequency distribution of the ratio R_E for charged-particle reactions. R_E is the ratio of the individual deviations of calculated thick-target yields from the mean to the errors stated in the publications. The upper part shows the integration of the histogram. Note: The abscissa is divided into 0.1 steps up to 1.0 and above by the corresponding reciprocal values.

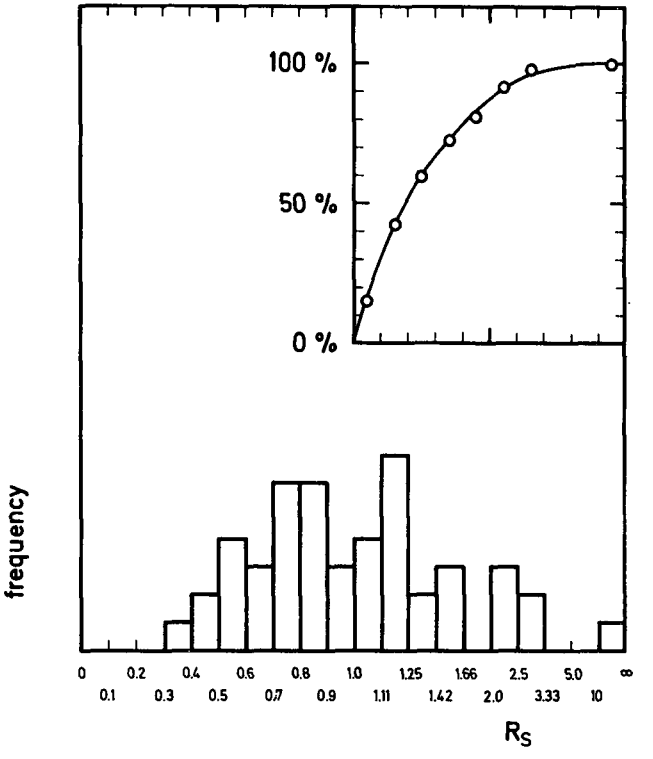

FIG. 3. Frequency distribution of the ratio R_S of thick-target yields obtained by systematics to experimental mean values for charged-particle reactions. The upper part shows the common integration of the frequency for factors above 1.0 and the corresponding reciprocals, respectively. Note: The abscissa is divided into 0.1 steps up to 1.0 and above by the corresponding reciprocal values.

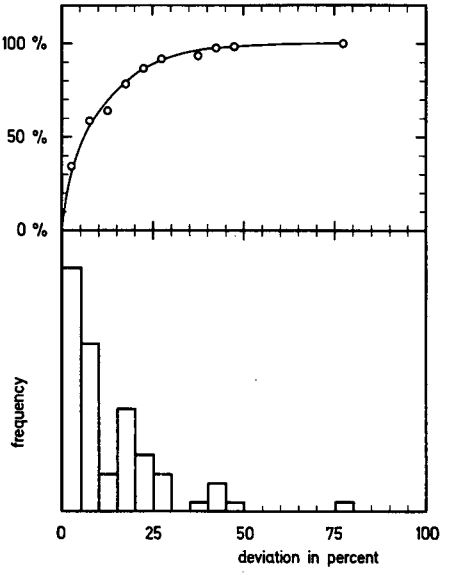

FIG. 4. Frequency distribution of deviations of the thick-target yields from the mean value for 14 MeV-neutrons. The upper part shows the integration of the histogram.

This comparison was performed with the systematics developed in our laboratory [2]. The distribution of deviations is shown in the histogram of Fig.3. In the upper part, the probability that deviations are less than a certain factor and less than its reciprocal value, respectively, is given. This probability was obtained by simply integrating the corresponding frequencies of the histogram. It is shown that in 50% of the cases the deviations for the excitation functions obtained by systematics are smaller than a factor of 1.25 and in 90% of the cases are smaller than a factor of 2. One extreme deviation occurs in the case of $^{54}Fe(\alpha, 2n)^{56}Ni$ where ^{56}Ni is a doubly magic nuclide.

Estimations similar to those for charged particles were carried out for 14-MeV neutrons. From the cross-sections published by different authors, the mean value was calculated for a given reaction. To judge the accuracy of the experimentally obtained cross-sections, they were compared with the mean value. As examples, the (n, 2n)-reactions were chosen. The distribution of the deviations of the individual values from the mean value is shown in Fig.4. To obtain the probability that the deviations from the mean are less than a given value, the histogram was integrated, as is shown in the upper part of Fig.4. There, one can see that in 80% of the cases the deviations between the mean values and the individual experimental data are less than 20%.

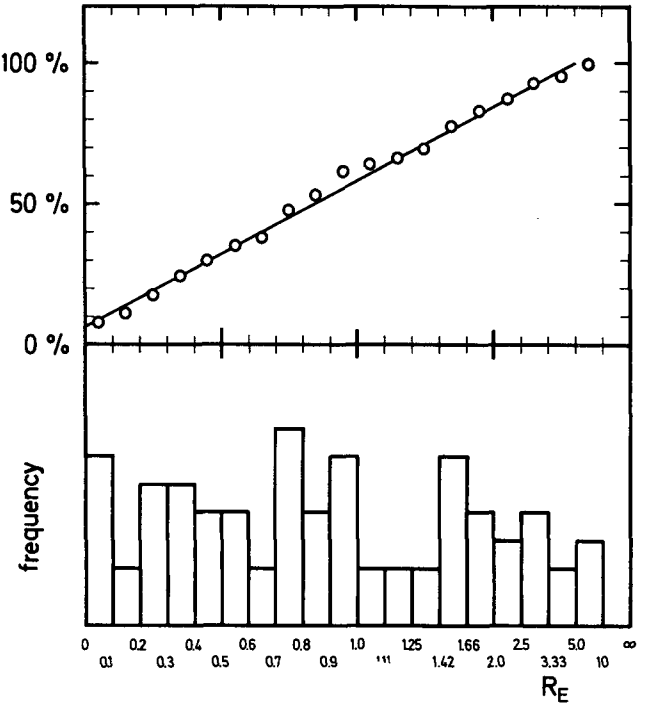

FIG. 5. Frequency distribution of the ratio R_E for 14-MeV neutrons. R_E is the ratio of the individual deviations of calculated thick-target yields from the mean to the errors stated in the publications. The upper part shows the integration of the histogram. Note: The abscissa is divided into 0.1 steps up to 1.0 and above by the corresponding reciprocal values.

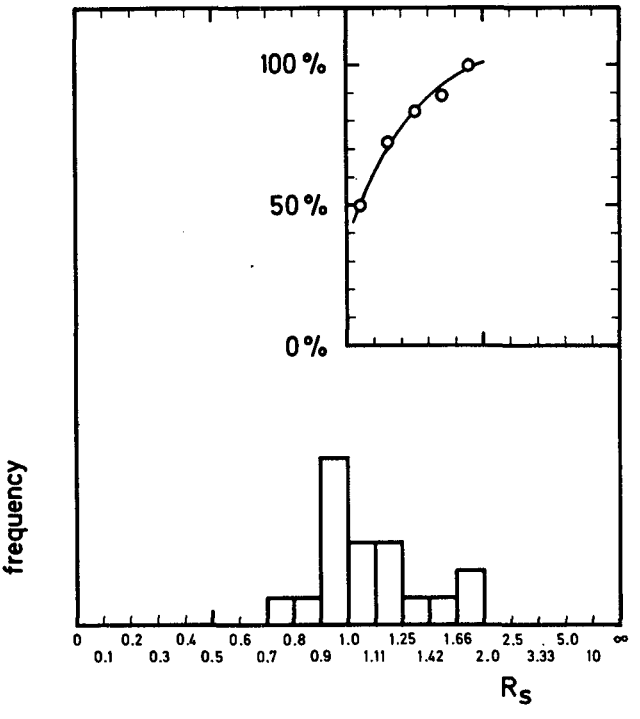

FIG. 6. Frequency distribution of the ratio R_S of thick-target yields obtained by systematics to experimental mean values for 14 MeV neutrons. The upper part shows the common integration of the frequency for factors above 1.0 and the corresponding reciprocals, respectively. Note: The abscissa is divided into 0.1 steps up to 1.0 and above by the corresponding reciprocal values.

A comparison of the errors stated in the publications with the deviations of the individual values is shown in Fig.5. The curve is very similar to that found for charged-particle reactions. Again, only in 60% of the cases, the deviations are within the errors stated by the authors.

The calculated (n, 2n) cross-sections of Pearlstein [3] were compared with the experimental mean value. The result is shown in Fig.6. The comparison shows that more than 70% of the deviations are smaller than a factor of 1.25 and all values are better than a factor of 2.

Thick-target yields for fast neutrons could not be calculated in the same way as was done for charged particles, because of the lack of experimental data. Systematics of excitation functions for (n, 2n) reactions were developed by Krivan and Münzel [4]. They showed that, with one exception, the agreement with experimental data was better than 25%.

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LES BESOINS EN DONNEES NUCLEAIRES DANS LE DOMAINE DE L'ANALYSE PAR ACTIVATION AVEC LES PARTICULES CHARGEES DE MOYENNE ENERGIE

J.L. DEBRUN, J.N. BARRANDON, Ph. ALBERT
CNRS, Laboratoire d'analyse par activation Pierre Sue,
Groupe d'application des réactions nucléaires à l'analyse chimique,
Centre d'études nucléaires de Saclay, France

Abstract–Résumé

NEED FOR NUCLEAR DATA IN ACTIVATION ANALYSIS USING CHARGED PARTICLES OF INTERMEDIATE ENERGY.

After a brief survey of the research work done by the group dealing with the application of nuclear reactions to chemical analysis (Saclay), the authors indicate the nuclear data needed in activation analysis using charged particles of intermediate energy. They show that often these data are not sufficient and sometimes even missing, and that there is a real need for both experimental work and data compilation and evaluation in this field of research.

LES BESOINS EN DONNEES NUCLEAIRES DANS LE DOMAINE DE L'ANALYSE PAR ACTIVATION AVEC LES PARTICULES CHARGEES DE MOYENNE ENERGIE.

Après avoir brièvement rappelé les études effectuées par le Groupe d'application des réactions nucléaires à l'analyse chimique (Saclay), les auteurs indiquent quelles sont les données nucléaires dont on a besoin dans le domaine de l'analyse par activation avec les particules chargées de moyenne énergie. Ils montrent que ces données sont souvent insuffisantes, parfois inexistantes, et qu'il existe un réel besoin, à la fois de travail expérimental et de travail de compilation et d'évaluation de données dans ce secteur de recherche.

EXPOSE DES RECHERCHES DU GARNAC

Le groupe d'application des réactions nucléaires à l'analyse chimique utilise les trois types de particules habituels: neutrons, photons et particules chargées. Ces dernières sont celles que nous utilisons le plus et nous ne parlerons pas des problèmes relatifs aux autres. Nos recherches ont deux buts principaux:

- l'élaboration de nouvelles méthodes ou possibilités d'activation
- l'utilisation de méthodes déjà connues, ou de méthodes à l'essai, pour résoudre des problèmes concrets.

Les chercheurs qui travaillent en analyse par activation avec les particules chargées se sont surtout attachés à doser les éléments légers (Be, B, C, N, O, F) et ont négligé les éléments de numéro atomique $Z > 20$, à l'exception de quelques-uns [1]. Les seuls radioisotopes utilisés étaient bien connus (^{11}C, ^{13}N, ^{18}F, ^{15}O, émetteurs β^+ purs) et pour les dosages on utilisait dans la majorité des cas des étalons de même nature que les impuretés à analyser. Enfin, des séparations chimiques étaient le plus souvent effectuées. Dans le cas des éléments plus lourds cités ci-dessus [1], il n'y a en fait jamais eu beaucoup de dosages réellement effectués et dans les cas où il y a vraiment eu une application,

il s'est toujours agi du dosage d'un seul élément à la fois, généralement avec séparation chimique après irradiation. Il n'y avait donc pas de difficulté du point de vue des données nucléaires. Actuellement, pour nous, le problème est complètement différent puisque nous mettons au point l'analyse non destructive d'un grand nombre d'éléments à la fois, à l'exclusion des éléments légers.

Nous avons jusqu'ici utilisé seulement les protons; ces particules semblent être les plus intéressantes et permettent de doser en analyse non destructive sans interférences une cinquantaine d'éléments dans une quinzaine de matrices différentes [2].

DONNEES NUCLEAIRES NECESSAIRES POUR L'ANALYSE

Nous avons ressenti le manque de données nucléaires en ce qui concerne aussi bien l'analyse qualitative que l'analyse quantitative. Pour l'analyse qualitative, les données nécessaires sont les périodes radioactives et les schémas de désintégration; pour l'analyse quantitative nous avons besoin en outre des taux de production (sections efficaces intégrées). On peut dire que si toutes ces données sont assez bien connues pour la majorité des radioisotopes utilisés en activation neutronique, il n'en est pas de même pour les radioisotopes obtenus spécialement par activation au moyen de particules chargées. Ces isotopes nécessitent pour leur production un accélérateur de particules et comme ces machines sont plus rares que les réacteurs, il est probable que c'est pour cette raison qu'il existe pour eux moins de données ou des données qui ne sont pas aussi récentes ni aussi précises que celles qui sont relatives aux produits d'activation neutronique.

L'ANALYSE QUALITATIVE

Pour cette analyse, qui est réalisée par spectrométrie γ avec un détecteur Ge(Li), l'expérimentateur a besoin de connaître avec une précision qui est fonction de la résolution de son ensemble d'analyse, les valeurs des énergies γ des radioisotopes qu'il a produits; ceci n'est d'ailleurs pas suffisant et il faut connaître aussi les rapports d'émission de ces rayonnements et faire pour plus de sûreté plusieurs mesures sur un même échantillon pour opérer une sélection grâce aux périodes radioactives

Les personnes qui travaillent en activation neutronique savent combien il est pénible d'opérer manuellement ce travail de détective qui consiste à identifier les radioisotopes; les calculateurs électroniques nous permettent maintenant d'effectuer ce travail automatiquement à la condition de disposer d'ensembles de données cohérentes entre elles. Cette dernière condition peut être considérée comme à peu près réalisée en activation neutronique; en activation avec les particules chargées, le gros du travail est à faire. En effet, même en admettant que l'on se satisfasse, pour une analyse qualitative, des schémas de désintégration et des périodes radioactives publiés à ce jour, il n'existe comme données utilisables en ce qui concerne les énergies que celles qui sont relatives aux radioisotopes formés à la

fois par réactions (n, γ), (n, α) ou (n, 2n) et par réactions avec les particules chargées; il y a aussi quelques rapports de Cline et Heath sur les isotopes déficients en neutrons [3].

Beaucoup des isotopes communs aux réactions avec les neutrons ((n, γ) par exemple) et aux réactions avec les particules chargées ((D, p) par exemple) ne sont pas utilisables en analyse par activation avec des particules chargées, à cause de nombreuses interférences, si bien que nous disposons à l'heure actuelle des spectres γ d'environ 50 radioisotopes alors qu'il y a environ 200 radioisotopes utilisables.[1]

Pour en terminer avec l'analyse qualitative, il nous semble important de souligner que, la nature et les sections efficaces des réactions nucléaires variant avec l'énergie des particules, il existe donc pour chaque élément naturel irradié avec une particule donnée un nombre illimité de spectres γ différents. (Ce problème existe à un degré bien moindre en activation avec les neutrons de réacteur.) Les catalogues de spectres actuels [4] s'efforcent de présenter des spectres de radioisotopes purs, et non pas des spectres de l'élément naturel irradié; une raison en est que suivant les temps d'irradiation et de refroidissement il y a là aussi un nombre illimité de spectres.

Nous pensons cependant qu'il serait utile de disposer, en activation avec les particules chargées, de spectres d'éléments irradiés à différentes énergies choisies en fonction des seuils de réaction.

L'ANALYSE QUANTITATIVE

Pour le dosage simultané de nombreux éléments dans un même échantillon, nous nous trouvons dans la même situation qu'en analyse neutronique utilisant la méthode du monostandard. En effet, nous ne pouvons pas irradier simultanément un étalon de chaque élément recherché, et l'échantillon; nous irradions un moniteur de flux et un échantillon, ou bien l'échantillon seul, et nous mesurons alors le nombre de microcoulombs reçus. Il est nécessaire de mesurer les activités spécifiques obtenues à partir d'étalons des éléments à doser, en fonction de l'activité du moniteur du flux. Nous n'avons en principe pas besoin de connaître les sections efficaces des réactions utilisées, nous travaillons relativement à un comparateur unique. Par contre, nous avons besoin de connaître les périodes des radioisotopes produits et les schémas de désintégration de façon à effectuer les corrections pour les temps d'irradiation et de décroissance ainsi que pour les problèmes de filiation. Les données dont il est question ont été compilées [5] mais il y a souvent des écarts qui nous laissent perplexes quant à la valeur à choisir. Il est clair que la précision de nos analyses dépend de la précision sur ces données et qu'un gros travail reste à faire dans ce domaine.

Nous avons dit qu'il était possible de s'affranchir de la connaissance des sections efficaces. En fait, si nous voulons effectuer des dosages aussi précis que possible, il ne nous faut pas utiliser les sections efficaces

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[1] Ceci était vrai au moment où ce rapport a été écrit. Les auteurs signalent qu'un nouveau rapport par Heath d'avril 1973 contient beaucoup des informations en question.

publiées, mais bien mesurer les taux de production relativement les uns aux autres, à l'énergie choisie. Néanmoins, il est intéressant de connaître ces sections efficaces au moins approximativement, pour les réactions que l'on n'a pas encore étudiées soi-même de façon à évaluer la possibilité d'un dosage, et à estimer les niveaux d'activité obtenus à partir d'un échantillon ou la valeur d'une interférence.

Qu'il nous soit permis de dire que les sections efficaces ont été mesurées par des physiciens et chimistes nucléaires et que la plupart du temps, elles sont inutilisables en activation (du moins directement) parce que présentées par exemple sous forme de sections efficaces différentielles (et non totales). Si nous consultons des compilations existantes [5], elles sont de notre point de vue trop touffues, contenant de nombreuses informations inutiles, et incomplètes car il manque des réactions ou des énergies que nous utilisons.

Enfin, pour clore le chapitre de l'analyse quantitative, il nous semble utile de parler brièvement du problème des programmes de traitement automatique des spectres. Il existe à l'heure actuelle d'innombrables programmes écrits soit en Fortran et utilisés avec de gros calculateurs, soit en assembleur et utilisés surtout avec de petits calculateurs du genre du PDP. 9.

Sans être spécialistes, nous pouvons nous rendre compte du fait que ces programmes ont des performances semblables sur certains points, mais que certains d'entre eux traitent mieux le problème de l'intégration des pics et la résolution des doublets, ou que d'autres traitent mieux le problème des petits pics. Il serait souhaitable d'étudier les différents programmes, d'en extraire les parties intéressantes et de les présenter dans un recueil spécialisé.

CONCLUSION

Comme toute autre discipline scientifique en plein développement, l'activation avec les particules chargées de moyenne énergie a besoin de constantes toujours plus précises et d'ensembles de valeurs bien connues les unes par rapport aux autres et que l'on n'utilise que dans cette discipline.

Comme types de constantes utilisées dans d'autres disciplines, nous pouvons citer les énergies γ, les périodes radioactives et les schémas de désintégration; il y a un important travail expérimental à faire dans ces domaines. Comme constantes qu'il n'est pas nécessaire de connaître avec précision, mais qui sont utiles pour faire des évaluations avant une expérience, nous pouvons citer les sections efficaces de réactions nucléaires; à l'heure actuelle il est sans doute encore valable d'extraire de l'abondante littérature de physique nucléaire les données utilisables en activation.

D'un autre côté, les valeurs ou renseignements utilisés spécifiquement en activation avec les particules chargées sont des spectres γ obtenus dans des conditions standards, des programmes de calculs et des taux globaux de production. Une meilleure connaissance de toutes ces constantes et données permettra de mettre au point une analyse par activation de plus en plus sûre et précise, ce qui est un des objectifs que cherchent à réaliser toutes les méthodes d'analyse.

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DISCUSSION

W.B. LEWIS: Your problem of the non-destructive analysis of a large number of elements present together seems to call for more sophisticated methods of display for observation – for example, maintaining a continuous display while the incident particle energy is made to pass over a threshold can reveal that threshold in the presence of a complex background. Do you not agree?

J.L. DEBRUN: Our problem is not the one to which you refer. We are not looking at groups of particles, we are doing trace analyses by activation and for the cases of interest to us no other nuclear method seems applicable.

A PROPOSED METHOD FOR ASSAYING SULPHUR BY PROTON ACTIVATION ANALYSIS USING A LOW-ENERGY ACCELERATOR

M. A. ESWARAN, N. L. RAGOOWANSI, M. ISMAIL
Nuclear Physics Division, Bhabha Atomic Research Centre, Trombay, Bombay, India

Abstract

A PROPOSED METHOD FOR ASSAYING SULPHUR BY PROTON ACTIVATION ANALYSIS USING A LOW-ENERGY ACCELERATOR.

A new method of proton activation analysis is proposed for assaying sulphur using the capture reaction $^{32}S(p,\gamma)^{33}Cl$. The method involves short irradiations of a few seconds by a mechanically chopped beam from a low-energy Van-de-Graaff accelerator, coupled with the measurement of the residual positron activity of $T_{\frac{1}{2}} = 2.52$ s, resulting from the decay of ^{33}Cl. A plastic scintillation detector was used for positron counting in conjunction with a multi-channel analyser operated in the multi-scaling mode with a dwell time of 40 ms per channel. The time for irradiation was 4 s and for counting was 10 s. The repeated irradiation-counting sequence was automatically controlled by a timer-relay unit which effects mechanical chopping of the beam. This activation reaction features a high-abundance target isotope (95%), and the method is highly selective since only the counts showing the correct half-life are included for the analysis.

This proposal is based on our detailed study (Phys. Rev. C5 (1972) 1270) of the excitation function for this reaction in the bombarding-energy range 3.3 to 5.4 MeV, applying this technique and using the model C-N Van-de-Graaff Accelerator at Trombay. A sensitivity of a few $\mu g/cm^2$ can be achieved by this method which is rapid and uses only a low-energy accelerator. This method can be fruitfully used in assaying sulphur in different materials, e.g. in petroleum products.

INTRODUCTION

Activation analysis using charged-particle beams has been based so far on the measurement of radioisotopes with half-lives ranging in most cases from minutes up to several days. Not much work has been reported on the analytical exploitation of nuclear reactions yielding radioactive species with half-lives of a few seconds. Such an approach offers, however, several advantages by providing for rapid and sensitive methods of analysis in maximizing the build-up of the radioisotope of interest close to saturation levels.

The determination of sulphur is of great importance in metallurgy, petroleum products, etc. It is, unfortunately, very difficult to determine the sulphur content by activation analysis either with thermal or fast neutrons because several elements interfere [1, 2].

In this paper, it is proposed that the determination of sulphur can be done by using the proton activation analysis, with the help of the capture reaction $^{32}S(p,\gamma)^{33}Cl$ (Q = 2.29 MeV) with a relatively low-energy accelerator. This activation reaction features (a) a target isotope of high abundance (95%), and (b) a product nuclide which emits high-energy positrons ($E_{\beta^+max.} = 4.51$ MeV) with a short half-life of 2.52 s [3].

This proposal is based on our study [4] of the excitation function for this reaction up to a proton bombarding energy of 5.4 MeV, using an activation technique.

EXPERIMENTAL DETAILS

Thin sulphur targets were prepared by evaporating natural antimony sulphide (Sb_2S_3) in vacuum to a thickness of about 300 $\mu g/cm^2$ on to a thick gold backing. This target was mounted at the centre of a 5-cm diameter thin-walled (0.8 mm) stainless steel chamber, coupled to the beam tube. The target was oriented at 45° to the incoming proton beam from the 5.5-MeV model C-N Van-de-Graaff accelerator at the Bhabha Atomic Research Centre at Trombay. The beam was collimated to a size of 5 mm by tantalum apertures. The beam was stopped in the target backing and monitored by a current integrator.

At 90° to the beam, a β-detector consisting of a 10-cm diameter × 2.5-cm thick plastic scintillator mounted on a XP1040 photomultiplier was placed with its front face 5 cm from the target. The pulses from this detector corresponding to a positron energy above 500 keV, selected by suitable electronics were fed to a Nuclear-Data 4096 channel analyser used in multi-scaling mode with 40 ms dwell time per channel. Using a beam of 2 μA, the operating cycle (bombard the target with the beam for 4 s and count the positrons with the analyser in the multi-scaling mode for 10 s) was repeated until a fixed amount of charge from the beam was accumulated as monitored in the current integrator. The periodical chopping of the beam was accomplished by a mechanical beam stopper operated by an electromagnet. The irradiation-counting sequence was

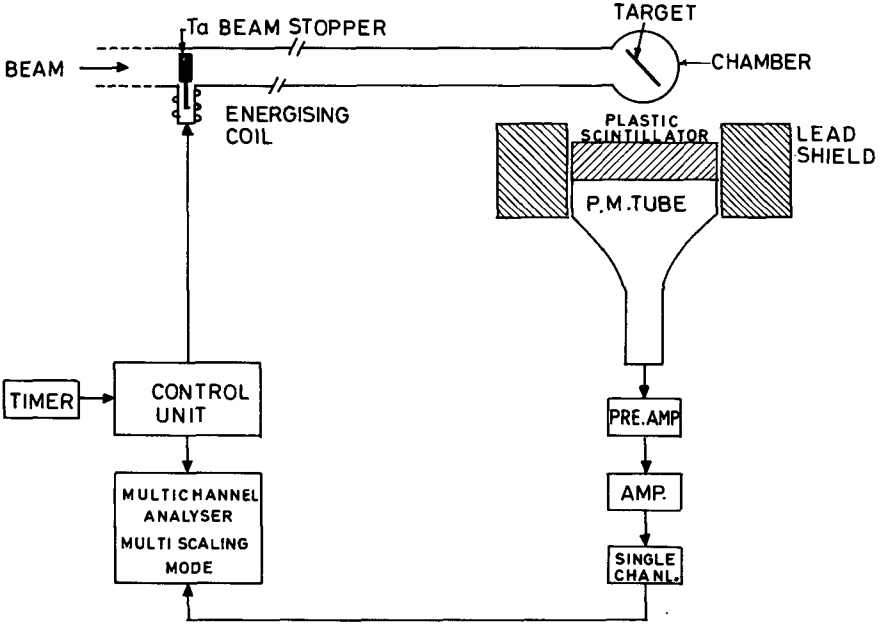

FIG. 1. Experimental set-up.

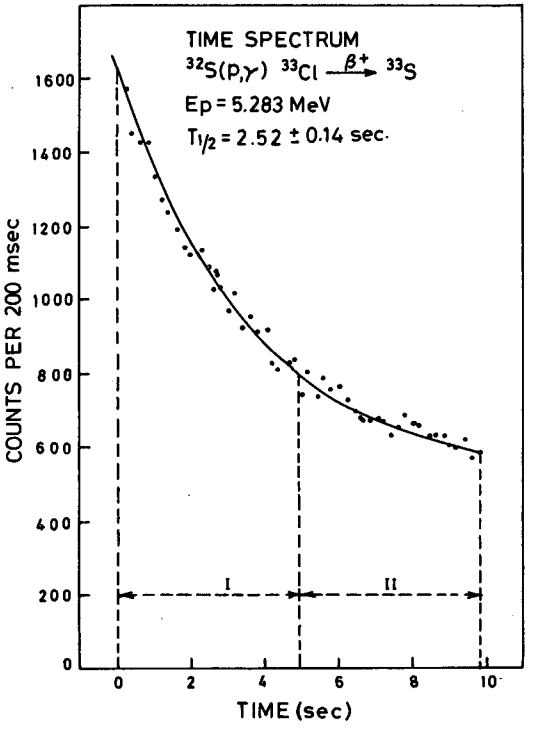

FIG. 2. Time spectrum of the positron decay of ^{33}Cl produced in the $^{32}S(p,\gamma)^{33}Cl$ reaction. The solid line through the experimental points is the computer fit obtained for an exponentially decaying function with constant background.

controlled automatically with a timer-relay unit, thus providing for the reproducibility needed to carry out repetitive irradiations on the same sample and accumulation of the decay data for achieving better counting statistics. The target, detector and counting set-up are shown in Fig.1.

RESULTS AND DISCUSSION

The resulting time spectrum from the positron detector, in addition to a constant background, showed the expected activity with a half-life of 2.52 s from the decay of ^{33}Cl produced in the $^{32}S(p,\gamma)^{33}Cl$ reaction. ^{33}Cl decays by positron emission to the ground state of ^{33}S with an end-point energy of 4.51 MeV. An example of the decay data, recorded in about 25 min with a bombarding energy of E_p = 5.283 MeV is shown in Fig.2. A computer fit of the data obtained by using an exponentially decaying function with constant background is also shown in the figure.

With this arrangement, the excitation function for the $^{32}S(p,\gamma)^{33}Cl$ reaction has been obtained in the following way. The counts in the decay curve in the first-5second period (region I in Fig.2) are added and from this sum the counts in the later 5-second period (region II) are subtracted.

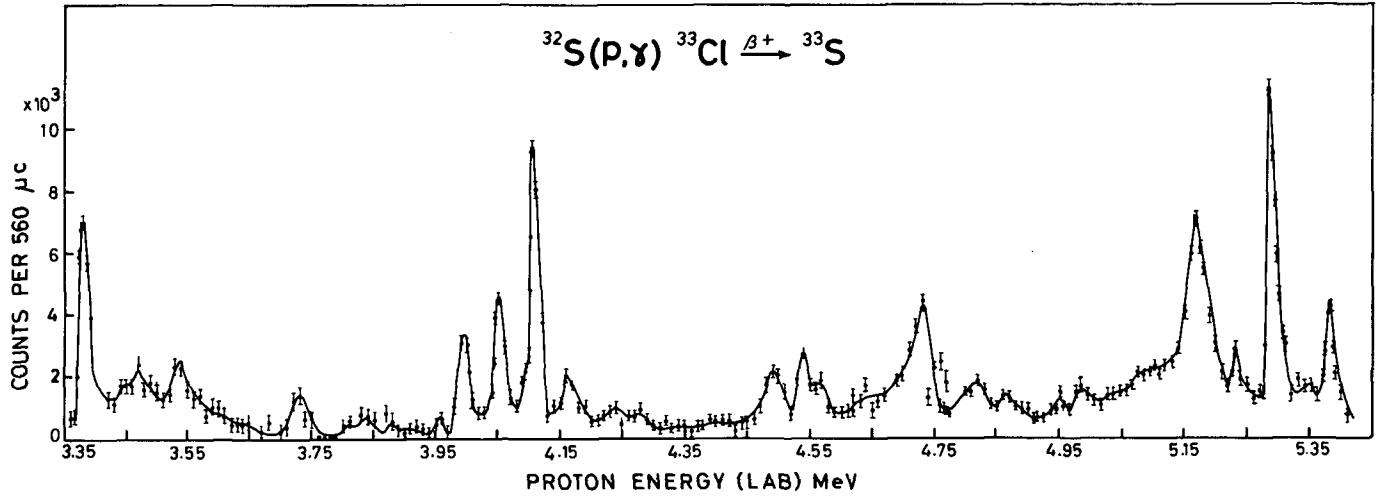

FIG. 3. Yield curve of ^{33}Cl activity resulting from the capture of protons by ^{32}S.

This difference gives, in effect, the measure of the yield of the $^{32}S(p,\gamma)^{33}Cl$ reaction. In this way, the yield curve was obtained for the bombarding energy range E_p = 3.3 to 5.4 MeV in steps of 10 keV using the target which was 14 keV thick for 5 MeV protons. This excitation function is shown in Fig.3.

As is clear in these measurements, the identification of the product isotope ^{33}Cl is made by its characteristic half-life of 2.52 s measured by counting the high-energy positrons ($E_{p,\,max}$ = 4.51 MeV) emitted in its decay. At the relatively low bombarding energies (< 5.5 MeV) used in these measurements there is no interfering reaction which will lead to the same activity. However, it is to be noted that if a silicon impurity is present in the sample the $^{28}Si(p,\gamma)^{29}P$ reaction will lead to ^{29}P which is a positron emitter with a maximum positron energy of 3.945 MeV decaying with a half-life of 4.23 s [3]. The contribution from this reaction can be eliminated if the discrimination level in the positron detector is raised to maximum of positron energy from this reaction at the expense of part of the contribution from the $^{32}S(p,\gamma)^{33}Cl$ reaction. Alternatively, the contribution from this reaction can also be estimated by suitable analysis of the decay data since the half-lives of ^{33}Cl and ^{29}P are different, i.e. 2.52 s and 4.23 s, respectively.

As a result of these measurements, it is proposed that this technique employing the $^{32}S(p,\gamma)^{33}Cl$ reaction can be fruitfully used in assaying sulphur in the surface layers of solid samples or in liquid samples such as petroleum products by comparison with suitable standard samples. The range of the 5-MeV protons in iron, e.g. is 65 mg/cm^2 [5]. Few tens of μg/cm^2 of sulphur can easily be estimated by this technique.

CONCLUSION

The method of proton activation analysis proposed here for assaying sulphur uses the radiative capture reaction $^{32}S(p,\gamma)^{33}Cl$. The method involves short irradiations of a few seconds by mechanically chopped beam from a low-energy (5.5 MeV) proton accelerator coupled with the counting of the residual positron activity resulting from the decay of ^{33}Cl with a half-life of 2.52 s. A simple plastic scintillation detector is used for positrons. The method features a high-abundance target isotope and is highly selective. The sensitivity and precision depends on the size of the detector and number of repetitive irradiations, for obtaining the counting statistics considered necessary. This technique is rapid, non-destructive and applicable to solid and liquid materials.

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DISCUSSION

J.A. CZUBEK: Can you tell me whether the work described in your paper is merely a demonstration of the possibility of detection of sulphur by this technique, or have you actually found it to be the better (from the point of view of accuracy, economy, etc.) than such methods of sulphur detection as, for example, inelastic fast neutron scattering using a pulsed neutron source, gas chromatography using ionizing detectors or X-ray fluorescence analysis?

M.A. ESWARAN: As I mentioned in my presentation, we were concerned in this experiment with its nuclear physics aspects and our only purpose here was to draw the attention of potential users to the activation technique as a method of assaying sulphur based on the use of a relatively low-energy accelerator. The merits of this method in comparison with others are not discussed in the paper.

THE SIGNIFICANCE OF STOPPING POWERS IN EVALUATING AND USING CHARGED-PARTICLE EXPERIMENTS

I.V. MITCHELL, K.B. WINTERBON, F. BROWN
Chalk River Nuclear Laboratories,
Atomic Energy of Canada Limited,
Chalk River, Ontario, Canada

Abstract

THE SIGNIFICANCE OF STOPPING POWERS IN EVALUATING AND USING CHARGED-PARTICLE EXPERIMENTS.

Charged-particle analysis, including nuclear reactions, Rutherford scattering and atomic processes such as X-ray emission, is now widely used for analytical purposes. Such analyses are frequently difficult because the stopping powers of charged particles must be used to interpret them correctly. A well known example is the problem of calculating thick-target yields. In the case of a sample of uniform composition, the analyst can frequently avoid the problem by using a properly chosen comparative standard. For samples in which the depth distribution of the impurity is not known the problem may require a more sophisticated approach. Similar situations arise when charged-particle analysis is used to determine the depth profile of an impurity or to measure the stoichiometry of a compound. The importance of stopping powers in measuring nuclear life-times by Doppler-shift techniques has also been recognized. This paper will emphasize the difficulties in obtaining good stopping power data that are really applicable to the particular system being investigated; especially in work involving single crystals where the phenomena of ion channelling or blocking can play a dominant role. There is also the related question of whether the data themselves contain errors because they came from an experiment that required stopping power data for its interpretation; a simple example is determining a target thickness from energy loss measurements. Another problem comes from the fact that nuclear data usually come from experiments in nuclear physics which have a purpose quite unconnected with analysis; for example, experiments on the relative abundance of γ-rays rather than their absolute intensities. Finally, there is the question of how the compiler might simplify the problems of the user. Data might be presented in more useful or extensive forms. A central authority for collecting, revising and distributing stopping-power data would be most useful. Suitable experimental procedures could be suggested since although the problems might be well recognized by a nuclear physicist they may not be obvious to users in other disciplines. Another approach would be to provide, or list, computer programs that would enable the user to apply the data to his particular problem.

The question of how fast, charged particles lose their energy to the medium through which they are passing has attracted the attention of theorists and experimenters for many decades. Since the formulation by Bohr [1] and its advancement by Bethe [2] many refinements have appeared (see, for example, refs. [3] and [4]). Understanding of the energy loss processes for slow-moving particles has also advanced. The roles of nuclear (elastic) and electronic (inelastic) mechanisms have been made clearer [5]. Experimental evidence ([6,7] and many others) for atomic size and electronic structure effects in projectile and/or target atom has stimulated a more microscopic description of the inelastic collision events responsible for energy loss [ref. 8 and others reviewed in ref. 9]. Semi-empirical "universal" stopping powers have been generated and appear in tabulated or graphical form [10-13].

We wish to use this opportunity to point out that despite the work that has been done, the situation is still far from satisfactory and particularly so for the many users of such data. In almost any charged particle experiment involving targets of finite thickness, stopping power data enter in a fundamental way and yet these numbers are frequently the source of the greatest uncertainty in the analyses undertaken. Two different types of example will serve to illustrate the point, one relating to work of an applied (analytical) type, the other to an important class of basic nuclear physics experiments.

First, let us look at a helium ion backscattering analysis of thin films to map the depth profiles of implanted atoms in a near-surface layer. In this particular example [14], the helium ion energy is 2 MeV, which is typical for this type of analysis and the matrix is anodic aluminum oxide. Changes in the depth distribution of implanted Xe marker atoms are used to measure the transport of Al and O ions under applied voltage as the anodic oxide film is thickened. Part of a typical energy spectrum is shown in Fig. 1. The energy scale can be converted into depth scales, one appropriate to each atomic species. Two are shown in Fig. 1, viz. for Xe and Al in Al_2O_3. The ordinate scale may be converted through the scattering cross-section to an atomic concentration. It is important to recognize that both these conversions require a knowledge of the stopping power for the analyzing beam and the precision with which the depth profile of the Xe atoms can be specified is directly related to the accuracy of the stopping power data[1]. For scattering from Xe at depth x the emergent particles have an energy

$$E_1' \approx k^2\left[E_o - \int_o^x S(E)d\ell\right] - \int_{x\sec\theta}^{o} S(E)d\ell$$

(A related example is provided by the very careful depth profiling of boron in As-doped silicon through use of the $^{10}B(n,\alpha)^7Li$ reaction [15]. The stopping-power for alpha-particles in silicon is needed to transform the alpha-particle energy spectrum into a boron profile). Unique information can be gained from particle-scattering microanalysis about thin film alloy formation, film stoichiometry, diffusion, etc. [16] with a precision and sensitivity that is often superior to that available by any other method. The limitation in absolute precision, as we have stated, currently lies in the stopping powers to be used.

[1] Here and throughout we do not take up the related problems of energy straggling.

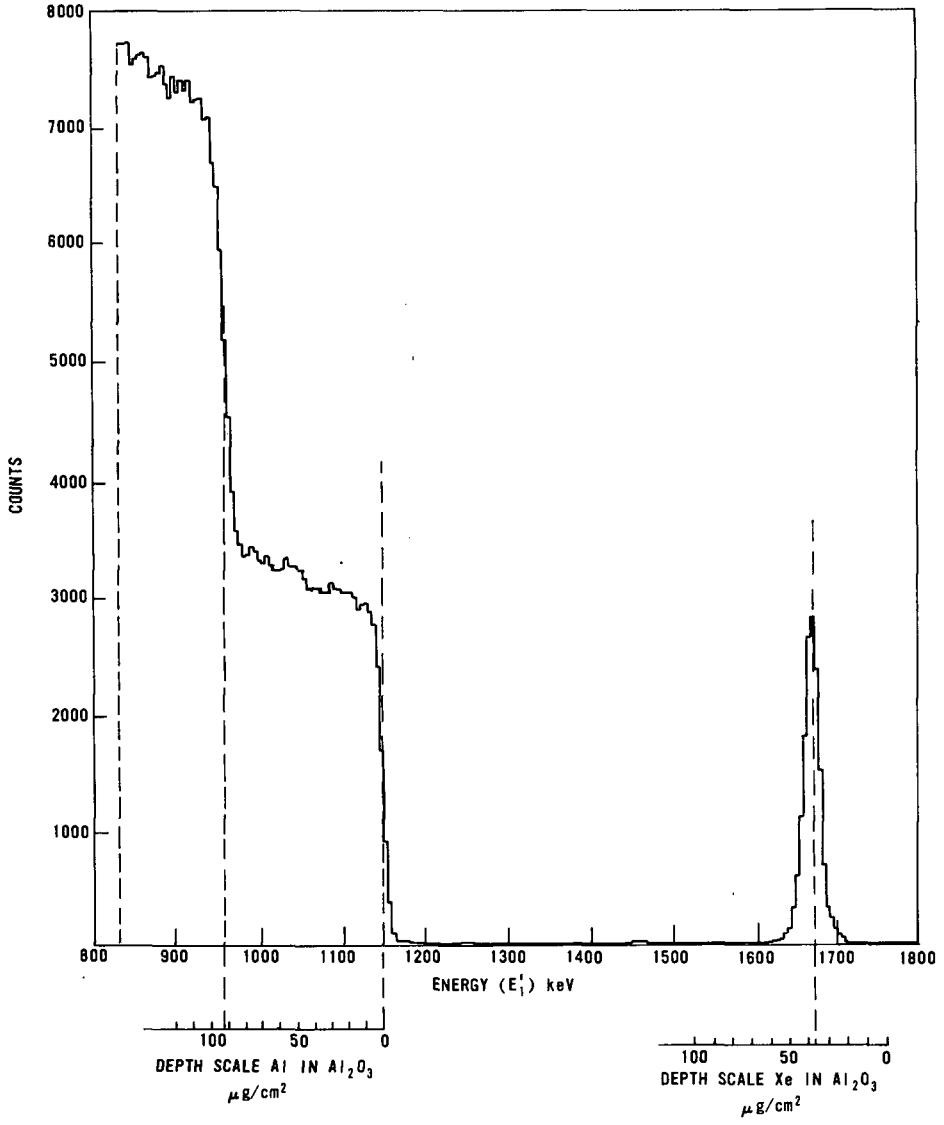

FIG.1. Spectrum from Xe buried in an oxide layer (95 $\mu g/cm^2$ thick) on the surface of Al; incident energy of the He^+ was 1975 keV.

Second, we cite two examples taken from an area of nuclear physics research and draw directly from experience at CRNL. The problem we are emphasizing is, however, common to all such programs. Thick target yields are related to the cross-section for the event of interest - here, heavy-ion induced Coulomb excitation - through an expression of the type

$$Y(E_o) = \int_o^R \sigma(E(x))\rho dx$$

where E_o is the incident ion energy, ρdx an element of the ion trajectory and R the range of the ion in the target. The ion energy E(x) at depth x is given by

$$E(x) = E_o - \int_o^x S(E)\rho dx$$

We see that any uncertainty in the stopping power is immediately felt as a corresponding uncertainty in the cross-section to be extracted.

In a related context Doppler shift attenuation measurements suffer from stopping power uncertainties in just the same way. A beam of fast heavy ions is used to Coulomb-excite target atoms. Recoil velocities of the order of 3-4% of c can be imparted. Gamma-decay of the excited state, during recoil, results in a maximum photon energy for forward (zero degree) emission of $E_\gamma = E_o\ (1 + \frac{v(t)}{c})$. The velocity distribution describes the slowing down process for the recoiling atom (t = 0 is the collision event) and is directly related to the stopping power for that recoil in the target:

$$v(t) = v(o) - \frac{1}{M} \int_o^t S(E\ (t_1))dt_1$$

neglecting changes in direction of motion of the recoiling nucleus. Analysis of the line shape of the Doppler-shifted spectrum yields a lifetime for the decaying state. An example is shown in Fig. 2 for 100 MeV ^{35}Cl excitation of ^{94}Mo [17]. The uncertainty in the extracted lifetime value $t_N = 4.5$ ps is thought to be > 10% and is attributed to the use of stopping powers which, although the best available, are not based on experiment. Many nuclear lifetimes suffer from precisely this lack of accurate stopping power data.

To the examples that we have mentioned we could add many others. One of the more important is charged particle activation analysis. Whenever a charged-particle-induced cross-section is required from thick target data the stopping power enters directly. As before, the reaction yield will be given by $Y = \int_o^R \sigma(E(x))\rho dx$, $E(x) = E_o - \int_o^x S(E)\rho dx$. This is as true for non-resonant as for resonant nuclear reactions and for non-nuclear (Coulomb excitation, particle-induced X-ray, etc.) as for nuclear cross-sections. The stopping powers are required

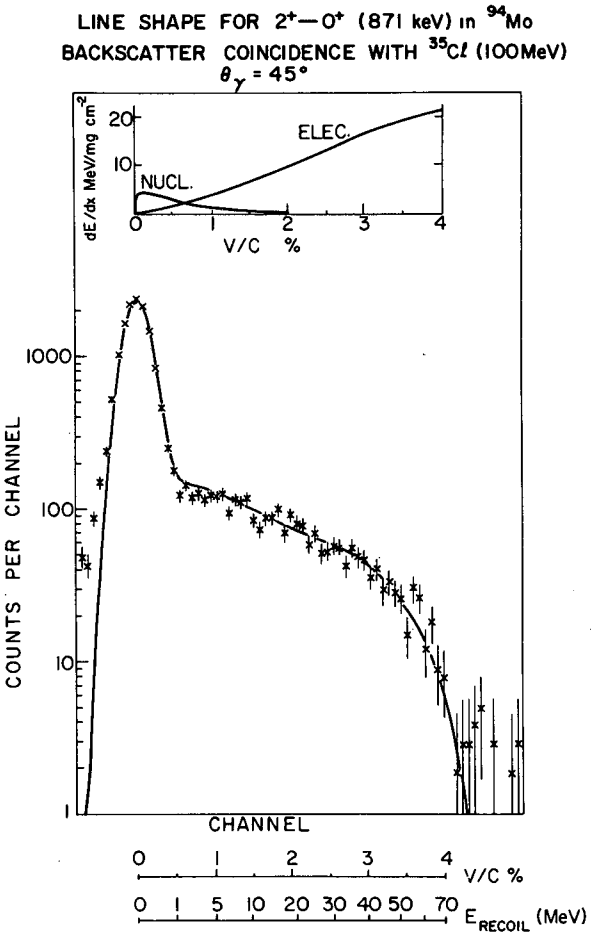

FIG.2. Illustration of the DSAM-method of measuring nuclear life-times. Assuming the stopping power curve shown in the inset for Mo-atoms in Mo, the best fit to the data points yields a mean life of τ_N= 4.5 ps for the 871-keV 2^+ level in ^{94}Mo. The accuracy would approach the 2% statistical value if the stopping powers were accurately known; from Ref. [17].

in profiling studies where the profile is to be extracted from the outgoing particle energy distribution. Similar remarks apply to charged particle ranges in any media. Stopping power inaccuracies affect the confidence with which ion ranges and energy loss rates may be predicted for applications as diverse as ion implantation into solids, simulation of radiation damage in reactor fuels and wall erosion rates for the plasma containment vessels of fusion reactors.

Some idea of the magnitude of the problem can be given by drawing from the foregoing examples. The energies at which

alpha-particle stopping-powers go through a maximum are of the order 0.5 to 2.0 MeV for most materials i.e. it is in this energy range that greatest depth sensitivity is reached. However, systematic data for this energy range, even for the more common elemental materials, are sparse. Further, while 10% errors are typically assigned, reported alpha-particle stopping powers at the same energy in Si differ from those in Al by as much as 30%. There is at present no published theoretical basis for such a large difference.

If anything, the situation is worse in the case of heavy ions (E/A < 10 MeV/amu). The highly regarded Northcliffe and Schilling tables "provide useful and reasonable estimates in the overwhelming number of cases where there are no data at all" but "it cannot be expected that all of these values will be of high accuracy" [13]. Isolated measurements undertaken for ^{35}Cl stopping powers in Ge and Ag depart from the tabulated values [13] by up to 10% in the range $10 < E_{Cl} < 100$ MeV [18]. (Absolute errors for these data are quoted to be less than 5%).

While it is patently clear that more data are needed there is also evidence that caution must be used in interpreting the stopping powers that are being measured. There are at least four areas of experimental and theoretical uncertainty.

1. Density Effect

It is not clear at the present time whether heavy ion energy loss in a gaseous and solid absorber of the same thickness will be different. The higher mean charge state for beams transmitted through stripper foils as compared with stripper gases may be understood in terms of the higher frequency of charge changing collisions in the solid [19]. It is also true that the ion beam must penetrate to a finite depth before charge state equilibrium is reached in a solid target (W. Brandt, private communication) and stopping powers measured in foils of such thickness can be expected to show anomalies. We are unaware of any published data on this subject. A group at ORNL has seen some evidence for this effect in transmission of oxygen beams (S. Datz, private communication).

2. Bragg's Law

This law states that the stopping cross-section of an assemblage of atoms is the sum of the stopping cross-sections of all component atoms e.g.

$$\epsilon_{mol}\ (M_xN_y) = x\epsilon_{at}\ (M) + y\ \epsilon_{at}\ (N)$$

Experimental stopping powers have been found to fall below the values predicted from the single element values [20, 21]; for anodic Al_2O_3 a 30% discrepancy has been reported for protons

$40 < Ep < 100$ keV [22] and for SiO_2 of ~ 10% [23] for alpha-particles of 1-2 MeV energy. In stopping-power studies, $0.3 < E_\alpha < 2.0$ MeV, Bourland et al. [24] showed that the values derived for stopping cross-sections in carbon were different for solid and gaseous hydrocarbon targets when Bragg's rule was applied. In a subsequent paper, Powers et al. [25] measured stopping powers in a variety of halogen-substituted hydrocarbon gases and concluded that using either two discrete values for the stopping cross-sections for carbon or two for hydrogen could satisfy the Bragg additivity rule to better than 3% in all 13 compounds studied. These results caution experimenter and evaluator alike. The conditions under which the stopping power can reveal information about chemical bonding in the absorber are obscure at the present time. No differences attributable to band structure have so far been observed in proton stopping powers (Ep ~ 4 MeV) in comparisons between semiconductors, metals and insulators [26], for example.

3. Shell Effects

At low velocities (E/A ~ 20 keV/amu) heavy ions will not be fully stripped and electron promotion mechanisms can cause inner-shell excitations during projectile-target atom encounters. Much more spectroscopic detail is embedded in the stopping power for this regime than in the case of fast completely stripped light ions such as protons and alpha-particles. Nevertheless, shell-like structure effects are seen in the stopping cross-section dependence on target atomic number for high energy protons [27] and for alpha-particles [28] in the energy range $0.3 < E_\alpha < 2.0$ MeV (see Fig. 3). These variations are accounted for in the modification by Rousseau et al. [29] of the theory of Lindhard and Winther [30], using Hartree-Fock wavefunctions for the target electron distributions rather than the structureless dress given by the Thomas-Fermi potential. The finer substructure in Fig. 4 is thought to be real [28]. Hence, tabulations based on smooth interpolations between data points should be used with caution.

Even more striking evidence has been reported for the oscillatory dependence of the (electronic) stopping power on projectile atomic number, Z_1 [6,7]. (Range measurements show corresponding results for the Z_2-dependence (J.L. Whitton, private communication)). In amorphous targets the stopping may vary by a factor two from a smooth, monotonic interpolation curve (E_{ion} ~ 500 keV). The effects can be enhanced by a factor 3 or more in single crystals (see later). This phenomenon has been shown to be a shell effect [8,9] perhaps best visualized as a periodic variation with Z in the size of the atom. For example, Ne, Cu, Ag, Au are small and have small stopping cross-sections whilst N, Ar, Kr, Xe are large and have large stopping cross-sections. This also implies that stopping powers in this regime may be sensitive to charge-state through ion "size".

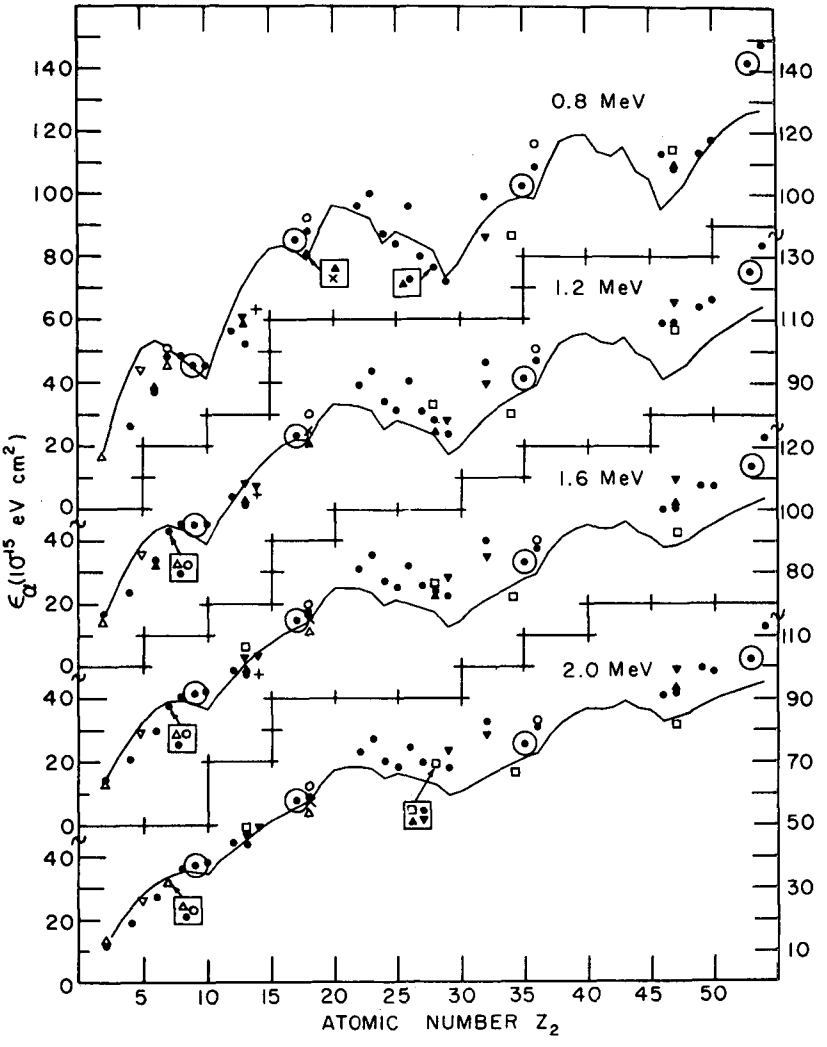

FIG. 3. Atomic stopping cross-sections ϵ_a as a function of the atomic number Z_2 of the stopping medium for alpha-particles at energies of 0.8, 1.2, 1.6 and 2.0 MeV. The solid curves are due to the theory of Lindhard and Winther [30] as modified by Rousseau et al. [29]. Solid circles represent measurements by Powers et al.,[25] and earlier work; the other symbols denote results by other experimenters. There is clear evidence for large structure and suggestive evidence for fine structure in ϵ_a versus Z_2.

4. Trajectories

In single crystal targets, successive collisions may be correlated, as in channeling [31] with a concomitant reduction in stopping and an increase in particle range. Up to a hundred-fold increase in the range has been seen for heavy ions.

In channeling, successive small-angle collisions steer the projectile away from any close collisions with target atoms so that in averaging the impact parameters used to define stopping, the small impact parameters have much-reduced weights. Channeled particle stopping powers depend on the channel and how well the particle was channeled in it. That fraction that is best-channeled spends least time in the electron-dense regions of the channel. For example, a stopping power of only one third the amorphous value has been observed for the best-channeled 4 MeV protons in single crystal Ge [26]. Stopping power measurements made in any solid, particularly metals films, in which large, coherent single crystal grains are often present, must allow for such contingencies. Conversely, users of stopping power data should recognize that the collision sequence for an energetic recoil in a single crystal or polycrystal target may not be the same as that in an amorphous target.

Hence stopping power data are most meaningful if the weighting of impact-parameters along the particle trajectory can be defined. In a backscattering measurement the particle has undergone at least one collision atypical of all others along the rest of its journey, namely the one that scattered it back. Inelastic energy losses, e.g. from inner-shell target electron ejection, will be high for that collision. In obtaining stopping by transmission measurements in thin targets one excludes those particles that have undergone large angle scattering, so one again ignores small impact parameters. For example, if an energy loss measurement is made only at very small scattering angles, much smaller than the width of the multiple-scattering peak, the measured "stopping power" is found to be proportional to thickness in the very thin target limit [32]. In the same way, the measured stopping of B in Si appears to depend on whether a range or an energy loss measurement has been made.

We have described a problem that affects a large community of users, in both fundamental and applied research. The purpose of this paper is to suggest that the continual evaluation, compilation and revision of stopping power data, in accessible form, is an undertaking proper to an international organization. When undertaken, it would benefit numerous users who, at present, find it difficult to judge the authority or applicability of the different tabulations and scattered data that are now available. Such a group would also coordinate the efforts of experimentalists working on stopping power measurements and could identify the more serious gaps in the data. Naturally, any compilation and evaluation would take advantage of the best present and future theoretical treatments.

It must be remembered that stopping power measurements or evaluations are sometimes subsidiary to the main purpose of a research project and it is not always obvious from the title

or abstract of a publication that stopping power information is contained in the text or references. A key-word survey, for example, would not reveal these data. A degree of publicity would perhaps be necessary in order to bring forth such 'hidden' data.

Another, perhaps more difficult problem, is to identify and evaluate errors in nuclear data, e.g. in reaction yields, that might be due to the use of erroneous stopping powers. Any future evaluation of such data should indicate the origin of the stopping powers used and the possible errors. In this way the user could revise his conclusions, if necessary, based on the best current information. Again, the cooperation of authors, in stating their source material explicity, will be helpful.

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DISCUSSION

G.A. KOLSTAD: You have exposed a rather fundamental and important problem in the field of nuclear data. I wonder if you have any suggestions as to what to do about it.

I.V. MITCHELL: I was hoping that the evaluators and compilers present might suggest what should be done.

M. LEDERER: Can the speaker or anyone else here suggest who might do an evaluation of stopping power data and semi-empirical calculations?

I.V. MITCHELL: Dr. Wei-Kan Chu at California Institute of Technology has told me that he is willing to give some help in this direction. This would be particularly in regard to proton and alpha-particle stopping powers. For heavy ions perhaps Dr. Schilling or Dr. Hans Betz could help.

Section XVI

APPLICATION-ORIENTED COMPILATIONS AND EVALUATIONS

Chairman

D.J. HOREN (USA)

CHARGED-PARTICLE NUCLEAR REACTIONS: COMPILATION, EVALUATION AND SYSTEMATICS OF EXCITATION FUNCTIONS

H. MÜNZEL, J. LANGE, K.A. KELLER, G. PFENNIG
Nuclear Chemistry Group, Institute for Radiochemistry,
Nuclear Research Centre, Karlsruhe,
Federal Republic of Germany

Abstract

CHARGED-PARTICLE NUCLEAR REACTIONS: COMPILATION, EVALUATION AND SYSTEMATICS OF EXCITATION FUNCTIONS.

In many fields of applied science and technology, there is a great need for cross-sections to be used for calculating the activity of nuclides produced by charged-particle bombardment. Therefore, many attempts are made in preparing compilations to ease the access to the data. In this paper, a survey is given on the present situation in respect to the available reference lists, the data compilations, the critical evaluation and the systematics of excitation functions.

Excitation functions for charged particle induced reactions are necessary to solve many different kinds of problems. To mention only some: Calculation of limits of detection in activation analysis, estimation of optimum irradiation conditions for isotope production or testing different assumptions in the theory of nuclear reactions. Practically, in all these cases it is not possible to predict which special excitation function will be needed in the near future. Therefore, an ideal compilation should contain the excitation functions for all possible target-projectile-combinations up to a projectile energy of about 100 MeV. In addition, it should contain very accurate data or should give at least some information about the errors to be expected. It is clear, of course, that such an ideal compilation could not be based only on experimental results. Therefore, one has to rely also on data obtained from a systematics of exitation functions.

The work which has to be done for preparing a comprehensive compilation can be divided into the following four steps which, of course, depend partly on each other:

I. Scanning the literature
II. Collecting experimental data, i.e. cross sections and excitation functions
III. Critical evaluation

IV. Development of the systematics of excitation functions.
In the following survey the situation in respect to the abovementioned aspects is discussed. But I will restrict myself to the discussion of cross sections for the formation of nuclides, whereas the formation of single states or angular distributions, etc. are not considered.

I. Scanning the literature

The first step is a comprehensive recherche of the available literature to collect the references for all publications. This is undoubtedly an important task, because all other compilations depend on this survey. In practice this means that one has to look up many journals etc. which is a very time consuming task. But, in despite of this fact, the situation is very satisfactory. This is due to McGowan and his collaborators who now publish every year such a compilation in form of a very extensive reference list [1].

The setup of this reference list is similar to the setup used in the well known CINDA-reference index. For each experimentally investigated reaction an entry is made. They are ordered according to the target nuclide. In the last two lists references are given also for theoretical investigations which deal with the analysis of experimental data. Each entry gives the type of reaction, the range of the projectile energy and some information about the kind of the data. In some cases the experimental data are given directly. The list of the keywords used shows that these indexes contain references of publications, which give results of all kinds of investigations about nuclear reactions, i.e. excitation functions, angular distributions, Coulomb excitation, total reaction cross sections, etc. Therefore, these reference lists have a considerable size. But due to the very clear arrangement they are still easy to handle.

Without going into details I would like to make some comments about these compilations. There is no doubt that it is very valuable to have such an easy access to the references for all publications one is interested in. This is the advantage of such a "charged particle CINDA". Other collections of literature references, like NSA or INIS, are less suitable. This is mainly due to the fact that they have a much wider scope and their keyword list is therefore not so adapted to the special purpose.

For the preparation of their compilations McGowan et al. scan many journals. Thirty of them are explicitly stated. Therefore, one can assume that these compilations cover this type of literature almost completely. But reports and conference abstracts are not scanned. In both

cases the data will nowadays most probably appear later in one of the covered journals, but with a considerable time lag. Therefore, it would have advantages to include references to this type of literature also.

II. Compilations of cross sections and excitation functions

A disadvantage, which is inherent to reference lists, is the fact that every user has to go back to the original publications to obtain the data. This task is not too time consuming, if i) the wanted cross sections are published, ii) the literature is readily available, and iii) the publication contains the relevant data in an easy accessible form.

Unfortunately, quite often at least one of these premises are not fulfilled. The difficulties are much larger, if the wanted excitation function was not experimentally determined, because in such cases one has to look up much more reactions to be able to estimate the cross sections by extra- or interpolation. Therefore it would be very helpful if a compilation about excitation functions existed.

The first broad collection of charged particle cross sections was published by Jarmie and Seagrave [2]. This collection contains reactions with H up to F. Later, Smith [3] prepared a compilation for reactions with targets ranging from Ne to Cr. McGowan et al. continued this type of work and published from 1964 till 1967 compilations for reactions with Mn, Fe, Co [4], Ni, Cu [5], Li, Be, B [6], C [7], and O, N [8]. In figure 1 an example is given to show the kind of presentation used. In the upper left the reaction, the reference and some additional information is given. In the example chosen, this information reads: "Synchrotron, stacked foil activation analysis, energy resolution 0.4 MeV. Tabular points obtained with data point reader from Fig. 1." The data are given in a table (lower left side) and a graph (right side). The compilations [2-8] contain also angular distributions etc. Therefore, the size is quite large. For instance, the C-compilation [7] has 241 pages. Unfortunately, this valuable work was presumably stopped because since 1967 no extensions or revisions have appeared.

Another compilation was prepared by Kunz and Schintlmeister [9] in the years from 1965 up to 1967 covering nuclear reactions with targets up to Z = 16 (Sulfur).

Few years ago we got for several reasons interested in the systematics of excitation functions of charged particle induced nuclear reactions in the low and medium energy range. For this purpose we needed many

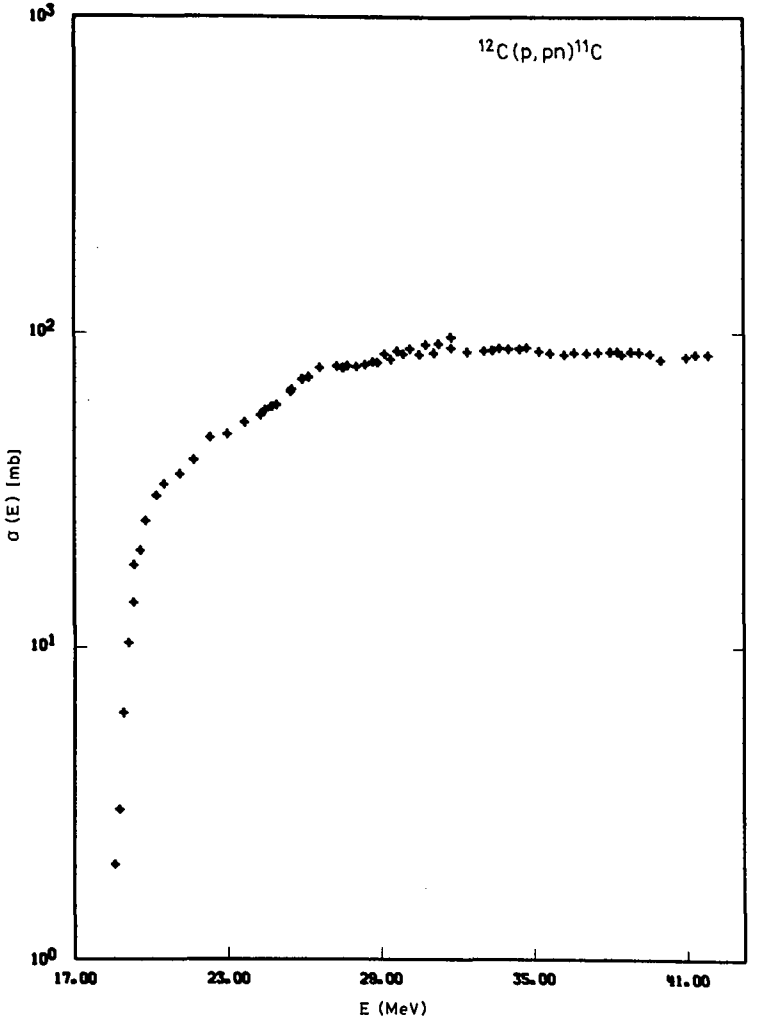

FIG. 1. Representation of the data in the compilation of cross-sections for charged-particle-induced reactions with carbon [7]. In the picture, the reaction $^{12}C(p,pn)^{11}C$ is shown. In the compilation, also numerical values and some additional information (see text) are given.

experimentally determined excitation functions. We have therefore collected many of them and published a compilation, which also included first results about the systematics [10]. This work was considerably extended in the last years, because we feeled that a comprehensive compilation for charged particle excitation functions was needed and the other abovementioned compilations were not continued. The result of this survey is now available as a volume I/5b of the Landolt-Börnstein, New Series [11]. This

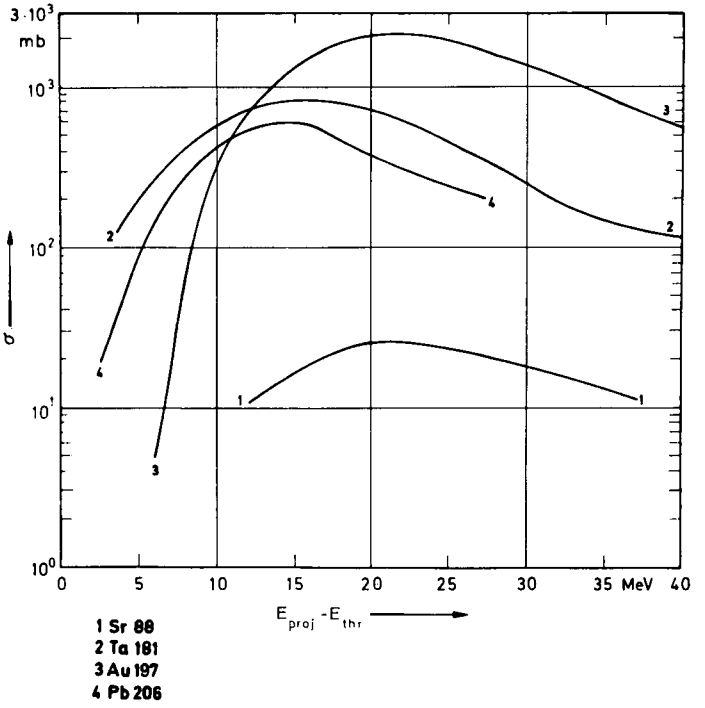

FIG.2. Representation of the excitation functions in the compilation prepared by Lange et al. [11]. In the example, the (p, 5n)-reactions with the listed target nuclides are shown.

compilation contains more than 1800 excitation functions for reactions of all kinds of target nuclei with charged particles, ranging from proton up to Ar-40.

In fig. 2 an example is given to illustrate the presentation used in this compilation. The excitation functions are given as smooth curves and no individual cross sections are shown. In general, the curves were obtained by reading off values from the curves drawn by the authors of the publications.

The ordinate gives the cross section in mb, whereas on the abscissa the difference between projectile energy and threshold energy is used. We are aware of the fact that due to this scale one has to read off in each case the threshold energy also given in a list of this compilation before the projectile energy can be calculated.

For this purpose the extensive Q-value table given in volume I/5a [12] can be used too. Nevertheless, we used this scale because it offers, as we think, two important advantages:

1) Large deviations between the excitation functions indicate the possibility that one of them may be in error. This assumption is based on the similarity of excitation functions for reactions of the same type in a limited range of the nucleon number A of the target nuclides. If there are no special reasons, like a relative large neutron excess or deficit of the target nucleus, then one should check carefully, if for instance wrong decay data were used for evaluating the cross sections. For such comparisons the systematics (see IV) may also prove to be very helpful.

2) Quite often, the wanted excitation function will not be found in the literature, i.e. is not experimentally determined yet. Then, an estimate can be made with the aid of the shown curves by extra- or interpolation. Again one makes use of the abovementioned similarity.

Fig. 2 which shows excitation functions for (p,5n)reactions can be used for an illustration: The curves 2 and 4 show the expected similarity and in addition agree approximately with the curves constructed with the aid of the systematics. The much lower cross section for the ^{88}Sr-reaction (curve 1) is due to the lower coulomb barrier of this target nuclide. Therefore the probability for proton emission is higher than in the reactions with ^{181}Ta and ^{206}Pu. Curve 3 deviates also considerably from the curves 2 and 4, for instance in respect to the position of the maximum, which is approximately 7 MeV higher than expected. This deviation is most probably due to the fact that a stacked foil target arrangement with an incident proton energy of 155 MeV was used to determine the cross sections at energies as low as 40 MeV. A slight error in the dE/dX-tables used to calculate the average energy in the individual foils or an error in the foil thickness determination may be the reason for the shift observed.

Although this compilation contains, as already mentioned, more than 1800 excitation functions, this does not mean that all available data are included. Some limitations had to be included in the preparation of the manuscript. So, for projectiles up to α-particles only an energy range of about 150 MeV was taken into account. In addition, starting from the threshold energy in general only the first 40 MeV, or if the maximum value of the curve would not be included in this range, the first 80 MeV are shown. But the most important limitation is that only such investigations were considered, where the energy interval of the measured excitation function exceeds 2 MeV. Accordingly, those publications in which

for a given reaction only one cross section is stated, are not contained in the compilation. This limitation was applied, because

1) the collection of this data would have delayed the preparation of the manuscript for quite a long time,
2) the inclusion of all data would have increased the size considerably, which would make the use cumbersome, and
3) the kind of presentation of the excitation function used now would not have been suitable for such an extension of the data.

Some of this limitations are due to the fact that we originally did not intend to prepare a compilation but were more interested in the systematics of the excitation functions.

All the compilations I mentioned up to now try to give a more or less comprehensive picture about the available data. The general intention was, as I believe, to serve all users and not only some specialists. Therefore, in one or another case it may be cumbersome to extract the wanted data from the available compilations. This situation can only be improved if a closer cooperation between users and compilers can be achieved.

III. Critical evaluation

The work necessary for a critical evaluation of the data can be divided into three steps:

a) collecting the data used for the calculation of the cross sections from the measured activities, for instance the decay data of the radionuclide,
b) comparison of these data with the presumably "best" data and, if necessary correction of the published cross sections.
c) Evaluating recommended values for the cross sections.

A survey of the available compilations shows that the situation in respect to all three steps of the evaluation is not satisfactory at all. In the CPX-reports, for instance, some additional information is given and in few cases corrections are applied. In our compilation no additional data are given. Recommended data are not available at all.

There is no doubt that the evaluation of recommended data is a very important task, but it is also a very time consuming job. The difficulties start already with the collection of the data, because, unfortunately, quite often one or more of the relevant values are missing or are at least not precisely stated in the publication. The subsequent comparison with the now available "best" values for monitor cross sections, decay

data, and so on, again is time consuming because quite often one first has to evaluate these "best" values itself. Authors and editors could ease the life of the evaluators very much if in future they could check more carefully if all relevant data are not only contained in the publications, but are also unambiguously stated. Of course, such a check can only be done effectively if the evaluators have stated clearly which data they need. Here, a closer cooperation between authors, editors and evaluators is necessary.

In the evaluation another difficulty comes up. Only in few cases excitation functions for a given reaction were determined experimentally two or more times. As a consequence, there are in general not enough experimental data for any kind of averaging procedure to obtain recommended values. Therefore, in these cases the only possibility to check the experimental data is to compare them with the excitation functions obtained from the systematics.

IV. Systematics

Already a rough estimate shows that up to now only few percent of the possible combinations of target nuclides, projectiles and reaction types are experimentally investigated. Even in the far future for many reactions only estimated or computed values will be available. Therefore, it is very important to develop and to test the applicability of such procedures.

A survey of the literature shows that in many publications experimentally determined cross sections are compared with calculated values. Quite often, the agreement found is remarkable good. Therefore, such calculation procedures should offer a possibility to obtain an estimate for unknown excitation functions. But the first difficulty which occurs is the selection of the most suitable procedure. There are several more or less distinct proposals available, which differ greatly in the sophistication of the theoretical assumptions and therefore also in the time needed for the calculations. We have compared the results given in about 30 publications to find out, which procedure would prove to be the best. This comparison did however show that the agreement between calculated and experimentally determined values depends only slightly on the complexity of the programs used. If this surprising result proves to be true, then that program should be used, which takes the shortest time for a reliable calculation of unknown excitation functions.

An even more important difficulty is the selection of the best value for the input data, like the level density parameter. A survey of

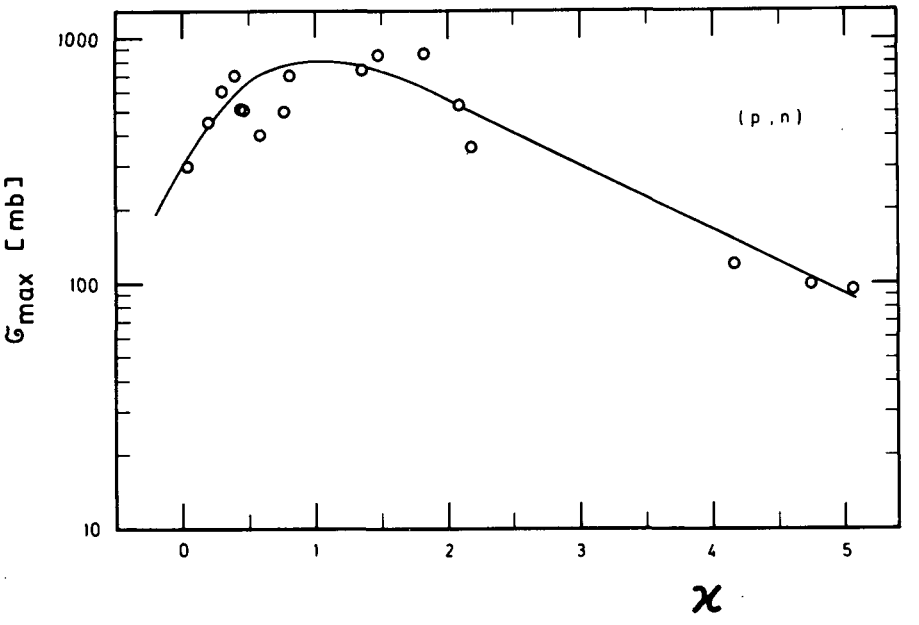

FIG.3. Dependence of the maximum cross-section on the parameter κ for (p,n)-reactions.

the literature shows that the actual values used differ considerably from each other. In many cases one or even more of the input parameters were treated as adjustable quantities, what of course improved the agreement between experimental and calculated values. Therefore, a selection of the best values for the input data and the most suitable calculation procedure has to be done, before reliable values of the cross sections can be calculated. We are just now working at such an analysis.

We have developed a semi-empirical procedure for estimating unknown excitation functions for p, d, ^{3}He and α-reactions, which can be applied without the use of a computer [10,13,14]. This systematics is based on the dependence of some characteristic values, like the position and height of the maximum of the excitation functions on a parameter $\varkappa$, which itself is a function of the binding energies of the last neutrons and protons and the coulomb barrier. Such a dependence is shown in fig. 3. With the aid of the values read off from those curves, which are visual fits to the points, many excitation functions were constructed. They will be published in volume I/5c of the Landolt-Börnstein [15], the manuscript is in preparation. In fig. 4 an example is given for the kind of presentation used in this case. Again, as energy scale the difference between

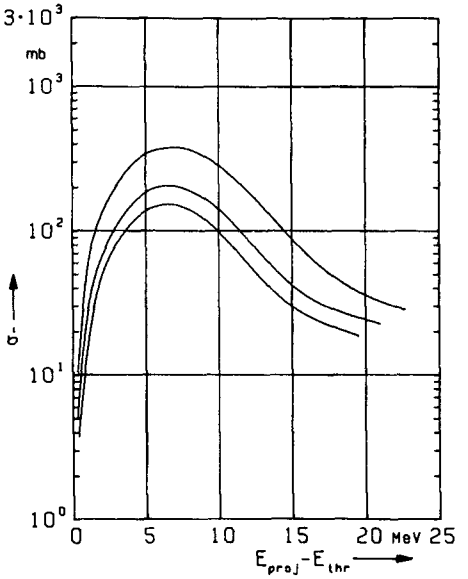

FIG.4. Representation of the excitation functions constructed with the aid of the systematics [15].

projectile energy and threshold energy is chosen. This volume will also contain thick target yields, which were calculated with the aid of these excitation functions. To use this compilation one has to read off $\varkappa$-values from a table and to interpolate between the given curves.

One important point in the application of such a systematics is the uncertainty of the estimated values. We therefore tested the accuracy in two ways. First, the deviations of the maximum cross sections from the predicted values were calculated for (p,xn)- and (α,xn)-reactions, with x ranging from 1 to 4. The mean values of the deviations obtained were less than 30 % [13]. Another test was performed to check the whole excitation function [16]. For this purpose thick target yields were calculated for (p,xn) and (α,xn) reactions, using experimental values and excitation functions obtained from systematics. The comparison shows that with one exception the deviations between corresponding thick target yields are less than a factor of 2. The exception is the reaction $^{54}Fe(\alpha,2n)^{56}Ni$, for which the value obtained with the aid of the systematics is by a factor of 20 too high. This is most probably due to the fact that ^{56}Ni is a double magic nucleus. We are just investigating, if the systematics could be improved by including shell effects in the calculation of $\varkappa$.

V. Summary

The situation can be summarized in the following way: The scanning of the literature is satisfactory. Probably one improvement could be the inclusion of reports and conference abstracts. In respect to the collection of cross sections, the situation is not so satisfactory. Although the newest compilation contains more than 1800 excitation functions, it comprises not all available data. Some limitations were mentioned above. The critical evaluation of the data is missing almost completely. An semi-empirical systematics is now available so that unknown excitation functions for several different types of reactions can be estimated. But, altogether, it still must be found out if the mentioned compilations satisfy the special requirements of the different users.

As this summary shows, there are many deficiencies or even gaps in this field of the compilation, evaluation and dissemination of cross section data for charged particle induced reactions. Many efforts have to be done to improve the situation. But this can be accomplished only if the conditions for the collection and evaluation of the data are improved too. I would like to mention four requirements only:

1) Considerable increase of the available man power in the various compiler groups.
2) Closer cooperation between the compiler groups.
3) Closer cooperation between authors, editors and compilers to make the data more readily accessible.
4) Closer cooperation between users and compilers to get a better understanding of the needs and requirements and, on the other hand, of the difficulties arising in the preparation of special compilations.

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DISCUSSION

D.J. HOREN (Chairman): Since it appears that basic researchers have little interest in making the types of measurements to which you have referred, do you feel that there is sufficient economic interest on the part of users to fund such work?

H. MÜNZEL: It is very difficult to answer this question. Our survey concerning data needs in activation analysis convinced us that data on excitation functions are lacking. If activation analysis is now considered to be a pressing need, then this is where the answer to your question lies. But I cannot say what will be the financial implications.

EVALUATION DES PARAMETRES DE SCHEMAS DE DESINTEGRATION

W. BAMBYNEK*, B. GRINBERG**, H. H. HANSEN*,
F. LAGOUTINE**, Y. LE GALLIC**, J. LEGRAND**,
E. DE ROOST*, A. SPERNOL*, A. H. WAPSTRA†, H. M. WEISS††

Abstract–Résumé

EVALUATION OF DECAY SCHEME PARAMETERS.

The use of radionuclides is partly a function of our knowledge of the parameters by which they are characterized. While in many cases an accuracy of 1 to 2% is sufficient, an accuracy of 0.1 to 0.2% is needed for certain applications. The latter accuracy can be achieved for many parameters, but only very few decay schemes are known with such an accuracy. This is the reason why the BCMN/IKO/PTB/LMRI group has decided to evaluate these schemes to a higher accuracy. The paper reports on the method of work and the evaluation rules adopted; some examples of evaluation are given.

EVALUATION DES PARAMETRES DE SCHEMAS DE DESINTEGRATION.

L'utilisation des radionucléides est en partie conditionnée par la connaissance des paramètres qui les caractérisent. Si dans de nombreux cas une précision de 1 à 2% est suffisante, pour certaines applications une précision de 0,1 à 0,2% est nécessaire. Cette dernière peut être atteinte pour de nombreux paramètres, mais très peu de schémas de désintégration sont connus avec une telle précision. C'est pourquoi le groupe BCMN/IKO/PTB/LMRI s'est proposé d'évaluer ces schémas avec une meilleure exactitude. Le mémoire rend compte de la méthode de travail et des règles d'évaluation adoptées, et donne quelques exemples d'évaluations.

INTRODUCTION

Depuis quelques années, des laboratoires européens essaient d'évaluer de manière approfondie les paramètres de schémas de désintégration des 150 radionucléides les plus importants. Le but de ce groupe est de fournir aux utilisateurs les meilleures valeurs ainsi que leurs incertitudes pour un grand nombre de paramètres. En effet souvent les utilisateurs regrettent de ne pas avoir à leur disposition une valeur recommandée et de devoir faire le choix eux-mêmes. Bien sûr il existe de nombreuses tables [1 - 4], mais malgré l'excellent travail effectué, souvent elles ne rendent pas le service attendu car la plupart sont des compilations. D'autre part, si dans quelques cas certaines tables recommandent une valeur, la nature de l'erreur qui lui est associée est rarement clairement définie; c'est pourquoi le groupe s'est attaché à définir d'une part une méthode de travail et d'autre part des règles d'évaluation de telle sorte que les incertitudes sur chacun des paramètres évalués soient déterminées de manière comparable.

Un autre but de ce travail est de mettre en évidence l'insuffisance de travaux expérimentaux pour la mesure de certains paramètres, et ainsi de susciter de nouvelles déterminations. Les quelques exemples que nous

* Bureau central des mesures nucléaires de l'Euratom, Geel, Belgique.
** CEA, LMRI, Centre d'études nucléaires de Saclay, France.
† Instituut voor Kernphysisch Onderzoek, Amsterdam, Pays-Bas.
†† Physikalisch-Technische Bundesanstalt, Brunswick, République fédérale d'Allemagne.

aurons l'occasion de traiter montreront que même pour des radionucléides qui semblent bien connus, certains paramètres fort utiles ne sont connus dans l'état actuel qu'avec une précision insuffisante.

1. MOTIVATIONS

L'utilisation des radionucléides est en partie conditionnée par la connaissance des paramètres qui les caractérisent. Si dans de nombreux cas une précision de 1 à 2% est suffisante, pour certaines applications une précision de 0,1 à 0,2% est nécessaire. Cette dernière peut être atteinte pour de nombreux paramètres, mais très peu de schémas de désintégration sont connus avec une telle précision. A cette occasion quelques exemples peuvent être cités: l'américium-241 est un radionucléide très utilisé; or il semble subsister quelques doutes sur sa période. En effet pendant de nombreuses années la valeur adoptée était de 458 ans environ; elle était déduite des travaux de Hall et Markin [5] et de Wallmann et coll. [6]. Une dizaine d'années plus tard, en s'appuyant sur les travaux de Oetting et Gunn [7], Stone et Hulet [8] et Brown et Propst [9], on peut être tenté d'adopter une période de 433 ans environ soit un écart de 6% alors que les différents auteurs revendiquent des précisions de 0,1 à 0,5%. La lecture attentive des différentes publications ne permet pas de déceler des erreurs qui permettraient de justifier les écarts observés. Si la période du ^{241}Am doit être connue avec précision, de nouvelles mesures sont nécessaires.

Un autre exemple concerne l'uranium-234, pour lequel une évaluation approfondie a conduit le Bureau central des mesures nucléaires (BCMN, Euratom, Geel) à entreprendre de nouvelles mesures de la période de ce radionucléide. La nouvelle valeur a entraîné une diminution de 2% de la section efficace de fission de l'uranium-235. Toujours dans le domaine des périodes, des travaux récents du National Bureau of Standards (USA) ont permis de déterminer avec précision celle du ^{63}Ni, alors que les valeurs publiées étaient séparées de près de 50%.

Des exemples similaires pourraient être cités pour d'autres paramètres tels que le rapport β^+/CE pour le ^{22}Na, l'intensité des photons de 59 keV du ^{241}Am, le coefficient de conversion interne de la transition de 14 keV du ^{57}Fe, de nombreux rendements de fluorescence pour la couche K, etc.

Il ne faut pas conclure à partir de ces quelques exemples que l'évaluateur met toujours en évidence une insuffisance des travaux expérimentaux; au contraire, très souvent il apparaît que les paramètres sont connus avec une précision suffisante pour l'usage souhaité. Le travail d'évaluation permet alors de définir une valeur et l'incertitude associée. Très souvent, lorsque les travaux sont nombreux, et les méthodes employées différentes, l'incertitude sur le paramètre évalué est bien meilleure que celle revendiquée par chacun des auteurs.

2. METHODE DE TRAVAIL

L'évaluation est effectuée, selon des règles clairement définies, par deux physiciens de deux laboratoires différents, qui ont eux-mêmes effectué quelques travaux sur les radionucléides étudiés. Ces conditions

sont apparues très importantes car il est difficile d'avoir un point de vue critique et de réévaluer des erreurs pour des techniques plus ou moins bien connues. Par exemple, pour la période du ^{241}Am, des précisions de 0,1 ou 0,2% n'auraient jamais été acceptées.

L'exposé des résultats comprend deux parties; la première est, pour chaque radionucléide, une étude approfondie avec des commentaires sur les publications examinées et tous les détails des évaluations. La seconde partie se présentera sous forme d'un tableau où seront reportés tous les résultats. Afin de rendre le maximum de services, il a été décidé de présenter d'une part l'évaluation des principaux paramètres: période, énergies, intensités des différents rayonnements, coefficients de conversion interne etc. et d'autre part une compilation de nombreux autres paramètres: sections efficaces de production, spins, parités, durée de vie des niveaux, valeurs de log ft etc.

Enfin, pour aider les utilisateurs, des spectres des différents rayonnements sont joints (α, e^-, γ et X selon le cas) ainsi qu'une table de décroissance calculée à partir de la période évaluée. Deux exemples de ces rapports sont disponibles, ils concernent le ^{58}Co [10] et le ^{51}Cr [11].

3. REGLES D'EVALUATION

Le détail des définitions et des règles est décrit dans l'introduction générale au travail entrepris [12]. Le problème le plus délicat a été de définir l'incertitude finale associée à la valeur du paramètre évalué. Finalement il a été recommandé que cette incertitude corresponde le plus possible à un intervalle de confiance de 68%, l'incertitude ayant dans le cas général la forme suivante:

$$\Delta = \frac{t\,s}{\sqrt{n}} + \frac{\delta}{3}$$

t: facteur de Student pour un intervalle de confiance de 68%
s: écart type
δ: erreurs systématiques

$$\delta = \sum \delta_i$$

$$\text{ou } \delta = \sqrt{\sum \delta_i^2}$$

La dernière relation n'est utilisée que si les erreurs systématiques sont nombreuses et du même ordre de grandeur.

Cette définition de l'incertitude est un compromis. Si elle est multipliée par 3, on obtient une valeur qui correspond pratiquement à un intervalle de confiance de 99,7%.

Les différentes étapes de l'évaluation sont les suivantes:
- compilation
- correction éventuelle des résultats

- détermination de nouvelles erreurs et rejet des résultats douteux
- estimation de la «meilleure» valeur et de son incertitude.

Toutes les publications, et, lorsque c'est nécessaire et possible, toutes les informations fournies par les auteurs, sont prises en considération. Les résultats publiés sont éventuellement corrigés si de meilleures valeurs ou de nouvelles formules sont connues des évaluateurs. C'est le cas par exemple des rendements de fluorescence qui conduisent à corriger certaines valeurs de coefficients de conversion interne, de l'énergie de certaines transitions α ou γ utilisées comme référence et qui ont été mesurées récemment avec des précisions bien supérieures à celles utilisées par les auteurs, etc.

Des incertitudes sont affectées à chaque valeur selon les règles fixées si celles données par les auteurs ne sont pas compatibles avec nos définitions. Chaque fois qu'un résultat est modifié, toutes les justifications sont fournies. Ensuite les résultats pour lesquels il n'a pas été possible de déterminer une erreur sont rejetés ainsi que ceux dont l'incertitude est trois fois plus grande que celle du résultat le plus précis. Cette règle n'est pas appliquée lorsqu'il n'y a qu'une valeur à laquelle il n'a pas été possible d'associer une erreur; dans ce cas elle est conservée et citée avec toutes les restrictions nécessaires.

Pour la détermination de la meilleure valeur et de son incertitude associée plusieurs cas peuvent être distingués qui sont repris en détail dans le texte d'introduction [12]. En effet, bien que les séparations puissent paraître arbitraires, les situations suivantes ont été imaginées et les calculs de la meilleure valeur et de l'incertitude ont été définis pour chacune d'elles:

- un seul auteur utilisant une seule méthode
- plusieurs auteurs utilisant la même méthode
- un ou plusieurs auteurs utilisant des méthodes différentes.

Cette dernière situation est la plus confortable puisqu'elle permet de minimiser les erreurs systématiques, la meilleure valeur est alors la moyenne pondérée. L'incertitude correspond à l'erreur interne; la validité de cette dernière est testée par le calcul de l'erreur externe qui doit lui être inférieure ou au plus égale. Si ce n'est pas le cas une nouvelle évaluation de l'incertitude sur chacun des résultats publiés est faite.

L'expérience effectuée sur le ^{51}Cr, le ^{58}Co et le ^{65}Zn a montré qu'il était rare de se trouver dans cette situation idéale, mais l'objectif de l'évaluation, quel que soit le cas dans lequel il se trouvera, sera de fournir une valeur avec une erreur associée qui corresponde le plus possible à un intervalle de confiance de 68%. Lorsqu'il aura été difficile, et même quelquefois impossible, de déterminer la meilleure valeur l'évaluateur proposera de nouvelles mesures.

4. EXEMPLES D'EVALUATIONS

Trois évaluations sont maintenant terminées; elles concernent le ^{51}Cr, le ^{58}Co et le ^{65}Zn. Les rapports sur les deux premiers radionucléides sont disponibles. D'autres évaluations sont en cours; elles concernent le ^{37}A et le ^{60}Co.

Pour le ^{51}Cr, la plupart des paramètres utiles sont connus avec une précision suffisante. Il serait cependant nécessaire que l'intensité de la

transition γ de 320 keV soit déterminée avec une meilleure exactitude, ce qui semble facilement accessible compte tenu des techniques disponibles.

Pour le ^{58}Co, le principal problème concernait sa période; en effet l'analyse des quelques publications a montré que les auteurs avaient mal tenu compte des impuretés souvent présentes. En effet, selon le mode de production, des quantités plus ou moins importantes de ^{57}Co et de ^{60}Co peuvent être formées. De nouvelles mesures ont été faites tant au BCMN qu'au LMRI; un excellent accord a été obtenu, ce qui a conduit les évaluateurs à recommander la valeur de 70,78 ± 0,10 jours, qui est sensiblement plus faible que celle généralement adoptée: 71,3 ± 0,2 jours. Le ^{58}Co est très utilisé pour les mesures de flux neutroniques et l'écart observé sur sa période entraînait des erreurs non négligeables. Pour tous les autres paramètres, les incertitudes obtenues par l'évaluation sont compatibles avec les différentes utilisations du ^{58}Co. De nouvelles mesures ne sont pas utiles.

Pour le ^{65}Zn, l'évaluation et les mesures complémentaires effectuées par le BCMN et le PTB montrent que tous les rapports de branchement sont connus avec une incertitude de 0,1%.

CONCLUSION

Le grand nombre de radionucléides à examiner implique des délais relativement importants. En effet, pour que ce travail soit correctement effectué, il est nécessaire d'y consacrer beaucoup de temps, ce qui en limite de façon importante le «rendement». Cependant s'il était possible de renforcer l'équipe existante par la participation d'autres laboratoires, les 150 radionucléides les plus utilisés pourraient être examinés en une dizaine d'années. Cette échéance peut paraître lointaine, mais il faut remarquer qu'après une évaluation il ne devrait plus être nécessaire de recommencer avant de nombreuses années à moins qu'entre-temps apparaissent des besoins en données plus précises.

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DISCUSSION

A.H. WAPSTRA: I would like to point out that things are wrong not only if the "external" error is much larger than the "internal" one, but also if the reverse is true. This, too, points to the presence of systematic components in the errors.

J. LEGRAND: I agree.

G.A. KOLSTAD: Among the three functions of a "data group", i.e. compilation, dissemination and evaluation, evaluation has until now had a lower priority than compilation and dissemination. Now that much more experimental data, and also computerized systems of data assimilation, are available, do you feel that the time has come to upgrade the level and quantity of evaluation, even if this means slowing down the compilation of new experimental data or increasing the turn-around time for putting out an evaluated mass chain, or similar evaluated set? Secondly, should the evaluation work be carried out at the data centres or by scientists working at their home laboratories with occasional interaction with a central co-ordinating group?

J. LEGRAND: It is true that with the large number of data now available, the time would seem to have come for more thorough evaluation. We would propose that this evaluation should be made by scientists who have personal experience of the parameters to be determined.

J.A. CZUBEK: What is the reason, from the point of view of statistics, for applying the formula $\Delta = t \cdot s/\sqrt{n} + \delta/3$ in your paper? It seems very strange to combine these two quite different types of error (by definition one of these is statistical and the other is constant), and to perform an algebraic summation (for errors a geometric summation is generally used). This question can be quite important if one of these errors is large.

J. LEGRAND: It is true that the random and systematic errors are of different nature and that the idea of summing them may seem strange. However, the user wants to have an uncertainty associated with each result and for this reason a compromise was necessary.

Y. LE GALLIC: I should like to add a few words to what Mr. Legrand has just said. Users want to know what uncertainty attaches to the data which they use. The problem is the same as that which exists for radioactive standards. Of course, the combination of random and systematic errors is subject to discussion. In the present case, the formulation adopted represents a convention by means of which the results presented can be brought into line while the user is provided with data which can be used directly.

B. ROSE: You stated you believe that the evaluation of the 150 most important nuclides would take about ten years. Given support from other laboratories, could you estimate how many man-years would be needed for this enterprise?

J. LEGRAND: On the basis of our experience, the average time required for the evaluation of one isotope is about six months (for two scientists).

C. WEITKAMP: The critical evaluation and generation of best, or recommended, values for properties of excited nuclear states has so far been limited to few and relatively isolated quantities, such as half-lives. How far do you plan to go with respect to the data to be evaluated? Will you, for example, try to evaluate γ-ray energies and intensities, of which many hundreds are sometimes published in a single paper? And if so, do you

expect that the data for the evaluation will be readily available. Is the six man-month estimate per nuclide still realistic?

J. LEGRAND: Not all the data are evaluated – only the most important ones (e.g. half-life, energies and intensities of the different radiations, internal conversion coefficient). However, the energies and intensities of all excited levels are not evaluated. It is for this reason that the average time for an evaluation can be put at six man-months.

D.J. HOREN (Chairman): I would like to congratulate the EURATOM people for carrying on this kind of work, because I think it is very necessary. Somebody, somewhere, has to do some precise work to meet established standards. However, if we are talking in terms of time, we might say that they are spending, if we take the numbers given by Mr. Legrand, roughly six man-months to get good, evaluated precision on, I would say, 20 numbers. If we were to try to evaluate every single datum generated in nuclear physics to a precision of better than 1%, it would be an impossible task. We therefore have to adopt some practical approach to the problem, and this is what is done in the various types of compilations. An average Data Sheet compilation takes something of the order of three to four man-months, sometimes longer. This includes examining around 10 000 numbers but certainly never in the detail with which the EURATOM type of evaluation is concerned. If in preparing a Data Sheet one were also to try and do experiments to determine the basis of inconsistencies in every number, one would of course never reach the publication stage. So I think we have to develop a perspective, but within this perspective there is certainly an urgent need for some very precise data. The other requirement is to force this type of need onto the experimentalists in that they will produce better papers.

NUCLEAR DATA COMPILATIONS OF UTILITY IN MEDICAL AND BIOLOGICAL APPLICATIONS

L.T. DILLMAN,
Ohio Wesleyan University,
Delaware, Ohio,
United States of America

W.S. SNYDER, M.R. FORD,
Health Physics Division,
Oak Ridge National Laboratory,
Oak Ridge, Tenn.
United States of America

Abstract

NUCLEAR DATA COMPILATIONS OF UTILITY IN MEDICAL AND BIOLOGICAL APPLICATIONS.

Detailed knowledge concerning the energies and intensities of all the radiations emitted as a result of a radioactive decay process is necessary in making radiation dose calculations. The principal author has previously published a compilation of nuclear data of this type for nuclides of interest in medical and biological applications. This paper reports on significant extensions of that work. We have recently calculated the bremsstrahlung spectrum associated with beta particles and with monoenergetic conversion electrons. Although only the inner bremsstrahlung is an invariable part of the radioactive decay process, we have calculated in addition the external bremsstrahlung for absorption in air, muscle, adipose tissue and skeleton. As a further aid to persons involved in internal radiation dose calculations, we propose to use decay scheme data as input and calculate specific effective energies (SEE) for many radionuclides. Specific effective energy is the energy absorbed per gram of target organ Y for each disintegration of the radionuclide in source organ X multiplied by qualifying factors associated with linear energy transfer values of the radiation. A computer program has been written which calculates SEE values for a large matrix of target and source organs. For each radiation, in addition to the intensity and energy, this program interpolates from a table to find the fraction of the energy absorbed in organ Y for a source uniformly distributed in organ X. These absorbed fractions have been determined by Monte-Carlo techniques using a non-homogeneous phantom representing standard man. The quality factors used in all calculations are shown explicitly so that the SEE tables may be used in calculations of absorbed dose as well as dose equivalent. All of the nuclear data and the static biological data are provided in the SEE tables, with the kinetic biological data which the user must provide being contained in the residence time of the radionuclide in the source organ.

Introduction

Nuclear data compilations such as the Journal, Nuclear Data, or the Table of Isotopes by Lederer et. al. do not list the energies and intensities of atomic transitions which occur as a result of radioactive decay. Nor do they list the average energy of beta particles in a given beta branch. However, information of this kind is required in dosimetry calculations. The principal author has previously published [1] detailed decay scheme data which included average beta energies and energies and intensities of atomic transitions. This compilation has been used by the Medical Internal Radiation Dose (MIRD) Committee of the Society of Nuclear Medicine and by others as the data base for radiation dose estimation. However, in medical and other biological applications the basic nuclear data is merely a beginning point for making radiation dose estimations. A larger fraction of the total effort in making radiation dose

estimations often comes after the basic nuclear data is determined. In view of this we have attempted to bridge the gap between the purely physical data on the one hand and the purely biological data on the other hand; our goal has been to put data in a form more directly and easily usable in medical and biological applications.

Lack of appreciation of the full significance of detailed information on basic nuclear processes can sometimes lead one to unwarranted conclusions. For example, the radiation dose due to bremsstrahlung photons accompanying beta decay cannot be neglected in some cases, particularly in estimating genetic dose. Hence a calculation of the bremsstrahlung spectrum has been incorporated recently as part of the decay scheme data. The methods used in this calculation and typical results will be discussed in this paper.

Presently the MIRD Committee is combining basic nuclear decay scheme data with compilations concerned with physical and biological aspects of the manner in which the various radiations interact with the human body to estimate dose from administered radiopharmaceuticals. This has led us to a concept we call specific effective energy and which we also discuss in this paper.

Decay Data and Nuclear Parameters

As a basis for the discussion of the significant extensions of and uses for the decay scheme data we have previously published we shall summarize the nature and extent of the previously published data. Determination of average beta energies is quite tedious and requires detailed information concerning the physics of this process. In the case of allowed beta decay, exact closed mathematical form calculation of the average energy of beta particles from the Fermi theory of beta decay is not possible and numerical methods of integration must be used. A high speed computer becomes an absolute necessity, particularly when one realizes that the theory of unique forbidden beta decay is considerably more complex than for allowed beta decay. The computer code developed by the senior author uses a library of data on such quantities as fluorescence yields, relative x-ray and Auger electron yields, theoretical internal conversion coefficients, electron binding energies, etc., in order to calculate the energies and intensities of x-rays, Auger electrons and internal conversion electrons. In the case of electron capture, theory is used to determine the distribution of initial vacancies in the various atomic shells.

Thus these data included average beta energies, x-ray intensities and energies, and Auger and internal conversion electron intensities and energies as well as data on nuclear transformations which may be found in standard nuclear data compilations. These data agree quite well with a similar publication by M. J. Martin and P. H. Blichert-Toft [2]. The break-down of x-ray and Auger electron data into the various groups is more complete in our publications than in [2]. A new publication by the MIRD Committee giving decay information of this type is in preparation. It will contain revised and updated data on the 54 radionuclides previously published [1], and will contain new data on an additional 65 radionuclides. In addition, information of this type for more than 400 radionuclides is on file at the Information Center for Internal Exposure, Oak Ridge National Laboratory, Oak Ridge, Tennessee, U.S.A. This information is being prepared for updating International Commission on Radiological Protection (ICRP) Publication II [3].

Bremsstrahlung Radiation

Bremsstrahlung radiation always accompanies beta emission or the emission of monoenergetic electrons. In dosimetry calculations it is often dismissed as representing a negligibly small fraction of the emitted radiation. Indeed, even for the rare case of a beta emitter with endpoint energy in the neighborhood of 10 MeV the total bremsstrahlung yield is not expected to exceed a few percent of the average beta energy if absorption of the betas takes place in tissue or other low Z material. However, there are cases where the radiation dose due to bremsstrahlung cannot be neglected. This is true, for example, in the case of immersion of persons in a pure or almost pure beta emitting radioactive cloud. The genetic radiation dose will be due almost entirely to bremsstrahlung radiation since the betas will be absorbed in the skin unless extremely energetic. A case in point is the concern with the environmental buildup of radioactive krypton-85 which is almost a pure beta emitter. There is a low intensity gamma-ray involved in the krypton-85 decay but approximately 20% of the genetic dose is predicted to be due to bremsstrahlung radiation. Another example where bremsstrahlung radiation may be important in dosimetry calculations is the case of a pure beta emitter confined primarily to the contents of the gastrointestinal tract.

Thus we have recently incorporated a calculation of the bremsstrahlung spectrum as a part of the decay scheme computer program mentioned in the introduction. There are two types of bremsstrahlung radiation. Inner bremsstrahlung radiation is created during the emission of the beta particle from the nucleus. Outer or external bremsstrahlung radiation is created after the beta particle is ejected from the nucleus and hence its characteristics are determined by the absorbing medium in which the radioactive particles are immersed. Although only the inner bremsstrahlung is an invariable part of the radioactive decay process we have calculated in addition the external bremsstrahlung for air, muscle tissue, adipose tissue and skeleton since these are the media of primary concern in medical and biological applications. These bremsstrahlung calculations have been made using the continuous-slowing-down approximation. This approximation assumes that the rate of energy loss of the beta particle along the entire track of the particle is always equal to the mean rate of energy loss. Actually the rate of energy loss fluctuates about the mean value. Also the continuous-slowing-down approximation does not include radiation from secondary electrons. However, their contribution would be quite small due to their predominantly very low energy. Berger and Seltzer [4] have compared bremsstrahlung radiation yields using the continuous-slowing-down approximation with more accurate methods which take into account the discontinuous nature of collision losses and bremsstrahlung losses as well as the production of secondary electrons. Their discussion ends with the assertion that the continuous-slowing-down approximation is quite serviceable and gives rise to errors of no more than a few percent.

The external bremsstrahlung spectral distribution has been calculated according to methods developed by Liden and Starfelt [5]. The spectral distribution of internal bremsstrahlung was calculated according to the theory developed in Knipp and Uhlenbeck [6]. In implementing the theory in both cases we have relied upon the detailed and comprehensive review of bremsstrahlung cross section formulas by Kock and Motz [7]. A detailed account of all formulas used and methods of implementation of the required numerical procedures is being prepared for publication. It is sufficient to say here that we believe the principal factor limiting the accuracy of the computed results is the accuracy of the cross section data base. The total bremsstrahlung spectra (inner plus outer) associated with

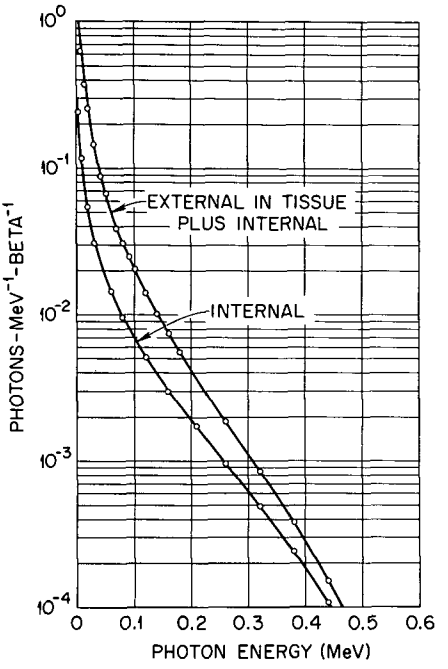

FIG.1. The internal and total (internal + external in a tissue medium) bremsstrahlung spectrum associated with the beta decay of krypton-85.

the beta decay of krypton-85 for a muscle tissue absorbing medium is shown in Fig. 1. Also shown for reference is the inner bremsstrahlung spectrum. The ratios of total bremsstrahlung spectrum of air, bone and adipose tissue to muscle tissue remain practically constant at 1.05, 1.24, and 0.90, respectively, up to an energy of 0.10 MeV. At higher energies these ratios change gradually to 1.009, 1.053 and 0.982, respectively, at an energy of 0.60 MeV. The general features of these spectra are representative of the spectra we have calculated for a number of different radionuclides.

For ease in using these data in dosimetry calculations we have simulated the continuous bremsstrahlung spectrum with a histogram where the abcissa consists of a number of discrete energy intervals. The number of energy intervals used increases approximately logarithmically with the beta end-point energy. Fifty is a typical number of intervals. The ordinate is the total number of bremsstrahlung photons in the energy intervals. Narrower energy intervals are chosen at lower photon energies because the bremsstrahlung photon yield increases quite rapidly at lower energies. This histogram method allows one to approximate the continuous bremsstrahlung spectrum with a series of photon intensities at a number of discrete photon energies, the discrete photon energies being the mid-points of the energy intervals. Each of these discrete photon energies can then be treated in a manner identical with gamma-ray photons which are a part of the normal decay scheme data. The only exception to this rule occurs for immersion in a radioactive beta emitting cloud. The bremsstrahlung produced in the skin from betas which are stopped in the skin differs from the bremsstrahlung associated with betas stopped in the surrounding air not only

because the absorbing media are different but also because the spectrum of betas which enter the skin differs from the equilibrium beta spectrum in air. A separate histogram is computed for this case.

Specific Effective Energy Tables

Even with complete nuclear decay scheme data, including information on bremsstrahlung radiation and atomic transitions which are not found in most nuclear data compilations, the task of accurate radiation dose calculations is formidable. This is particularly true in internal dosimetry where the radioactive source may be distributed in several internal body organs and the radiation dose to these same and other organs may be in question. Suppose, for example, one wishes to calculate the genetic dose, whole body dose, and the dose received by each of the source organs when the source is distributed in varying degrees in five organs. Then for each radiation emitted in the decay and for each source organ one must calculate the genetic dose, whole body dose, and the dose to every source organ. If there are, say, 100 penetrating radiations this will involve 100 x 7 or 700 calculations for each of the source organs or 3500 calculations in all. One-hundred penetrating radiations is not atypical although at first glance it might seem so. Taking into account the bremsstrahlung spectrum by the methods described above accounts for 50 penetrating radiations in an average case. Furthermore, the state of the art in gamma-ray spectroscopy is such that many radionuclides have been found to contain a plethora of low intensity gamma-rays. As an aggregate these may be quite significant in dosimetry calculations although any particular gamma-ray alone makes a small contribution to the overall radiation dose. Each calculation requires knowledge of the fraction of the energy emitted in source organ X which is absorbed in target organ Y, the absorbed fraction. Absorbed fractions have been calculated recently [8] by Monte Carlo techniques using a nonhomogeneous phantom representing standard man [3]. These calculations take account of the size, shape, density, elemental composition and relative positions of all of the major organs and parts of standard man as well as accounting accurately for the physical processes by which gamma radiation interacts with matter. For small target organs it is sometimes the case that the statistical accuracy of the Monte Carlo estimate of absorbed fractions is too poor to be meaningful. In such cases absorbed fractions have been estimated by alternate techniques which involve buildup factors and other principles which have been discussed by Snyder [9]. These absorbed fractions are tabulated at 12 energy values ranging from 0.01 to 4.0 MeV and thus an interpolation must usually be performed to obtain the absorbed fraction at the desired energy. The prospect of 3500 interpolations and other simple arithmetic operations adds up to a most tedious and laborious task.

As an aid to persons involved in internal dosimetry calculations we propose to publish tables of specific effective energies (SEE) for a large number of radionuclides, including those nuclides of particular interest in medical and biological applications. Specific effective energy (SEE) is defined as the energy absorbed per gram of target organ Y for each disintegration of the radionuclide in source organ X multiplied by Quality factors related to the linear energy transfer of the radiations involved. The Quality factors are included to estimate dose equivalent values (rem dose) as opposed to absorbed dose values (rad dose). A computer program has been written which calculates SEE values for a matrix of 22 source organs and 31 target organs for a total of 3410 entries. For each source-target combination the SEE value is tabulated for each of 4 kinds of radiation and for the sum of the 4 kinds

TABLE I. SOURCE AND TARGET ORGANS FOR SPECIFIC EFFECTIVE ENERGY TABLE

Organ[a]	Source	Target
Bladder Contents (BLAD CON)	x	
Bladder Wall (BLAD WAL)		x
Stomach Contents (S CONT)	x	
Stomach Wall (S WALL)		x
Small Intestine (S.I.)	x	x
Upper Large Intestine Contents (ULI CONT)	x	
Upper Large Intestine Wall (ULI WALL)		x
Lower Large Intestine Contents (LLI CONT)	x	
Lower Large Intestine Wall (LLI WALL)		x
Kidneys	x	x
Liver	x	x
Lungs		x
Nasopharyngeal Region (NP REG)[b]	x	x
Tracheobronchial Region (TB REG)[b]	x	x
Pulmonary Region (P REG)[b]	x	x
Pulmonary Lymph Nodes (LYMPH)[b]	x	x
Muscle	x	x
Ovaries	x	x
Pancreas	x	x
Skeleton		x
Cortical Bone (COR BONE)[c]	x	x
Trabecular Bone (TRA BONE)[c]	x	x
Red Marrow (R MARROW)[c]	x	x
Yellow Marrow (Y MARROW)[c]	x	x
Total Marrow (T MARROW)[c]		x
Cortical Endosteal Cells (COR E CL)[c]		x
Trabecular Endosteal Cells (TRA E CL)[c]		x
Total Endosteal Cells (T E CL)[c]		x
Skin		x
Spleen	x	x
Testes	x	x
Thymus		x
Thyroid	x	x
Uterus		x
Total Body (T BODY)	x	x

[a] Quantities in parentheses are abbreviations used in computer output.

[b] Subregions of the lungs.

[c] Subregions of the skeleton.

of radiation. The four kinds of radiation are: (1) gamma and x-ray radiation, (2) beta and monoenergetic electron radiation, (3) alpha radiation, and (4) recoil nuclei associated with alpha emission. In the output of the computer code these are designated by the symbols G, B, A and R, respectively. The total of all radiations is designated by the symbol T. Although not explicitly shown in the computer output, the Quality factors currently in use are 1 for gamma and x-ray radiation and for betas and monoenergetic electrons, 10 for alpha particles and 20 for recoil nuclei. Thus a user of these tables who is interested in absorbed dose

TABLE II. SPECIFIC EFFECTIVE ENERGIES FOR IODINE-131[a]

SOURCE → / TARGET ↓		S CONT	S. I.	ULI CONT	LLI CONT	LUNGS	THY-ROID	T BODY
S WALL	G	77.6	4.78	6.53	2.44	2.21	0.157	2.31
	B	389.						2.78
	T	466.	4.78	6.53	2.44	2.21	0.157	5.09
S.I.	G	3.86	44.0	19.9	23.9	0.380	0.0221	2.53
	B		243.					2.78
	T	3.86	287.	19.9	23.9	0.380	0.0221	5.31
ULI WALL	G	4.76	31.9	42.4	5.68	0.400	0.0579	2.64
	B			441.				2.78
	T	4.76	31.9	484.	5.68	0.400	0.0579	5.42
LLI WALL	G	1.30	20.9	7.55	50.3	0.103	0.0158	2.43
	B				719.			2.78
	T	1.30	20.9	7.55	770.	0.103	0.0158	5.20
LUNGS	G	2.32	0.332	0.340	0.0943	19.4	1.44	1.90
	B					194.		2.78
	T	2.32	9.332	0.340	0.0943	214.	1.44	4.67
OVARIES	G	1.05	13.6	5.43	12.7	0.628	0.0194	1.73
	B							2.78
	T	1.05	13.6	543	12.7	0.628	0.0194	4.51
TESTES	G	0.190	0.768	1.78	8.31	0.0965	0.0033	2.69
	B							2.78
	T	0.190	0.768	1.78	8.31	0.0965	0.0033	5.47
THYROID	G	0.190	0.0423	0.0514	0.0158	1.17	613.	1.67
	B						12100.	2.78
	T	0.190	0.0423	0.514	0.0158	1.17	12800.	4.42
T BODY	G	2.55	2.83	2.74	2.74	1.92	1.68	1.83
	B	2.78	2.78	2.78	2.78	2.78	2.78	2.78
	T	5.33	5.61	5.52	5.51	4.69	4.45	4.61

[a]Table entries are in units of eV-dis^{-1}-g^{-1}.

as opposed to dose equivalent can easily factor out the Quality factors. A majority of radionuclides do not emit alpha radiation and in such cases absorbed dose and dose equivalent are identical.

A list of the target and source organs is shown in Table I along with the abbreviations used in the computer output for those cases where abbreviations are necessary. It will be noted that the lungs and skeleton are subdivided into smaller regions which may have widely varying biological retention functions or radiation sensitivities for a given radionuclide. The tremendous advantage to the user of the SEE tables becomes apparent by referring to the example above where 3500 calculations were required. The number of calculations required is reduced to 7 since the summation over all radiations has already been made. Furthermore, no interpolations are necessary since all absorbed fractions are implicit in the SEE values.

TABLE III. SPECIFIC EFFECTIVE ENERGIES FOR STRONTIUM-85[a]

SOURCE-> / TARGET		S CONT	S. I.	ULI CONT	LLI CONT	LUNGS	COR BONE	TRA BONE
S WALL	G	106.	6.40	8.49	3.41	2.91	1.03	1.03
	B	18.3						
	T	124.	6.40	8.49	3.41	2.91	1.03	1.03
S. I.	G	5.13	63.5	26.1	34.3	0.526	1.38	1.38
	B		11.4					
	T	5.13	74.9	26.1	34.3	0.526	1.38	1.38
ULI WALL	G	6.52	45.7	58.9	7.65	0.536	0.955	0.955
	B			20.8				
	T	6.52	45.7	79.7	7.65	0.536	0.955	0.955
LLI WALL	G	1.71	29.9	10.1	73.3	0.120	1.45	1.45
	B				33.9			
	T	1.71	29.9	10.1	107.	0.120	1.45	1.45
LUNGS	G	3.17	0.489	0.523	0.141	31.9	1.46	1.46
	B					9.15		
	T	3.17	0.489	0.523	0.141	41.1	1.46	1.46
T E CL	G	0.947	1.31	1.98	1.76	1.64	6.70	6.70
	B						0.597	0.951
	T	0.947	1.31	1.98	1.76	1.64	7.30	7.65
R MARROW	G	1.48	3.03	2.34	3.78	1.77	6.96	6.96
	B						0.0346	1.05
	T	1.48	3.03	2.34	3.78	1.77	6.99	8.01
OVARIES	G	1.71	20.7	6.57	19.9	0.269	0.434	0.434
	B							
	T	1.71	20.7	6.57	19.9	0.269	0.434	0.434
TESTES	G	0.273	1.13	2.43	10.5	0.0463	1.32	1.32
	B							
	T	0.273	1.13	2.43	10.5	0.0463	1.32	1.32
T BODY	G	3.50	3.86	3.74	3.72	2.69	2.54	2.54
	B	0.131	0.131	0.131	0.131	0.131	0.131	0.131
	T	3.63	3.99	3.88	3.85	2.82	2.67	2.67

[a]Table entries are in units of eV-dis^{-1}-g^{-1}.

The information which the user must provide is the kinetic biological data related to the time integral of activity[1] of the radionuclide in each source organ. All the decay scheme data and static biological data such as organ weights, sizes and locations are contained implicitly in the SEE tables. Furthermore, data related to the basic interactions of nuclear radiation with tissue and bone are contained implicitly in the SEE tables. The transition from SEE values to radiation doses is simple. One merely multiplies the SEE value for target organ Y times the time integral of activity of the radionuclide in source organ X times a constant factor

[2]Time integral of activity is a number proportional to the total number of disintegrations which have occurred and is often measured in μCi-h or μCi-d.

to convert the final answer to the units of rads in the case of absorbed dose or to the units of rems in the case of dose equivalent. In order to find the total radiation dose to target organ Y one must, of course, use this simple procedure and sum over all source organs which contribute any dose to Y.

The SEE values for iodine-131 and strontium-85 are illustrated in Tables II and III. In order to bring the size of the tables within manageable proportions, rather than show the direct computer output which is quite voluminous we show only those source and target organs which are known to be significant in the metabolism of iodine-131 and strontium-85 in the human body. Furthermore, the entries in the direct computer output for alpha radiation and recoil nuclei have been suppressed in Tables II and III since no alpha particles are involved. The conversion of the SEE values in Tables II and III to radiation dose, while simple in principle as discussed in the immediately prior paragraph, is difficult in actual practice. This is because time integrals of activity in the various source organs depend on the method of administration of the radionuclide and on the compound administered. Furthermore, human subjects exhibit rather wide statistical variation for the retention of a given radionuclide in a given source organ. The emphasis in this study has been to interrelate basic nuclear data with certain static biological data in a useful way for internal dosimetry calculations and we feel the SEE are at present the best available vehicle for producing accurate dose estimates. It is fair to say that the variability of kinetic biological data is the limiting factor on the accuracy of radiation dose estimates in nearly all cases if the SEE tables are used.

As expected in the case of iodine-131 the data of Table II coupled with time integrals of activity indicate that the thyroid receives by far the largest radiation dose when the intake is oral or by inhalation. However, in the therapeutic use of iodine-131 the dose to the other target organs mentioned in Table II is very useful information. For strontium-85 the dose commitment to the target organs mentioned in Table III is more nearly uniform. Hence, the organ at greatest risk is not obvious and depends on the method of administration of the radionuclide and the nature of the compound administered.

We recognize that the SEE tables do possess certain limitations. They are based on the assumption that the radioactivity is uniformly distributed in the source organs. This may sometimes not be the case. However, since the position of localized hot spots in a particular organ of a given individual is usually impossible to anticipate, the assumption of uniform concentration is perhaps justified. Our method does not preclude taking into account non-uniform source distributions in an organ but at present the experimental data are too meager to warrant such a procedure. Also, since the SEE tables implicitly contain information on body organ weights, sizes and locations, these tables will not apply to children or adults who physically vary significantly from standard man. However, we feel the SEE tables to be a major step forward in providing the data base for the best possible radiation dose estimations with a minimum of effort.

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DISCUSSION

G.A. KOLSTAD: While the title of your paper is "Nuclear Data of Utility in Medical and Biological Applications", it is limited to radioisotopes for diagnosis, therapy or other in vivo applications. However, it is also important for radiation therapy to develop sound compilations of evaluated nuclear and biological data for the treatment of animals and human patients by radiations from particle accelerators – for gamma radiation, high-energy electrons, protons, alphas and heavier ions and even for π^- or possibly K^- mesons. What is the medical community doing to ensure that nuclear and biomedical data of this other class are being compiled, evaluated and made available in useful forms?

L.T. DILLMAN: I do not feel competent to answer the question. My contacts with the medical community have been confined primarily to internal dosimetry problems.

G.A. KOLSTAD: It would appear that there is a major gap which needs to be filled.

D.J. HOREN: I think this is probably true. There is a gap but some work is being done. At Oak Ridge, Alsmiller is trying to do calculations to compare the various types of particles for radiotherapy – neutrons, protons, heavy ions, pions, etc. I think the main problem is going to be that of trying to co-ordinate three groups: (1) the physicists, who will have to measure specific nuclear properties (and there are proposals for some work of this kind, especially for π-mesons), (2) the medical people and (3) the calculators (people like Alsmiller or Dillman).

G.A. KOLSTAD: It seems to me that it calls for a more organized and considered effort than has apparently been made so far.

D. BERÉNYI: Mr. Dillman has spoken at some length about internal and external bremsstrahlung. However, most of the data which he presented appear to have been calculated data. Could he indicate what he considers to be the really missing experimental measurements in this area?

L.T. DILLMAN: Very little experimental information is available concerning the interaction and production of bremsstrahlung in media of medical interest, such as bone and tissue. However, experimental bremsstrahlung measurements have been compared with theoretical calculations for specific elements and the comparison indicates that the theoretical values are in good agreement with experimental results, the errors being no larger than a few per cent. I believe, therefore, that one can proceed with confidence in applying theory for the complex media of biological interest. Certainly the errors will be smaller than the error in many biological parameters which must also be used.

DEVELOPMENT OF A COMPUTER-BASED NUCLEAR DATA COMPILATION – TABLE OF ISOTOPES*

M. LEDERER, J. M. HOLLANDER
Lawrence Berkeley Laboratory,
University of California,
Berkeley, Calif., United States of America

Abstract

DEVELOPMENT OF A COMPUTER-BASED NUCLEAR DATA COMPILATION – TABLE OF ISOTOPES.

On the basis of a proposal made by the authors in 1968, they are proceeding to apply computer techniques to the production of the next (7th) and future editions of the Table of Isotopes.

The use of computers is intended to accomplish several goals: (1) to reduce the probability of errors and simplify the compiler's job by improvements in the data-input procedure, by computer checks for all possible machine-detectable errors in the syntax and in the physics of the input data, and by elimination of redundant entry and proofreading of data re-used in succeeding editions of the Table of Isotopes; (2) to shorten the time delay between the cut-off data for acceptance of new literature and the publication of the compilation by nearly complete automation of the production process; (3) to enable the authors to produce automatically sorted or inverted compilations, such as tables of gamma rays from all isotopes produced by neutron capture in natural targets, ordered by energy; (4) to eliminate redundant entry of abstracted data by different compilers whenever possible.

An important part of the computer automation is the development of a text input and editing system for direct keyboarding of complex text. This system is being used for input of all tabular data. Graphical data (level schemes) are input in tabular form on punched cards and plotted by computer. A major saving in effort and improvement in accuracy should thus be obtained because level schemes can be modified easily and redrawn automatically, without the need for extensive drafting and redundant proofreading.

In 1968, shortly after the 6th edition of the Table of Isotopes was published, Lederer, Hollander and Meissner proposed a computer-based system for data compilation [1]. Although this system was motivated by our primary goal, the production of future editions of the Table of Isotopes, the use of computers includes some new techniques that are applicable to other types of data compilations and, more generally, to multi-purpose data bases.

Even though the topic of this paper was to be the computer-based system, I think that some readers are more interested in the nuclear data than the methodology of compilation. I shall make a few remarks about the status of the Table of Isotopes project, before returning to the discussion of some aspects of the automation.

The dramatic growth of nuclear data at the close of the last decade ended a pleasant era in which one or two people could compile all the data encompassed by the Table of Isotopes. Consequently, in 1968, we envisaged an expanded project that included more compilers and the use of computers to simplify the process of updating and printing. Actual compilation work was started in 1971 when initial funding was obtained.

*Work performed under the auspices of the US Atomic Energy Commission and the US National Bureau of Standards.

$^{131}_{53}I$

$t_{1/2}$ 8.085 5 d (Iso 22 715)
8.054 6 d (PMB 2 255)
8.067 7 d (PMB 2 360)
8.073 8 d (Iso 19 475)
8.070 9 d (NSEg 32 46)
no modification of $t_{1/2}$ by chemical environment (PR C3 1699)
others(RRou 13 485, RRL 2 41, CJC 31 120, PR 90 443, Nucl 11n3 14, PR 81 643, Nat 167 365)

Ψ β^- (PR 54 775)
Class A; Ident: chem (PR 54 775); chem, genet (PR 57 363)
Prod. fission (PR 56 1, Nwis 27 529, PR 54 775, Nat 158 163, PR 57 363, NNES 9 984, CJR 25B 371, Nat 161 520, NNES 9 1368)
I 7/2; μ +2.738 1; q −0.40 1; atomic beam (PR 119 2022)

β^- $\beta_{0.81}$ (0.6%), 0.6065 7(86%), 0.336 7(13%), mag (Phca 17 637)
0.807 10 (1.4%), 0.6059 12 (84.7 3%), 0.339 10 (13.3 3%), mag (PR 86 863)
0.810 5, 0.606 4, 0.335, 0.250, mag, $\beta\gamma$ coinc (PR 84 585, PR 81 642)
0.812 15 (0.7%), 0.608 5 (81%), 0.335 15 (9%), 0.250 20 (≤9%), mag, $\beta\gamma$ coinc (PR 86 212)
average β^- energy: 0.19, ion ch (PR 86 82)
others(ZP 179 62, Phil 43 648, Phil 43 221, Nat 170 916, PR 86 82, PR 83 860, PR 82 103, PR 81 482, Phca 17 658, PR 78 179, PR 76 94, PR 75 1270, PR 74 1879, PR 74 1640, PR 61 686)

γ (see also ^{131m}Xe)
0.080164 9, 0.284307 49, 0.364467 50, cryst (PR 91 1027)
0.080165 14 ($\dagger_\gamma$2.72 15, e_K/γ 1.33 16), 0.16398 2 (K/L 1.83 9, L_I/L_{II} 6.1 7, L_I/L_{III} 1.2 1, with ^{131m}Xe), 0.17723 3 ($\dagger_\gamma$0.36 2, e_K/γ 0.155 16, K/L 5.1 5, L_I/L_{II} 1.8 7, L_I/L_{III} 2.7 13), 0.2723 5 ($\dagger_\gamma$0.8 1), 0.284307 49 ($\dagger_\gamma$7.05 40, e_K/γ 0.041 4, K/L 5.5 5, L_I/L_{II} 2.9 4, L_I/L_{III} 3.4 5), 0.3180 4 ($\dagger_\gamma$0.11 2), 0.3250 4 ($\dagger_\gamma$0.04 1), 0.32578 5 ($\dagger_\gamma$0.45 3, e_K/γ 0.0236 28, K/L 6.2 13), 0.3585 5 ($\dagger_\gamma$0.020 4), $\gamma_{0.364}$ ($\dagger_\gamma$100, e_K/γ 0.018 1, K/L 6.3 6, L_I/L_{II} 4.5 8, L_I/L_{III} 5.0), 0.4048 5 ($\dagger_\gamma$0.080 7), 0.50299 8 ($\dagger_\gamma$0.36 2, e_K/γ 0.0085 13, K/L >5), 0.63699 9 ($\dagger_\gamma$8.0 4, e_K/γ 0.0043 4), 0.6430 4 ($\dagger_\gamma$0.18 2), 0.72289 10 ($\dagger_\gamma$2.10 15, e_K/γ 0.0037 4), [$\gamma_{0.364}$ (γ 82.2%) from level scheme (CML)], Ge(Li), Ge−scint $\gamma\gamma$ coinc, mag conv (PR 153 1321, NP 39 613)
$\gamma_{0.080}$ ($\dagger_\gamma$3.10), 0.17702 17 ($\dagger_\gamma$0.31 2), $\gamma_{0.284}$ ($\dagger_\gamma$7.42 40), 0.32568 10 ($\dagger_\gamma$0.28 2), $\gamma_{0.364}$ ($\dagger_\gamma$100 6), 0.50274 17 ($\dagger_\gamma$0.45 6), 0.63630 24 ($\dagger_\gamma$9.08 90), 0.72292 6 ($\dagger_\gamma$2.05 23), Ge(Li) (NP 82 289)
$\gamma_{0.364}$ (L_I/L_{II} 3.63 62, L_I/L_{III} 6.18 105, M/L 0.195 27, N+O/M 0.25 4), mag conv (APHu 28 13)
others(RaAc 10 1, CR C264 944, ZP 179 62, NP 43 650, NP 40 566, NP 31 456, NP 24 318, Magy 2 n3, IzF 23 206, PR 101 746, ArkF 8 21, Phca 20 243, PR 90 849, CJP 30 715, PR 88 884, PR 86 863, PR 86 212, CJP 30 35, Nat 170 583, ArkF 5 427, Phil 43 648, Phil 43 221, Phca 17 637, PR 84 585, PR 83 860, PR 83 679, PR 82 277, PR 82 103, PR 81 642, PR 81 482, Nucl 7n5 24, PR 76 94, PR 75 1615, PR 75 1544, PR 74 1640)

$\gamma\gamma(\theta)$ ZP 244 332, NP 82 289, ArkF 23 49, PR 90 849

$\beta\gamma(\theta)$ PR 145 907, PR 79 728
βe transverse polariz(θ): YadF 5 1037, NuoC 25 942
$\beta\gamma$ circ polariz(θ): ZP 179 62
nucl align: PR 120 1777

0.150 level of ^{131}I: $t_{1/2}$ 0.94 3 ns, delay coinc (DUzb n4 24)
0.95 5 ns, delay coinc (PR 140 B536)
0.76 5 ns, delay coinc (NP A161 471)
others(IzF 23 1445, ZETF 37 314, ArkF 11 110A, NP 1 281)
μ 2.77 50 if $t_{1/2}$=0.95 5 ns, IPAC (NP A102 203)

1.797 level of ^{131}I: $t_{1/2}$ 5.9 2 ns delay coinc (PR 140 B536)
g −0.16 5 if $t_{1/2}$=5.9 2 ns, IPAC (NP A102 203)

FIG. 1. Sample tabular listing for the Table of Isotopes, 7th edition.

We plan to maintain the traditional form and scope of the Table of Isotopes, with the following modifications:

1) Measured uncertainties will be included on all quantities.
2) Tables I and II of the 6th edition will be combined into a single table. Figure 1 shows an example of how this table will look. In place of the partial listing of "major radiations" that appeared in Table I of the 6th edition, we have included some derived information on X-rays and absolute γ-ray intensities, where not directly measured, in the detailed radiation data.
3) Reaction levels will be included for all nuclei. The detailed schemes for each nucleus will be drawn separately from a skeleton diagram for each mass chain, which shows the decay relationships and Q-values. A sample is shown in Fig. 2.

Although we believe that this is generally a very useful type of compilation, we realize that many important applications require simpler tables and more highly evaluated or reduced data – for example, best values or absolute conversion electron intensities. We are now compiling data for the next edition of the Table of Isotopes, with concurrent development of computer automation, as our primary goal. As a secondary goal, we intend to produce such additional tables by use of the computer and the addition of some data to the data files.

I turn now to the computer system. I emphasize from the outset that the computer cannot and should not replace the intellectual aspect of compilation and evaluation work; rather, it enhances this by the removal of much unnecessary clerical work that a data-evaluator must otherwise do. The major tasks we demand of the computer are the elimination of redundant copying, proofreading, and production work, and the detection of many types of errors that the computer can recognize from the syntax, the context, or the physical laws and probabilities applicable to a given kind of text or data. These are important tasks when one must deal with large volumes of data – of the order of 10^5 quantities.

At the heart of any data bank is a stored data file, mechanisms for entry, storage, and retrieval, and printing or plotting of the data, and a process we call "flagging" or identification. I do not want to dwell on these features, which are common to all data banks, but I should mention a few important aspects of flagging.

Flagging labels a piece of information by its type and possible use, thus facilitating retrieval. As a simple example, one "item" in our data file contains data on one isotope, in one data category (such as half-life), from one reference. The isotope, data category, and reference code are flags; in addition, the item is flagged to show method of measurement, compiler, typist, date of entry, and "status". "Status" is actually a collection of flags; these include intended use in the Table of Isotopes (i.e. the order of listing for this item among all $t_{\frac{1}{2}}$ entries for this isotope), whether the entry is a measured or an adopted best value, whether to use it in some other table, etc. In complex entries, e.g. a gamma-ray spectrum, the data may also contain internal flags identifying specific quantities, such as energies, relative or absolute intensities, conversion coefficients, and subshell ratios.

Two novel aspects of our computer development effort are (1) the use of an improved system for entering and editing data, and (2) extensive checking for errors by the computer. Both aspects are aimed primarily at improvement of the accuracy of the data base. The input-editing system also

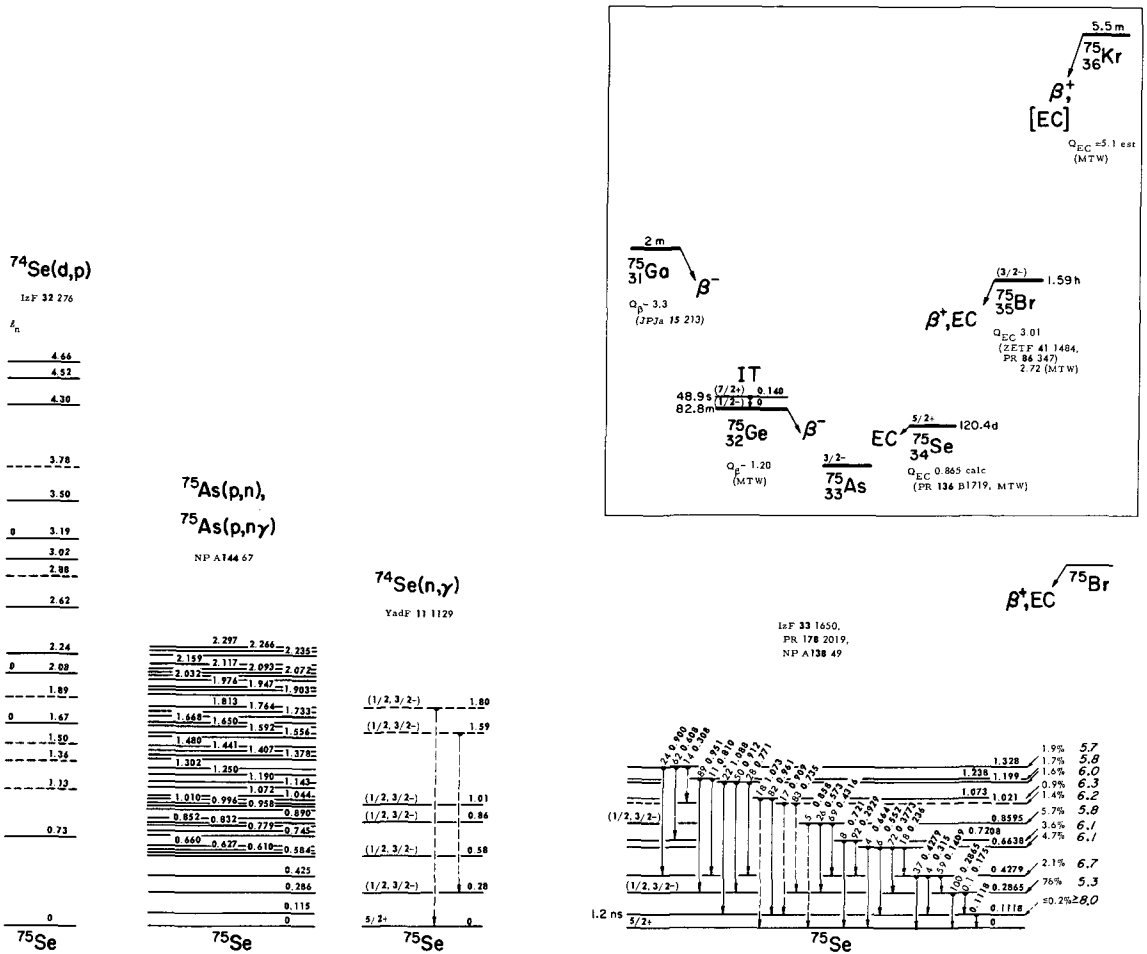

FIG. 2. Sample level-scheme drawing for the Table of Isotopes, 7th edition.

FIG. 3. Keyboard-display console, Table of Isotopes text input-editing system. (The keyboard shown in this photograph is an earlier prototype version.)

simplifies the entry process and avoids unnecessary re-copying of data by the compilers.

The input-editing system is an outgrowth of earlier developmental work in text processing at Lawrence Berkeley Laboratory, motivated by the Table of Isotopes and the Particle Data Compilation projects. Figure 3 shows the input console in use. The keyboard is an expanded typewriter keyboard, and the display is a storage oscilloscope. The console is connected to a small computer, which can drive multiple terminals. Figure 4 shows the keyboard layout. The character set includes complete Roman and Greek alphabets in upper and lower case, numbers, and a large number of special symbols. Fonts include "normal" and combinations of superscripts or subscripts, italics and boldface.

Some useful features of this sytem are the following:

1) It permits easy editing, both when entering data and later, after the data have been entered and proofread. A moving cursor can be positioned anywhere in the text and insertions or deletions readily performed. (Insertions appear at the bottom of the screen, and are re-placed in the proper part of the text by striking the redisplay key.)

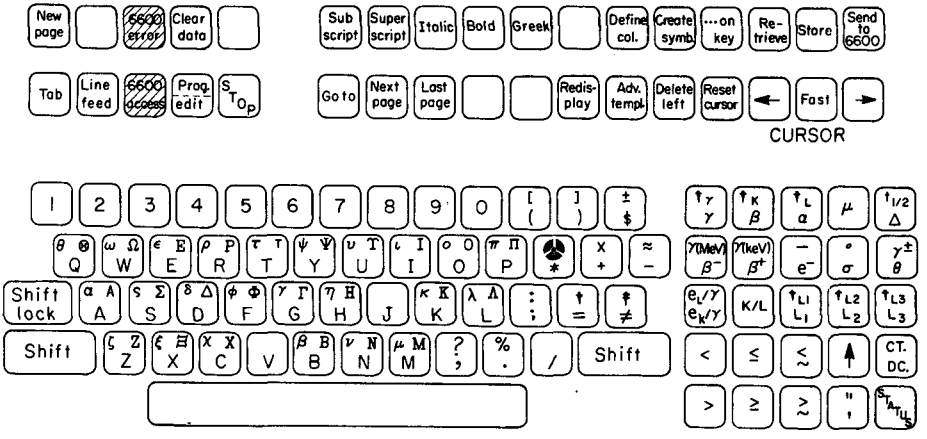

FIG. 4. Schematic layout of the keyboard. Special function keys in top two rows are for
a) position control ("Tab", "Line feed", "New Page")
b) display control ("Last Page", "Next Page", "Go to" (page number) "Send", and "Redisplay"
c) editing (Cursor keys, "Delete left", "Reset cursor", "Clear data")
d) retrieval of a specified item ("Go to" (item number X) "Send") (X = f for identifier flags, d for data)
e) storage and retrieval of a specified block of items or mass chain ("Store", "Retrieve")
f) template control ("Advance Template")
g) re-definition of a key ("Create symbol" (keys) "on key" (key))
h) column number identification ("Define column")
i) fonts ("Subscript", "Superscript", "Italic", "Bold", "Greek"). Foot pedals can also be used for subscript or superscript.

2) Compound symbols such as e_K/γ can be entered with a single key. In fact, the typist can create such a compound symbol and assign it to a key while typing.
3) A "template", or prompting routine ensures the inclusion of required data-identifiers (flags). The identifiers for each data item are entered via this routine; the upper portion of Fig. 5 shows an example of one completed template. Note that the format flag, which I did not mention before, defines the number of columns and their headings for a columnar entry.
4) A true columnar format can be used to enter data directly from published tables. (Positioning is controlled by the tab and line feed keys.) An example of such a table is shown in the lower portion of Fig. 5. Columnar data can be edited in the same manner as non-columnar data.
5) The system provides direct access to the stored data files via a high-speed link between the keyboard system and the main computer facility at the laboratory.
6) When not in use as a text editor, a console can be used as a standard teletype terminal connected to the main computer system.

We find that use of the keyboarding system serves not only to save time, but also to eliminate errors. It eliminates much error-prone re-copying by

ITEM: 630
TAPE: 30028
ISOTOPE: 115-Ag-47(20_m)[619]
DATA CAT.: γ(keV)
REFERENCE: NP_A143_289//70Hn01
METHOD: Ge(Li)_scint-Ge(Li)_γγ_coinc_Ge(Li)-scint_γβ_coinc
FORMAT: $E_\gamma \pm \Delta E_\gamma \_ \uparrow_\gamma \pm \Delta \uparrow_\gamma$
A OR C: A[619]
COMPILER: EB[619]
DATE: 11/20/72[619]
TYPIST: AMN[619]

****

1	131.4±0.2↑	14.4±0.6↓
2	213.6±0.2↑	24.8±0.6↓
3	229.7±0.2↑	100↓
4	237.1±0.4↑	2.2±0.2↓
5	243.6±0.4↑	1.8±0.3↓
6	277.4±0.6↑	0.4±0.2↓
7	303.3±0.2↑	3.4±0.2↓
8	326.6±0.2↑	10.6±0.8↓
9	360.9±0.2↑	3.3±0.4↓
10	372.6±0.2↑	10.2±0.8↓
11	389.3±0.2↑	1.9±0.3↓
12	417.3±0.4↑	1.7±0.6↓
13	473.2±0.2↑	18.7±0.9↓
14	507.7±0.2↑	7.1±0.7↓
15	539.6±0.3↑	0.7±0.2↓
16	548.3±0.3↑	1.1±0.2↓
17	586.0±0.3↑	0.8±0.2↓
18	649.9±0.2↑	13.8±0.9↓
19	699.2±0.2↑	10.1±0.8↓
20	719.0±0.4↑	0.4±0.2↓
21	750.0±0.4↑	0.6±0.2↓

FIG. 5. Sample output of part of a data item typed on the input-editing system. The upper portion contains the identifier flags, typed in response to the questions "ISOTOPE":, "DATA CATEGORY": etc. (Identifiers labelled with superscript numbers in brackets were defined during entry of a previous item.) The data (lower part of figure) are in columnar format, with the column headings given in the "FORMAT" identifier. _, $\frac{T}{T}$, and $\frac{L}{F}$ are symbols for blank, tab, and line feed.

hand, re-copying that would otherwise be necessary to convert the data to a special input format. It is flexible enough to allow the data compiler to use the symbols he knows and remembers – the language of physics.

The second novel feature of our computer development is the use of the computer to perform extensive checks for errors of form or syntax, and for violations of the laws and probabilities of physics. For example, the letter O is similar to the digit zero, but the computer knows that the letter is not an acceptable part of a number. Of course, the computer cannot always know a 2.375-MeV γ-ray should have been 2.357, but it will tell us if a list of energies is out of order. It also tells us that a 23.57-MeV γ-ray is highly improbable, and that 2.3 ± 0.05 is not an acceptable number. (It would be helpful if all scientists also understood this.)

The important point is that proofreading, just like data entry, is subject to human error; as any compiler knows, proofreading detects only a certain fraction of the actual mistakes. Checking by the computer is not a substitute for proofreading, but it can greatly improve the overall accuracy of the data base by warning the compiler of definite or probable errors.

One type of error readily detected by the computer is the violation of physical laws. This is especially relevant to highly correlated data, such as a nuclear level scheme. In the case of a level scheme, a misplaced transition, an incorrect level of transition energy, or an incorrect spin or multipolarity can frequently be detected because it violates energy, spin or parity conservation.

The system that we are developing will allow for easy exchange of data with other groups via format conversions that can be done by us. In addition to the benefits from sharing data, the checking features of our programs would thus become available for checking other data bases.

I have not mentioned some of the interesting applications of computerized data files, because I wanted to emphasize novel features we have developed for the purpose of creating a more accurate data base with less difficulty. But inverted or sorted tables, such as a table of gamma rays of isotopes produced by a specified activation process, ordered by energy, magnetic tapes of such data, and on-line retrieval of data are obvious applications that are possible once the data are stored and properly flagged in computer-readable form.

REFERENCES

[1] LEDERER, C. L., HOLLANDER, J. M., MEISSNER, L. P., A Computer-Based System for the Nuclear Data Compilation "Table of Isotopes", University of California, Lawrence Berkeley Laboratory Rep. UCRL-18530 (October 1968).

DISCUSSION

C. WEITKAMP: I am very glad, and I think most of the audience is too, that Dr. Lederer has made a clear statement as to what evaluation means, at least for his compilation, namely selection of data. I wonder whether this is the definition of other compilers as well?

M. LEDERER: Perhaps I didn't make myself clear. I define "evaluation" as comprising the two processes: (1) selection, and (2) the creation of new data ("best" values or weighted averages, derived or

calculated quantities, etc.). My intention was to draw attention to these two aspects, and to emphasize the fact that, except for data selection, evaluation means the creation of new numbers.

Miss K. WAY: There has been much confusion at this meeting in the use of certain words, such as "best value", "adopted value", "recommended value", and that horror of horrors, "evaluated value". I have even heard "evaluated best value". It would be very helpful if consistent definitions could be proposed and adopted by the nuclear community.

M. LEDERER: I hope I have not muddied the waters. My definition was merely an operational one. I wanted to draw attention mainly to the fact that when the compiler does any kind of evaluation, he is creating a new number.

Miss K. WAY: This is a novel point of view, which is somewhat shattering.

M. LEDERER: It becomes especially shattering when, after having been added to the literature, the new number proves to be wrong. People are more upset than when an experimenter remeasures a value and gets a poorer number than before, because they believe in evaluators more than in experimenters.

M. LAMMER: Let me perhaps attempt a sort of definition to distinguish between compilations and evaluations. I would call a compilation a collection and listing of experimental data. An evaluation should involve an examination and critical selection of experimental data resulting in a set of single values for single quantities. The performance of such an evaluation would then depend on what the result is needed for. If some data are needed quickly, for practical applications, one would merely select a few sets of data from the literature which seem to be the most reliable and average them. In a more careful evaluation, one would have to correct for standards used and experimental methods. All evaluators know the problems that arise with gamma-ray intensities, where uncertainties are often assigned without regard to whether a standard was available for calibration in the particular energy range or not. A careful evaluation has to take account of all this, even though such a procedure is time-consuming.

M. LEDERER: Yes, but I think one must always be careful to note that the selection of data itself is an expert process. For example, if in the Table of Isotopes one half-life is given for a certain isotope, we are in a very real sense recommending that as a best value to use. If we give two half-lives which agree fairly closely and have a comparable limit of error, we are saying that they are comparable and that you can choose between them or you can average. If we list two values that disagree, we are giving you a warning. If we list five values with a two-and-two disagreement and the fifth value is much less precise than the others or falls in one group or the other, we are giving you a very definite warning that the question hasn't been settled. So, in effect, we are making judgements in this way. In principle, our preference, given the time constraints, is to do this by data selection, at least in the tabular data.

Z. SZATMÁRY: I would like to express the point of view of users of compilations. We can discuss definitions of evaluations, recommended values and so on, but if I am a simple user of nuclear data, I cannot introduce five or ten values in a calculation but only one. Therefore, I would like to be able to rely on the opinion of an expert as to which single value I should use. Otherwise, I myself have to do the evaluation without having the necessary

scientific background for the job. This multiplication of the number of evaluations all over the world can lead to an explosion more dangerous than a nuclear explosion.

M. LEDERER: Yes, that is a very important point. I hope I haven't lost the user in this discussion. I think that from the standpoint of the Table of Isotopes we're giving you a simple answer. I'm saying: take the first value listed and you're probably all right. If there's a disagreement, I'll leave you to do the evaluation if you so desire. If we really believed that we could decide between one value or another or get a better value by averaging the two, we would have done so ourselves. I realize this is not a complete answer because the Table of Isotopes cannot cover the field in depth. The ultimate answer is that somebody has to decide in every case whether the best value is one of the measured values or whether some weighted average is necessary. In an ideal world we would have reduced every quantity down to a best choice, whether it be some measured value or "evaluated value".

B. ROSE: It must be remembered that there is not a single class of user for any one set of tables, but at least 30 different classes with different needs. I would guess that Professor Lederer's Table is directed more to the professional nuclear physicist than to the "simple user". It seems to me quite impractical to produce a single table which can satisfy the needs of the thirty different classes.

M. LEDERER: Yes. I think when I came to this meeting I had the idea that the Table was used by professionals as a quick reference – something that he could keep in the laboratory or take to a conference, and by the applied user more generally. More and more I have gained the impression here that our Table is perhaps used by the expert applied user in preparing specialized tables in different fields.

D.J. HOREN: Even to define compilation is difficult. For instance, is it to be defined as just taking the data that appear in a table of the author's paper, or does it also include reading the text and modifying such data if the text so indicates? As I mentioned in the presentation of my paper (IAEA-SM-170/47), it is important for the user to learn something about the compiler of the compilation he intends to use.

NUCLEAR DATA TABLES – A MAJOR DATA OUTLET

Katharine WAY
Duke University, Durham, N.C.,
United States of America

Abstract

NUCLEAR DATA TABLES – A MAJOR DATA OUTLET.

The importance of Nuclear Data Tables as a major outlet of nuclear data is, nearly seven years after initial publication, emphasized, and achievements to date and hopes for the future are discussed. Historical notes are given to provide background and perspective.

The completion of ten volumes of Nuclear Data Tables nearly seven years after initial publication in December 1965 seems an appropriate time to discuss achievements to date and hopes for the future. These ten volumes have presented 80 data compilations in a total of over 6000 pages. Nuclear Data Tables has thus become in the last few years one of the major publishers of nuclear data. It is hoped that a report on the journal to this Symposium will elicit suggestions for ways to make it more valuable to the international scientific community.

A few historical notes will give some perspective. In the late 1950s, the Nuclear Data Group, then under the aegis of the National Academy of Sciences – National Research Council, with the mission of organizing nuclear data by nucleus, felt so acutely the need for data organized by nuclear properties that it fostered and published data collections on abundances, masses, moments, etc. However, the effort was so time-consuming that it interfered with the main project of the Group. In 1962, N.B. Gove and Katharine Way suggested to the American Institute of Physics the idea of a compilation journal as a medium for the publication of data collections, but AIP decided that the inauguration of such a journal would be too risky. Fortunately, when the idea was brought up again at the sessions on Scientific Information at the Paris meeting in 1964, it caught the interest of commercial publishers and by the end of 1965, arrangements had been made with Academic Press to commence publication of Nuclear Data, Section A, Tables, which was to be devoted to compilations by property or topic produced by contributors anywhere in the world. Section B, Sheets, was to be devoted entirely to the output of the Nuclear Data Group on data organized by nucleus. In 1968 the sections were separated into two distinct journals, Nuclear Data Tables and Nuclear Data Sheets. The publishers felt they could operate without any page charge. Prices were to be kept low by using printout and other photoready copy for tables and graphs.

In 1965, a survey had shown that a number of excellent data collections already existed as reports. Therefore, one early goal was to attract this report literature to the journal, to make sure it conformed to the standards of refereed publications, and to reduce its often ungainly bulk by techniques generally unavailable to the issuers of reports.

Of the 80 compilations published in the first ten volumes of NDT, about 15 were issued originally in a report form which, however, was often very

different from that of the final publication. As the journal became known, many authors, who would previously have issued reports, submitted their compilations directly to NDT. A few manuscripts have resulted from suggestions of the editors but the great majority has been conceived by the authors themselves and submitted (in skeleton form if very long) in the same way that research papers are submitted to primary journals.

Opinions are often voiced that the task of compilation is so uncongenial to physicists that special inducements to undertake it must be offered in the form of invitation by distinguished committees. The editors, however, believed that data organization is attractive to many physicists, at least sometime in their careers, and that they will undertake it in the independent spirit in which they undertake pure research if the satisfaction of publication and recognition can be expected. The continued success of NDT shows that the standing invitation for manuscripts of a conscientious journal has been enough to produce a flow of distinguished compilations.

The 6000 pages of the first ten volumes have presented, as already mentioned, 80 data compilations. Since each volume has been divided into six issues the average issue has contained 1.33 papers with an average length of 75 pages each. The longest paper consisted of 292 pages, the shortest of 6.

The 116 authors have been citizens of 15 different countries, 65 of the USA, 14 of Germany, 9 of the USSR, 8 of Canada, 5 of the United Kingdom, 3 of Japan, and 1 or 2 each of Argentina, Belgium, Czechoslovakia, Finland, France, Iran, Mexico, The Netherlands, and Switzerland. The journal is thus becoming truly international.

Table I shows that of the 31 papers from countries other than the USA (some with multiple authors) 65% have originated in universities and 35% in "national laboratories". The proportions for the USA are just the reverse, 27% from universities and 73% from national laboratories.

Table II displays the number of papers the journal has published on different topics and the number of these which were tabulations of theoretical quantities rather than collections of experimental data. The theoretical papers have amounted to some 40% of the total.

Rather striking in Table II is the small number of neutron cross-section papers. Excellent international coverage of cross-section data has been achieved by the four cross-section centres in Brookhaven, Gif-sur-Yvette, Vienna, and Obninsk and much evaluated information is available from them in computerized form but the average nuclear physicist still has a hard time finding the values he needs. We hope that more cross-section tables designed for the non-reactor scientist will soon be available.

TABLE I. NUMBER OF NUCLEAR DATA TABLE PAPERS BY TYPE OF INSTITUTION

	University	National Laboratory	Total
USA	13	36	49
Other countries	19	12	31
Total	32	48	80

TABLE II. NUMBER OF NUCLEAR DATA TABLE PAPERS BY TOPIC

Topic	Number	
	Total	Theoretical
Angular-momentum and angular-correlation coefficients	11	11
Beta activity	1	
Beta-decay process	2	2
Capture gamma rays	4	
Cross-section, charged particles	8	1
Cross-section, neutron	5	
Fission	2	
Fractional parentage	2	2
Gamma activity	4	1
Gamma-ray conversion coefficients	8	7
Isobaric analog states	1	1
Masses and mass differences	8	
Moments	2	
Nuclear models	5	5
Miscellaneous nuclear properties	13	2
Penetration through matter of electrons	1	1
Penetration through matter of gamma rays	1	
Penetration through matter of heavy particles	2	
Total	80	33

Nuclear Data Tables has worked hard to make its tables easy to use. The editors believe that most users turn immediately to a table and try to figure out how to use it without first reading the introduction. The editorial policy is therefore to put all necessary explanations of policies and abbreviations in very concise form just in front of the tables. All that is left for the introduction is the historical comments, comparisons with other tables, mathematical developments, and other discussion not essential to the immediate use of the table. This arrangement is particularly helpful to those whose first language is not English and to scientists in neighbouring fields.

One can think of nuclear data collections as being divided into two classes, primary and secondary, the first consisting of collections of directly measured quantities with recommended values based on critical evaluation of the measurements, the second of compilations made from other compilations. An example of the latter might be a collection of values of the activation cross-sections, gamma-ray energies, and half-lives needed by activation analysts, all derived from recommendations in primary compilations. In general, the editors of Nuclear Data Tables have fielt that it is their first mission to publish primary compilations but they have recently accepted a

collection of neutron activation cross-sections based on other compilations because, as mentioned above, these cross-sections are difficult for the average physicist to locate.

Good secondary compilations depend, of course, upon the existence of good primary compilations. The speed and accuracy with which these can be assembled depend upon the ease with which the compiler can locate the relevant research papers and then extract the information. An international keyword system for help in finding all papers on a given topic is needed. It could be developed from the system proposed by the Nuclear Data Group in 1962, adopted some time ago by Nuclear Physics, and soon to be in use in Physical Review C. For help in extracting information on measured quantities, once the paper is located new ideas are needed. I imagine special formats or styles of presentation so that the main results can be found at a glance. This conference might designate a committee to develop and propose such styles. If the committee succeeds in suggesting something that is eventually adopted, primary compilations will be better, faster, and much, much cheaper.

In conclusion I would like again to invite comments on Nuclear Data Tables and suggestions of any kind for ways to make it more useful to scientists working in the fields represented at this Symposium.

DISCUSSION

M. LEDERER: I agree with your comment about putting necessary notes on each page, and would go one step further – one should, wherever possible, use universally understood symbols that need no explanation.

F. F. DYER: Has the format of the new Wakat table* been established?

Miss K. WAY: Not finally.

*M. A. Wakat, Nucl. Data A8 (1971) 445.

SYMPOSIUM SUMMARY
AND
ROUND TABLE DISCUSSION

SYMPOSIUM SUMMARY

W. B. LEWIS
Atomic Energy of Canada Limited,
Chalk River, Ontario,
Canada

This Symposium was called essentially to consider how the users of nuclear data can best be served by the measurers and compilers of such data. Considering first the users, they appeared to include almost all the producers and frontier-problem solvers of the world. The problems of the world are set by a tremendous total population, who by modern communications are aware of what rewards can come to a human life. That tremendous population is still rapidly increasing.

The average age of the world's population is probably in the teens and so for most, the main action in their lives is yet to come. For them to achieve and accomplish, and to be able to afford the accomplishment, nuclear data evaluated and compiled play more of a role than most people suspect. The contribution of nuclear data ranges into the mundane essentials of staying alive, keeping fed, and keeping warm and healthy, as well as into the most exotic projected activities such as sending eight men to land on the planet Mars (IAEA-SM-170/53). Another major world problem of humanity, that of eliminating terrorism, appeared to look to nuclear data chiefly in its most sophisticated aspects of implementing safeguards against terrorism by nuclear bombs (IAEA-SM-170/1, 54, 78). Minor applications, however, also appear in forensic science.

There are problems of choice all along the route because measuring and compiling nuclear data does not automatically provide all that the measurer and compiler seek in life. With the division of labour that has been established in the world and seems necessary to sustain so large a population, it falls to others to supply the food and life support of their servants, the measurers and compilers, who are not most productive if treated as slaves.

The keynote address to this Symposium assumed that the first hurdle has been passed and that the scientists exist and have completed their basic training, so their problem is to choose the activity that is good science because it can be seen as the "art of the soluble", but mostly the chosen paths must also be seen to yield a tangible benefit within a practical time-scale; a scale that now seems to be somewhat shorter than a single scientific career but could be much longer if the politicians of the world who steer and fund the division of labour exercised vision extending that far.

My philosophical introduction is, however, becoming too long and I must follow the keynote address and come to grips with some of the practical choices, assuming that the choices are to guide skilled measurers and compilers to be their most effective.

Let me take first the medical users. As presented, it would seem that their greatest need is for a science of radiobiology worthy of the name.

We were shown how inadequate are the concepts of radio-biological effectiveness (RBE) and the elementary ideas of microdosimetry and track structure to explain the extraordinarily large biological effects a small amount of ionizing radiation can have (IAEA-SM-170/59). Consider a gamma radiation dose of 500 rad to the whole human body. It is an energy density so small that it is measured in mere tens of thousands of erg per gram – it is 50 000 erg per gram – not enough to raise the temperature by more than 1/800th of a degree, yet it is likely to lead to the death of a human being. Yet similar amounts applied in therapy can have beneficial biological effects; it is as if radiation has not to be assigned an individual effect but rather a contribution in parellel with temperature, pressure, oxygen supply, etc.. which all together have an optimum but can be lethal when too far off that optimum. Radioisotopes play their role in medicine both in diagnosis and in therapy (IAEA-SM-170/92, 97). A number of isotopes, because of their convenient availability and suitable half-lives, appear in the various pharmacopoeia of the world (IAEA-SM-170/70), but there is room for more; that is more nuclear data, together with their evaluation and compilation, are looked for. Another very striking effect in therapy comes from pursuing the ideas of RBE and linear energy transfer (LET), making it attractive to apply high-energy protons and α-particles, as well as the more exotic artificial products, the π-mesons as specific agents against localized tumours. That whole field of the mesons produced by "high"-energy (or "intermediate" energy on the nuclear scale) particle accelerators introduces the nuclear data measurers and compilers to a new region of their science which is still mostly wilderness.

I would like to pursue this metaphor of the wilderness and the garden further as it seems so apt in this matter of the choices facing the nuclear scientists. Fortunately, they are many and have individual preferences. Some will find more satisfaction in pursuing their work in the well-ordered gardens, realms where the nuclear data are extensive and embodied in major compilations, programmed for interrogation by enquirers with all the data evaluated and recorded on magnetic tapes. Others will prefer pioneering with 800-MeV protons and the mesons, neutrons and spallation products (IAEA-SM-170/9) they produce, exploring in the wilderness (IAEA-SM-170/35, 45). The region of the 14-MeV neutrons from the D-T-reaction is on the edge between the wilderness and the garden, calling for careful planning to make the most meaningful measurements and evaluations (IAEA-SM-170/22, 31).

Turning to that major user-realm, namely, nuclear power generation which is needed to supply the food and energy of the growing population, we were reminded in the keynote address that a small problem, of such major concern that some 22 000 pages of testimony have been recorded during the past year, is calling for better experimental information on the energy released from the short-lived fission products immediately following the shut-down of a reactor from high power. In general, the application of nuclear data to nuclear power generation is very extensive and fairly satisfactory (IAEA-SM-170/8, 18, 79) for the established types of reactor, so very little was reported at the symposium. On the other hand, for the prospective fast breeder reactors nobody appears satisfied, and many papers relate to the outstanding discrepancies between the calculations based on the evaluated nuclear data and the results of experiments on reactor assemblies (IAEA-SM-170/69). The data are characterized as

differential data since they are related to neutron interactions with atoms in very narrow ranges of the total neutron energy spectrum. The experimental results, on the other hand, from reactor assemblies are characterized as integral data (IAEA-SM-170/19, 20). Reconciling the differential and integral data for fast reactors allows little play for the models and methods of calculation (IAEA-SM-170/38) so changes of differential nuclear data are still looked for (IAEA-SM-170/7, 30, 50, 67, 91).

There appears to be a major gap in time likely to occur before the large-scale exploitation of nuclear fission power receives any assistance from nuclear fusion. The assistance may not come as a contribution to energy so much as a contribution to the overall neutron and fuel cycles. Hybrid fission-fusion reactors (IAEA-SM-170/56) and sub-thermonuclear fusion chains (IAEA-SM-170/49) were shown to be still largely wilderness areas but perhaps deserving cultivation.

Before discussing the needs of some of the many other users of nuclear data, it may be well to note that speaking as we have of measurers and compilers can be misleading, for one and the same scientist is needed to pay attention not only to those two activities but also to evaluation and to broadening the range of the science itself. To quote the keynote address, there is "depth and complexity" in compilation, making it necessary for it to be supported by scientists "at a level sufficient for the compiler to be au courant with the data and to maintain his contacts with the nuclear community", so that "significant new nuclear science will come from nuclear compilations".

Many of the users of nuclear data were only identified by a few chance words. The applications of radioisotopes as tracers to study ways of improving the growth of crops and of adjusting feed to the optimum in animal husbandry are well known, but the advances of nuclear technology are making available even more convenient and suitable isotopes. Their choice, taking into account production, transmittal and application, makes use of many of the nuclear data compilations. Moreover, under the guise of chemistry, another whole technology using stable isotopes to serve as the tracers was discussed (IAEA-SM-170/96). These stable isotopes can then be identified and measured by one of several nuclear activation techniques with the aid of neutron sources such as ^{252}Cf, or experimentally convenient nuclear reactors (IAEA-SM-170/32), or particle accelerators bombarding the sample prepared in the form of a suitable target, or compact accelerators producing 14-MeV neutrons in the field or higher-energy accelerators including microtrons and betatrons (IAEA-SM-170/75).

Some special users were identified in fields of environmental science, in oceanography (IAEA-SM-170/36), in mining (IAEA-SM-170/15, 17) and many other fields of industry and applied science (IAEA-SM-170/42, 46, 65).

Moreover, some advances in technology were illustrated and foreshadowed, made practical by byproducts from the large-scale growth of nuclear energy. In particular, ^{238}Pu, the 90-year half-life isotope, will become available and its use was referred to not only in small energy sources for heart pacemakers (IAEA-SM-170/64), but possibly also as a fissile material augmenting its energy output by fission in a miniature reactor (IAEA-SM-170/39).

The experiments on controlled nuclear fusion lead to considerations of the regeneration or breeding of tritium that require an extension of not only energetic neutron but also particle reactions that produce tritium

(IAEA-SM-170/21). New technology is required for tritium retention and recovery which may be monitored by nuclear techniques.

Very extensive studies are opening up on the detailed structures of protective surface films on which so much depends in industrial processes. Amongst these techniques are nuclear backscattering and thick-target nuclear reaction studies (IAEA-SM-170/33).

Hot-atom chemistry was shown to need some compiled nuclear data (IAEA-SM-170/28).

The science of plasmas and lasers interrelates also with nuclear science in some aspects both currently and prospectively (IAEA-SM-170/53).

NUCLEAR DATA COMPILATIONS

It became apparent at the Symposium that not only the external users but also the nuclear scientists who formed the great majority of the participants were largely unaware of very many of the large number of nuclear data compilations available and their special merits (IAEA-SM-170/60). A few years ago, the IAEA published a bibliographical compilation of the compilations that included a total of 40, whereas now 90 have been identified. Many papers listed and explained the relevance of these compilations. To remind you, let us look again at a set of slides[1] presented in one of the papers. This historical sequence followed a line of interest to oceanographic applications of neutron activation analysis (IAEA-SM-170/36). A sequence of interest to medical users would be somewhat different because of the need to include consideration of the radiations of the daughter radionuclides. Moreover, the penetrating power and the relative biological effectiveness of the individual radiations is significant in medical and biological applications (IAEA-SM-170/46, 43).

The oceanographic nuclear scientists concluded that their need was best served by a compilation presented on magnetic tape so that it could be amended, extended and updated constantly by a computer (IAEA-SM-170/36). Some other users call for the opposite extreme, a compact tabulation that presents only the information special to their needs. Both would agree that they seek accurate information. The evaluation of nuclear data for accuracy is a major work requiring all the facilities acquired by the compilers of the major basic tabulations. It is necessary to preserve a strong line of communication between these basic compilations (IAEA-SM-170/47, 48) and the multiplicity of smaller special-purpose compilations (IAEA-SM-170/83). It can be very frustrating to a user of small compilations to find different values quoted in different sets of tables for the same nuclear constant, with no guide to his choice.

Much can be done by the measurer to help the evaluators in their difficult task. In general, much more detail needs to be given concerning the assessment of errors both concerning the methods, possible systematic errors and points of reference where measurements are relative. It is the general experience of evaluators that different measurements do not agree within the supposed limits of error. The condition makes it difficult to recognize the exceptions where the work has been so carefully done,

[1] Not published in the paper in question.

and so closely related to accurate standards, that the results should not be adjusted to a value outside the limit of error. On the other hand, a plea may be entered that where there is room for any doubt the measurer should be brought into consultation and should regard his work as unfinished until agreement is reached with the evaluator.

The nature of the nuclear data required in compilation is continually extending. The introduction since 1963 of the high-resolution germanium γ-ray spectrometer extended the need for detailed γ-ray spectra (IAEA-SM-170/37, 44, 62, 76), and as activation analysis extends to the use of higher-energy neutrons (IAEA-SM-170/77) γ-photons (IAEA-SM-170/66) and charged particles (IAEA-SM-170/6, 68, 66), the range of nuclear levels and isomeric states with different half-lives grows broader (IAEA-SM-170/44, 60, 93). Users repeatedly requested absolute intensities of γ-rays. Moreover, despite the high resolution, interferences are important and should be readily found in the tabulations (IAEA-SM-170/32). Techniques of analysis are constantly extending, involving coincidences not only between γ-rays but also with X-rays and Auger electrons.

The range of nuclei of interest not only includes the direct fission products but also nuclides resulting from neutron capture by fission products. Several papers discussed tabulations of fission product data (IAEA-SM-170/12, 13, 34, 63, 74, 94)[2]. There is interest also in the heat released by fission products (IAEA-SM-170/58).

The half-lives and independent yields of the fission products are of importance in the non-destructive analysis of spent reactor fuel (IAEA-SM-170/12, 54).

Standard sources for the calibration of the energy scales of spectrometers also need to be well chosen (IAEA-SM-170/23).

A plea was entered that stopping power data not only for protons and α-particles in various solids but also for heavier particles should be measured and included in evaluated compilations (IAEA-SM-170/16, 33).

The successful use of theoretical relations to extend nuclear data was mentioned in several connections (IAEA-SM-170/16, 30), including unresolved activation resonances, especially neutron resonances in reactor components (IAEA-SM-170/2) and excitation functions in charged-particle nuclear reactions (IAEA-SM-170/10, 11).

A growing collaboration is evident between regional nuclear data centres. This appears most advanced in the field of neutron-nuclear data for reactors (IAEA-SM-170/4, 50). Similar collaboration is sought in other fields (IAEA-SM-170/25), especially in activation analysis, which is becoming so complex (IAEA-SM-170/24).

The current trend in university teaching towards interdisciplinary activities is well exemplified by the field of activation analysis. It introduces students to the limitations of compilations so they not only swell the ranks of compilers but learn of the difficulties (IAEA-SM-170/3). I am sure that all participants in this Symposium will leave with greater appreciation and sympathy for the compilers.

The data evaluators and compilers indicated a desire to reach internationally-standardized formats as well as key reference data, such

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[2] Paper IAEA-SM-170/94 by E. A. C. Crouch presents a new compilation of published fission product yields from 15 heavy nuclides.

as the neutron yield in ^{252}Cf fission. They also would like to sharpen the meanings attached to "evaluated data" and other loose phraseology used by compilers. Moreover, their work would be assisted by a wider adoption of keywords in the scientific journals (IAEA-SM-170/83, 11). It was warned, however, that keywords cannot uncover all the hidden but related data (IAEA-SM-170/33).

EPILOGUE

In the oral presentation of this summary I avoided mentioning names. The present version, however, is annotated with references to the original papers. We have been most fortunate to have had the benefit of the presence and participation of so many of the creators of nuclear data evaluations and compilations at the symposium. In fact, it can be said that we have had leading representatives from the major centres of these activities throughout the publishing world. What has emerged from the symposium is a general awareness of the magnitude and importance of the complex task in which we enjoy their leadership.

I appreciate the privilege of having had to work hard to distil an essence from the learned detail that is embodied in the papers and was contributed in the discussion. I am aware that I am reporting very little of the scientific detail and solid substance. I hope that those who look for more will have the time and opportunity to find it in the published Proceedings.

ROUND TABLE DISCUSSION

A. FINKELSTEIN (Chairman): Ladies and gentlemen, we shall now start the round table discussion which will conclude the Symposium. The members of the discussion panel will be Drs. Lewis, Kolstad, Yankov, Grinberg, Wapstra and Ferguson. I should like first of all to ask Dr. Lewis whether he has anything to add to the summary of the Symposium which he has just presented.

W. B. LEWIS: I gather that this round table is to address itself to the question of where we go from here. At the end of my summary there were some indications that we should be seeking further international collaboration between compiling centres. Presumably the mechanisms for this actually exist within the International Atomic Energy Agency and the International committees. The International Nuclear Data Committee is a body that can have sub-committees and the Agency is an organization that can have consultants. Hence either of these mechanisms would seem appropriate for bringing people together to reach the type of common agreement they feel is necessary.

A. FINKELSTEIN: I think it would be useful at this point to hear from Dr. Kolstad, because he was Chairman of the International Nuclear Data Committee during the years 1970-1971.

G. A. KOLSTAD: We are all increasingly aware of the emphasis these days (sometimes ad nauseam) on the relevance of what we are doing. I think that the relevance of the subject we are dealing with has perhaps been underestimated by society as a whole and even by the nuclear science community itself. We are witnessing the development of problems in many areas of science and technology, referred to by Dr. Lewis in his summary, where nuclear data are becoming increasingly important. Yet some of the responsible people in those areas find it difficult to recognize the importance of such data for their activities. Take, for example, the question of fusion reactors. The people in the United States concerned with research and development relating to controlled thermonuclear fusion are beginning now to do the neutron calculations that they will be faced with as they move into the fusion reactor area. These neutronics calculations, as Dr. McNally has pointed out (IAEA-SM-170/49), are highly dependent on a great deal of nuclear data which at present do not exist. The problem is going to be one of educating those responsible to fund work in nuclear data measurements, compilation and evaluation which feeds their particular technology or discipline.

We have had the problem of declining support of research. I think this has been not only an American but virtually a world-wide phenomenon. This support reached its peak in the United States around 1967 and since then has declined, both in terms of real dollars and, even more sharply, in terms of dollars corrected for the cost-of-living index, until this year, when we are hitting what I hope is the bottom. Things seem to be levelling off as far as support of nuclear physics is concerned and therefore I would

hope, with the increasing importance being assigned to the area of nuclear data, that we shall find it possible to augment some of these activities. We have broadened our recognition of nuclear data to a very considerable extent, as described by Professor Havens (IAEA-SM-170/4). We have restructured the United States Nuclear Data Committee in a way which reflects, perhaps not completely, but at least to a better degree than before, the needs of other areas of science and technology for nuclear data. We have done this in such a way as to keep the parent committee small while, at the same time, establishing committees for the various disciplines and sub-committees which are concerned with particular applications and which will report back to the policy level group. It is hoped that with this approach we shall be able to develop, in an organized fashion, the mechanism for input from the technologically based industries, from the various sciences and from the research and development areas which will need nuclear data. We have also proposed that a similar approach be followed by the international committees, under the auspices of both the NEA and the IAEA.

This problem of handling the broader scope of nuclear data is currently being considered by both the European American Nuclear Data Committee and the International Nuclear Data Committee. The latter body has been acting jointly with a recently established working group on nuclear structure and reaction data and this has stimulated it to move along more rapidly in this direction than might perhaps otherwise have been the case. But we need to organize better input mechanisms from a broader range of users than exist at the present time. This is very difficult to achieve. We have found at this meeting, for example, that many users in the fields of space science, accelerator development, shielding and so forth, have explained how nuclear data were needed but they have not provided the measuring community with a quantitative set of data requirements which could then be met by a concerted programme of measurement. This is a matter which obviously needs further attention.

We also need to strengthen the data centres and to organize them better in order to meet the needs of society. Up till now data centres have been seeking to satisfy three main types of requirements. The National Neutron Cross-Section Centre at Brookhaven was responding mainly to applied needs in the fission reactor field. The Nuclear Data Project founded by Dr. Way was motivated primarily by the needs of basic science. Many of the other data compilation efforts were motivated by the compilers' personal research interests. When all these things are taken together they constitute a wide variety of needs, all of which have to be met. I think the nuclear data centres will have to recognize all three of these very legitimate demands for adequate compilation and evaluation.

The question of whether or not the evaluation is adequate is another matter that came up at this meeting and my feeling is that a relatively larger fraction of the effort should be devoted to evaluation, even if it possibly means a slowing down of the compilation activity to some extent.

Lastly, I would like to mention that we need to recognize areas other than those which were represented at this conference, some of which were referred to by Dr. Lewis, e.g. agriculture and various branches of industry. I think this is where a good deal more can be done in providing data services. We might also even consider tapping them as sources of funds for this kind of work.

A. FINKELSTEIN (Chairman): I have noted with great interest Dr. Kolstad's suggestion concerning a number of possible new financial sources. We must explore these very diligently albeit with limited hopes.

G.B. YANKOV: It seems to me that the Symposium constituted an extremely important forum of evaluators, measurers and users of nuclear data and I hope that it resulted in each of these three groups having gained a much better understanding of each other's problems. We have been confronted with evidence of a great expansion in the utilization of nuclear data. Earlier we were concerned only with neutron data but at this meeting much attention was paid to non-neutron data as well. The increasing flow of information will, of course, bring with it an increase in difficulties. In these circumstances the role of international co-operation will also become more important, because it provides the only basis for coping with all the tasks that are facing us. We in the Soviet Union realize this and we have set up two non-neutron data centres to assist the Obninsk Neutron Data Centre. They are still very young and are not yet completely organized, but we are placing a great deal of hope in them. We consider that activities in connection with all types of nuclear data should constitute a single united effort and should not be broken down into individual mechanisms. Each centre should do its own work but this work should then be co-ordinated so that there will be no duplication of effort and so that the whole range of activities can be covered.

I believe the time has come to prepare lists of nuclear data requirements for other fields, e.g. applied fields. Some experience is already available along these lines, although it is related only to neutron data. During the last two or three years, with the help of the International Atomic Energy Agency, the formulation of data requirements in connection with safeguards problems and thermonuclear reactions has been undertaken on an international basis. I believe that in still other fields where nuclear data are used, we must now begin drawing up individual lists of requirements rather than running them all together.

At this point I should like to touch on a problem which is closely related to the success of our endeavours. Various speakers at this Symposium have referred to the difficulties which we face. It must be remembered that most of the things we decide on here will have to be carried out by young scientists and it is a known fact that young people and older people have rather different opinions about the world. Therefore some of the difficulties that have been mentioned here may reflect such different points of view. It may be that the young scientists do not consider these difficulties to be so great. Many of these young people are represented at this Symposium and it would be very interesting to hear their opinion. In conclusion, I should like to express the hope that, by making an effort to reach agreement on our problems, we shall be able to cope with all of them.

A. FINKELSTEIN (Chairman): Thank you very much, Dr. Yankov. No doubt we are all young in heart and in spirit, but let us hope that young people will be able, increasingly, to travel to various international meetings.

B. GRINBERG: Being in complete agreement with what my fellow speakers have said, I have very little to add. I think we can consider that this Symposium was an important step forward, due largely to the efforts of the International Atomic Energy Agency and the International Nuclear Data Committee. I am sure we all hope that it will be followed by other steps, otherwise it will only have been informative in character, making

us aware of the problems but without pointing the way to solutions. For me, at least, the Symposium has brought out the fact that, along with the many users who are aware of the needs in the domain of nuclear data that are not always satisfied, there are many researchers in various disciplines who are still unacquainted with the problems involved.

In order to take the next step forward, it is necessary, as Dr. Kolstad has said, to set up appropriate mechanisms. I must admit that I personally do not quite see what these mechanisms will be, considering the great diversity of nuclear data needed by the different disciplines. The people who will have to evaluate cross-sections are not going to be the people who will be evaluating parameters of decay schemes. In these circumstances it is obvious that the International Atomic Energy Agency, which has already played an important role in the matter of neutron data, is an essential centre for a future action. To be sure, I have no illusions regarding the financial resources that the Agency will be able to make available for this kind of work. If we want to develop compilation and evaluation activities, the implication would seem to be that the interested Governments should make the necessary funds available to international groups. Therefore these Governments need to be made aware of the problem and perhaps this Symposium could be of help in that process.

However, regardless of the mechanisms set up and the resources allocated, the task is beyond the capabilities of any single country, so that there will be a need either for co-ordination among national groups or at least for a division of work. One example of this is the collaboration between certain United States and Soviet groups which has been arranged with the aid of the International Atomic Energy Agency. What has been done in a restricted area should, I believe, be extended to cover all fields requiring the compilation and evaluation of nuclear data. Although the need for evaluated data is obvious in the field of metrology, it has emerged from the various papers presented at this Symposium that they are needed for many other disciplines as well.

A. FINKELSTEIN (Chairman): Thank you, Dr. Grinberg. I think it is precisely in the matter of co-ordinating national activities that the Agency will have an increasingly important part to play, particularly in those fields which are not yet well covered by its Nuclear Data Section, such as all the non-neutron data areas. Of course, this will have to be done with the funds at present available because it will obviously be some time before we can expect a supplementary budget for this purpose. Nevertheless, the co-ordination function could be exercised even at this stage to deal with some of the problems you have raised.

A. H. WAPSTRA: I shall confine myself to a few scattered remarks. I would first like to say something about availability of compilations. Dr. Münzel has described to me the great appreciation expressed by recipients of his questionnaire for that part of the questionnaire which contains a list of compilations now available. I myself have had the strange experience that now, nearly two years after the publication of my latest mass table, people are still using the earlier one in which the precision is much lower. So this is a matter that needs some attention. Could not Dr. Way perhaps publish in the first issue of every volume of Nuclear Data Tables a kind of compilation of current compilations in the field? It would not need to be an evaluated list, but it should not include out-dated compilations or at least not include them without comments.

Then there is the matter of evaluations. Some objections have been raised to unnecessary evaluations and the question has been asked whether we could limit ourselves to mere compilation. I think the answer is no. I remember, for example, a paper which included, amongst other things, an alpha decay energy I needed. The value given in the abstract was different from that given in a table and when I checked the figure showing the spectrum, it contained a third value different from both. I felt I just had to evaluate. That was not too difficult in this case because in the figure two digits had evidently been transposed and the error quoted in the abstract was absolutely impossible, but yet some evaluation was evidently necessary. One can, as Dr. Lederer has shown in a slide, list several values for a half-life along with their errors, but then one must make sure that one does not mix standard errors with limits of errors which are about three times as large. Again, some evaluation is necessary. I would almost prefer to go to the other extreme and always publish a recommended value, the sole exception being the case where you take the best value only if, in your evaluated opinion, the next best one is at least three times worse.

I would next like to make a remark about request lists. Berényi has made an interesting study of a request list for nuclear data. He found, if I remember correctly, that many of the requested data were already available, either with the requested accuracy or not very much worse. In any case, I have certain reservations about the requested precisions. I cannot prove this, but I strongly suspect that, in order to be on the safe side, people often ask for accuracies one or two orders of magnitude higher than really necessary. And sometimes exaggerated requests for accuracy are compounded, as in shielding calculations. Designers of accelerators, for instance, often apply shielding formulae to get an order-of-magnitude better results, i.e. lower radiation levels, than officially required. Since the allowed radiation doses are, in turn, devised with an order-of-magnitude safety margin, we get very dependable shielding but we are also wasting the taxpayers' money. Once again, I think we should take some care in this respect.

I was quite impressed by the amount of new work done in activation analysis, but I would like to think that further development is possible here at an increase in cost by a factor of not more than about two. Large-volume Ge/Li counters can be mounted in anti-Compton shieldings, improving peak-to-background ratios by an order of magnitude and thus opening up a new range of possibilities for non-destructive analysis. It may be well worth someone's trouble to prepare tables and atlases of spectra analogous to those of Dr. Heath, for use with this new tool.

To close with a few general remarks, in my opinion the importance of this Symposium has been threefold: first, it has helped acquaint participants with many compilations, perhaps with more than they knew existed; second, it has made them aware that a lot of work still remains to be done; third, it has made compilers — or at least some of them — aware of how necessary it is to devote great care to the presentation of data, especially for users in other fields.

A. T. G. FERGUSON: I shall follow Dr. Wapstra in talking to rather specific points. First of all, I would like to remark that, as in the United States, the national Nuclear Data Committee of the United Kingdom is being restructured to take account of more broadly based nuclear data requirements and to co-ordinate the needs of a much wider range of users. In

moving from customers who are concerned purely with the reactor business to this broader clientele, we must realize that we have an important educational function to perform and that the degree of sophistication we can expect from these users is limited, as regards both the formulation of requests and the realism, for example, of the accuracy which they demand. It will take some time before we can have the rather rigorous standards which have over the years become characteristic of the reactor request lists.

Another fact that has struck us as we attempt to broaden our base is the appalling ignorance of the vast majority of potential users of nuclear data as to what is available. As a community responsible either for the production of nuclear data or its compilation, I think we have the further responsibility of selling our product to this market, which we are now beginning to explore in some detail. Dr. Wapstra has made a suggestion about listing the available compilations in a widely read journal like Nuclear Data. With all due respect for this proposal, I think that effective advertising has got to be much more widely spread, so that it will reach many of the people who would certainly benefit by the availability of these compilations but have never heard of Nuclear Data or anything else at this level of sophistication. I feel that it is our duty as a scientific community to get articles describing the type of material which we have available into much more popular publications and technical trade journals.

One of the lasting impressions which I have gained from this Symposium is the importance of the basic evaluated compilations to this whole field. They represent the source material from which everything else is drawn, and all the other user-orientated compilations depend on them.

I have been working for many years in the neutron data field and even now I still sit through meetings with reactor physicists in which the whole burden of the discussion is complaints about the terrible problems of codes for translating from this format to that format. The fact that fairly substantial libraries have grown up independently in different parts of the world has a perfectly adequate historical justification. There is every excuse for the fact that there are differences and that we have to live with these problems. However, in view of the way this field is widening, I feel we should make it one of our most urgent endeavours to ensure that in five years' time I am not sitting through further meetings listening to the same kind of discussions about codes for translating other types of nuclear data from yet one format into another. This, in my opinion, is one of the most urgent tasks facing the community represented at this meeting.

A. FINKELSTEIN (Chairman): I would suggest that we now open the discussion to questions and comments from the other participants in the Symposium.

Miss K. WAY: As someone who has been in the nuclear data business a long time, I think it is time for the distinguished committees to give some thought to the need of providing the compiler with the proper tools. You have been considering new subjects which could be tackled but nobody has said a word so far about easing the life of the compiler. I think that until this is done, there is no chance of getting more, better and cheaper compilations. As I said in the presentation of my paper (IAEA-SM-170/83), what the compiler needs is to be able to find the papers and to be able to extract the data from them in a reasonable time. I think this implies the use by authors of standard formats for presenting certain types of data.

Much thought and experiment should be devoted to this. Decisions should not be reached hastily. Formats should be tried out to see how they work. But until this can be achieved, the poor compiler is going to have to go through the terrible drudgery with which he has had to cope all along. This is a foolish situation.

An even simpler matter, and one in which some signs of progress are now apparent, is the location of the papers. I am referring here to an indexing or key-word system. Such a system can be coded and, I would hope, be adopted on an international scale. This would be very helpful. It would also be valuable to have more standard abbreviations. These are all things which can be done only by international groups of the kind you are proposing – except that you haven't proposed that they should do any of these things. So, I would urge you to consider points like these.

B. GRINBERG: I fully share the views of Miss Way and I think she was right to draw attention to this problem, which was already raised within the International Working Group on Nuclear Structure and Reaction data (IWGNSRD), which met at Vienna in March 1972. I believe that the recommendations made by that Group* to the authors of scientific papers would simplify and ease the life of both compilers and evaluators because, as things now stand, they are losing considerable time in seeking the information they need. The Working Group requested that authors themselves should take the trouble to evaluate the accuracy of their findings.

W. B. LEWIS: Taking up Dr. Way's suggestion, I think there is something that can be done. All the scientific literature is abstracted and I am sure that those who compile the abstracts and submit them sometimes feel just as annoyed as the compilers with the material they have to work with. So I suggest that somebody should provide these abstractors with a specific statement about the requirements of compilers and that, where appropriate, they make an annotation to the effect that the abstract was prepared in the face of difficulties arising from the fact that the manuscript did not meet the prescribed conditions. This would help to get the information out to the authors.

D. BERÉNYI: I have a few short comments. First of all, on the subject of request lists, the formulation of a good list is not an easy matter. For example, at the first session of the IWGNSRD meeting to which Professor Grinberg referred, several different request lists were presented. For example, there were lists of about 15 isotopes for which half-lives were required or lists of around 20 isotopes for which the gamma yields were needed. In most cases, of course, this information was already published, so that request lists of this kind were of no use.

I think the first step is the preparation of application-oriented compilations, because if we have good compilations for, say, activation analysis or safeguards, we can expect a response as to whether or not the accuracy of the data is sufficient for the people working in those particular fields.

My second comment is concerned with a quite different problem. If I remember correctly, Dr. Horen said in the presentation of his paper (IAEA-SM-170/47) that his group can evaluate about 20 mass chains per year. Of course, this rate is very slow and I think it is essential to distinguish here between the collection of primary data and the evaluation.

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* The Working Group's recommendations are contained in the report INDC(NDS)-46/U+W and have been quoted in part in an editorial in Phys. Rev. Lett. 29 5 (9 October 1972) 973-974.

It is very important to have as complete a collection of data as possible; we need data files or, as a first step, reference files, as in the case of neutron data. The evaluation of the data is a somewhat different problem, I think, and it would be good if these evaluations could be carried out by groups other than the Nuclear Data Group at Oak Ridge. The evaluations could still be published either in the well-known periodicals Nuclear Data Tables or Nuclear Data Sheets. If we have good centres as sources of the primary data, then one group could be responsible for evaluating a particular mass chain and another group another mass chain.

The third and last comment has to do with the need for atomic data. It is a matter which I have already mentioned at this meeting, but I should like to emphasize it once again. This is a field where the interdisciplinary aspects are very important because a large number of phenomena are involved, for example, internal conversion, electron capture and heavy-ion reactions with different targets; there are also some applied aspects, such as X-ray fluorescence, spectroscopy for chemical analysis and so on. Data are needed, for example, on yields for bombarding targets with electrons or with X-rays. There is no relevant compilation here, or else the available data are full of gaps. Of course, here again, the first step is to examine the compilations to see what are the really missing data.

J. A. CZUBEK: Being a representative of the users, my interest is in getting accurate nuclear data. There has been a great deal of discussion at this Symposium on whether one should calculate, evaluate or estimate — that is to say, on which value one can take as a true value. This is important because we users are very often obliged to provide results with an accuracy of better than 1%. For example, a situation where the half-life of an isotope is known with an accuracy of only 10% and the scatter of the experimental data may also be about 10%, or even 20%, is very unsatisfactory for our work. It therefore seems to me that it would be a good thing for a round table discussion to be organized under the auspices of the Agency in an effort to establish some criteria on how data should be processed in order to get the most accurate value.

D. J. HOREN: On the question of youth, I agree heartily with the comments of Dr. Yankov and I might report in this connection that it was by our soliciting the opinion of youth that we really succeeded in getting the key word system accepted by the Physical Review. It was only by solicitation of the membership of the Nuclear Physics Division that we were able to get enough weight behind our opinion to promote this idea.

Dr. Wapstra has expressed concern about advertizing and I fully sympathize with him. We have tried to do something about this, too, seeking to advertize his mass tabulation through the Radiation Shielding Information Center (RSIC), which, unlike ourselves, has extensive communication with the user community. Another way in which the Data Project is trying to advertize, if you will, is by personal contacts with people in the applied areas, including industry, laboratories, etc.

As far as internationalization of the data collection and compilation effort is concerned, the Data Project has been involved in this for many years, as witness the number of compilers that we have in foreign countries, including the USSR. In the organization of international efforts, I agree, every effort should be made to ease the burden of the compilers who are actually engaged in the work, since they are so far behind already. We would have some ideas on how to go about doing this.

One point made by Dr. Münzel in passing seems to have gone unnoticed. I am thinking of the need to make data producers realize the potential uses of their information in applied areas. There are many occasions on which the data producers actually measure quantities which they never report and which would apparently be very useful in applied work.

As we look to the future, I think we have to be realistic in discussing the collection and tabulation of data. Enormous quantities of information are involved and before sitting down to decide how to go about this work we must ask the question: How many times does any one datum ever get used? I believe that examination of this problem in detail will show that the answer varies from data type to data type. The question will then arise of whether it is worth the cost to have a big organized union collecting every piece of information. In my opinion, there are certain data that have extensive uses. This is true of decay data, as has been demonstrated over and over again at this meeting, and of a few other selected types of data. However, there are vast numbers of data for which this is not true: to take only one case, there are the 2000 gamma rays just in neutron capture. Of course, there are many more cases which we haven't even mentioned, to say nothing of those which haven't been discovered yet. It isn't at all clear whether anyone is ever going to look at these things. You can tabulate them but a justified need for an evaluation may never arise. There will be large quantities of such data available, measured to probably very high degrees of accuracy.

In conclusion, I should like to make the point that we have no problem finding the data. We feel that our system is very effective, and I think that it has been demonstrated at this Symposium that, at least as far as much of the data contained in the papers we look at is concerned, the reference system seems to be working quite well.

N. M. SPYROU: As I mentioned in my paper (IAEA-SM-170/3), a nuclear activation group is being formed in the United Kingdom amongst British universities, medical schools and institutes of higher education. The comments made by the compilers have been noted and we hope to submit our particular problems and needs to them in the form they request. However, it must be pointed out that such a group is really there to make life easier for the poor users, rather than for the compilers. I wonder whether the formation of such consumer groups is not eventually going to make life much more difficult for the compilers when they come up against such organized pressures.

J. J. SCHMIDT: I would like to come back to Dr. Horen's point that we should be realistic about the future. I agree, but I also believe that something useful could be achieved. I would like to cite the example of the neutron data field, in which, on the basis of a continuing assessment of the requirements, international mechanisms for compiling and exchanging of information have been set up and do work. I gather from many comments and suggestions made during this Symposium that there is a feeling that a start should be made on setting up similar mechanisms in the field of nuclear structure and decay data, for example, to develop computer files for such data, which would be available to everyone upon request. However, setting up the mechanism and having the data available is one thing, the priority of what one should actually put into the files is another. I agree that one needs to ask how the data are actually used, how often and what is their priority.

D. J. HOREN: I may be rather bold, but I am already assuming that we have or will shortly have, the answer to the data handling aspect of that problem. Obviously, I am including the decay data. This should be computerized. This is something I have been maintaining for a long long time. Essentially, we are already doing this: all our decay schemes are drawn up with computers, which means that they are effectively computerized, at least the parts that we have evaluated.

J. J. SCHMIDT: I think this would be a very good starting point for other people as well. I understand, for example, that the two new nuclear data centres in the Soviet Union mentioned by Dr. Kondurov (IAEA-SM-170/25) and Dr. Chukreev (IAEA-SM-170/24) are planning to use your keywords and your reference files. In fact, I believe they have already started. I am sure there will also be interest in developing countries to participate in such an international endeavour.

G. A. KOLSTAD: If I might comment somewhat facetiously about what has been said on the subject of youth, I would remind you that someone once said that all wisdom does not spring from a bassinet. I think that youth has a good deal to offer but I also think there is a great deal to be gained from the experience of the kind of people sitting around this room, most of whom are over thirty.

I think that Dr. Berényi's comment was particularly pertinent. As you will recall, he specified two tasks to which we would have to address ourselves immediately as we move forward in the attempt to establish discipline-oriented sub-committees: the first is the development of application-oriented request lists; the second is the preparation of supplemental atomic and molecular compilations, needed to carry out work, for example, in controlled thermonuclear fusion. I can think of many other areas as well. In biology and medicine there is certainly a need for a great deal of that kind of information. In safeguards, supplemental X-ray information represents a natural augmentation of nuclear data. There are obvious needs in agriculture. And in the domain of nuclear physics, as pointed out in paper IAEA-SM-170/33, we need a better understanding of the stopping power problem. I think all these matters represent vital and valid charges to these yet-to-be established groups.

Finally, I would like to mention a person who has been largely ignored at this meeting, namely, the person who makes the measurements. On the whole, persons working in nuclear physics have in the past been motivated by reasons unrelated to filling the gaps in nuclear data compilations and, once they have the data they need to develop their basic science, they move on to other areas, going into heavy-ion physics, meson physics or high-energy physics or something else. There is going to be a problem of motivating the nuclear physicist at universities and elsewhere to do the kinds of measurements which many of use need but which have rather little basic nuclear physics content. I don't know how that problem is going to be answered.

B. ROSE: In reply to Dr. Kolstad's question, might I suggest, as a first practical step, that the IAEA should fund a few fellowships for this specific purpose. A number of fellowships are being used for training at various laboratories and if a few of them could be devoted to some of these particular tasks, it might perhaps be a step in the right direction.

H. JUNG: In connection with what Dr. Ferguson said concerning the presentation of nuclear data especially for the use of non-experts, I would

like to point out, as an example, that it may be a great advantage to have access to large computer files containing thousands and even millions of data, such as those on neutron cross-sections. For the non-expert, however, I believe the old BNL-325 is still a good book.

H. MÜNZEL: In the discussions at this Symposium it has been claimed on several occasions that a detailed request list is the most important thing that we should aim for. I would like to speak a few words of warning on this subject. I am fully aware that it would be very convenient to have detailed lists of this kind: they would ease our life very much because we could sit back and say we have to measure these and these data and we don't have to think about why we need them. However, I can think of certain fields for which such a detailed request list cannot be obtained. In activation analysis, for instance, when you have to solve a problem and don't have the data, you have two choices: you can either make the necessary measurement yourself or you can decide to forego activation analysis and apply some other method. You will not state that you will need to know the half-life of a particular nuclide, in, say, three years' time because the chances are that by the time the measurement was made and the result evaluated, you would not be interested in it any longer. In these types of applications, therefore, it will be very difficult to get specialized or a detailed request list. If one sought to determine requirements, all you would perhaps get is the answer: we need all data for all nuclei and we need them with high accuracy. The point of my warning, then, is that if one assigns priorities to data, one should not do so solely on the basis of detailed request lists available.

A. FINKELSTEIN (Chairman); Since there seem to be no further contributions to the discussion, I should like to propose that we bring the Symposium to a close. When the IAEA Secretariat began organizing the meeting it felt some concern over the large number and diversity of subjects to be taken up, assuming that this would automatically mean that some participants would have less interest in certain of the sessions. However, applying the yardstick of audience participation and of duration and intensity of discussion, I think this Symposium can be considered to have successfully reached its objective. Of course, credit for this does not belong to the Secretariat. A meeting is successful when it can draw on good specialists in the fields considered, and this has been true here.

Of course, we shall consider and study carefully the papers and summaries of the discussions we have heard here in an effort to obtain guidance in defining the Agency's programme for the years ahead. Evidently many of the participants hope that increased efforts will be made in matters relating to evaluated data and that work will be continued along these lines. Many would also like to see more work on non-neutron data, in particular decay schemes. This is a subject which we are going to take up with deliberation and prudence so that we can establish suitable co-operation with the existing groups and, in the light of the results obtained, increase our activities in this sector to the greatest extent possible.

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LIST OF PARTICIPANTS

ARGENTINA

Ricabarra, G.	Comisión Nacional de Energía Atómica, Av. Libertador 8250, Buenos Aires

AUSTRIA

Eder, O.J.	Österreichische Studiengesellschaft für Atomenergie, Lenaugasse 10, 1080 Vienna
Lammer, M.	Österreichische Studiengesellschaft für Atomenergie, Lenaugasse 10, 1080 Vienna

BELGIUM

Beets, C.	Nuclear Energy Centrum, Boeretang 200, 2400 Mol
de Corte, F.	Institute for Nuclear Sciences, Ghent University, Proeftuinstraat, 9000 Ghent
Demildt, A.	Nuclear Energy Centrum, Boeretang 200, 2400 Mol
de Wouters d'Oplinter, R.	BelgoNucléaire S.A., 25, rue du Champ de Mars, 1050 Brussels
Franeau, J.	Université de Mons, 17, Place Warocqué, Mons
Garnir, H.	Université de Liège, Institut de Physique Nucléaire, B 400 Liège
Piron, J.	Centre National de Documentation Scientifique et Technique, 4, Bd. de l'Empereur, 1000 Brussels
Schepers, J.C.	Nuclear Energy Centrum, Boeretang 200, 2400 Mol

BRAZIL

Herdade, S.	Comisão Nacional de Energia Nuclear, Rua General Severiano, 90 Botafogo, Rio de Janeiro

CANADA

Bartholomew, G.	Atomic Energy of Canada Ltd., Chalk River, Ontario
Lewis, W.B.	Atomic Energy of Canada Ltd., Chalk River, Ontario
Merritt, W.F.	Atomic Energy of Canada Ltd., Chalk River, Ontario
Mitchell, I.V.	Atomic Energy of Canada Ltd., Chalk River, Ontario
Thorson, I.M.	Simon Fraser University, Burnaby 2, B.C.
Walker, W.H.	Atomic Energy of Canada Ltd., Chalk River, Ontario

CZECHOSLOVAK SOCIALIST REPUBLIC

Kott, J.	Skoda Concern, Pilsen
Simáne, C.	Faculty of Nuclear Sciences and Physical Engineering, Brehova 7, Prague 1
Zbytovsky, A.	Skoda Concern, Pilsen

DENMARK

Mortensen, L.	Research Establishment, Risø, 400 Roskilde

EGYPT, ARAB REPUBLIC OF

Ahmed, Abdel-Rassoul	Atomic Energy Establishment, Cairo

FINLAND

Silvennoinen, P.	Technical Research Centre of Finland, Reactor Analysis Group, SF-02150, Otaniemi

FRANCE

Abou El-Ela I. (Mme)	Centre d'études nucléaires de Grenoble, Foyer Concordie

Barrandon, J.M.	Centre national de recherches scientifiques, Saclay, Laboratoire P. Sue
Barré, J.Y.	Centre d'études nucléaires de Cadarache, B.P. n°1, 13115 St. Paul-lez-Durance
Beauge, R.	Centre d'études nucléaires de Saclay, B.P. n°2, 91 Gif-sur-Yvette
Berger, R.	Centre d'études nucléaires, B.P. n°6, 92 Fontenay-aux-Roses
Berquez, D.	FRAMATOME, 77, rue du Mans, Courbevoie
Bertrand, F.J.	Centre d'études nucléaires, 29, rue de la Fédération, Paris 15ème
Blachot, J.	Centre d'études nucléaires, B.P. n°85, Hôtel Métropole, 38041 Grenoble Cédex
Blons, J.	Centre d'études nucléaires, 29, rue de la Fédération, Paris 15ème
Bouchard, J.	Centre d'études nucléaires, B.P. n°6, 92 Fontenay-aux-Roses
Boucharec, J.L.	FRAMATOME, 81, rue du Mans, 92 Courbevoie
Cance, M.	Centre d'études nucléaires de Saclay, B.P. n°2, 91 Gif-sur-Yvette
Cohen, Y.	Centre d'études nucléaires de Saclay, B.P. n°2, 91 Gif-sur-Yvette
Communeaj, F.	Centre d'études nucléaires, 29, rue de la Fédération, Paris 15ème
Costa, L.	Centre d'études nucléaires de Cadarache, B.P. n°1, 13 St. Paul-lez-Durance
Coulomb, P.	Centre d'études nucléaires de Saclay, B.P. n°2, 91 Gif-sur-Yvette

Debrun, J.L.	Centre national de recherches scientifiques, 16, quai A. France, Paris
Delaunay, B.	Centre d'études nucléaires de Saclay, B.P. n°2, 91 Gif-sur-Yvette
Devillers, C.	Centre d'études nucléaires, 29, rue de la Fédération, Paris 15ème
Doan, N.V.	Centre d'études nucléaires de Saclay, B.P. n°2, 91 Gif-sur-Yvette
Doutriaux, D.	FRAMATOME, 77, rue du Mans, 92 Courbevoie
Engelmann, Ch.	Centre d'études nucléaires de Saclay, B.P. n°2, 91 Gif-sur-Yvette
Fiche, Ch.	Centre d'études nucléaires, 29, rue de la Fédération, Paris 15ème
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Legrand, Jean	Centre d'études nucléaires de Saclay, B.P. n°2, 91 Gif-sur-Yvette
Lott, M.	Centre d'études nucléaires, 15, rue de la Fédération, Paris, 15ème
Martinet, M.	Groupement atomique alsacien atlantique, 20, avenue Edouard Henriot, 92 Le Plessis-Robinson
Martinot, M.	Centre d'études nucléaires de Saclay, B.P. n°2, 91 Gif-sur-Yvette
Moreau, J.	Centre d'études nucléaires de Cadarache, B.P. n°1, 13 St. Paul-lez-Durance
Nguyen, V.D.	Centre d'études nucléaires, B.P. n°6, 92 Fontenay-aux-Roses
Nimal, Bernadette	Centre d'études nucléaires, 29, rue de la Fédération, Paris 15ème
Nimal, J.C.	Centre d'études nucléaires, B.P. n°6, 92 Fontenay-aux-Roses
Ouvry, F.	Centre d'études nucléaires, 29, rue de la Fédération, Paris 15ème
Panisset, J.C.	Centre d'études nucléaires de Cadarache, B.P. n°1, 13 St. Paul-lez-Durance
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Roy, M.	Centre d'études nucléaires, B.P. n°6, 92 Fontenay-aux-Roses
Samak, N.	Centre d'études nucléaires, 29, rue de la Fédération, Paris 15ème
Soleilhac, M.	Centre d'études nucléaires, 29, rue de la Fédération, Paris 15ème
Tellier, H.	Centre d'études nucléaires de Saclay, B.P. n°2, 91 Gif-sur-Yvette
De Tourreil, R.	Centre d'études nucléaires, B.P. n°6, 92 Fontenay-aux-Roses

GERMANY, FEDERAL REPUBLIC OF

Bächmann, K.	Lehrstuhl für Kernchemie, Technische Hochschule Darmstadt, Hochschulstrasse 4, 61 Darmstadt
Buerk, K.	Kernforschungszentrum Karlsruhe, Zentralstelle für Atomkernenergie Dokumentation, 7501 Leopoldshafen
Debertin, K.	Physikalisch-Technische Bundesanstalt, Bundesallee 100, D-33 Braunschweig
Ebel, G.	ZAED, Kernforschungszentrum Karlsruhe, 7501 Leopoldshafen
Filges, D.	Kernforschungsanlage Jülich, 517 Jülich
Gilles, P.	Kernforschungsanlage Jülich, 517 Jülich

Göttel, P.	Kernforschungsanlage Jülich, 517 Jülich
Henkelmann, R.	Institut für Radiochemie, Technische Universität München, D-8046 Garching bei München
Hübner, H. (Mrs.)	Interatom, D-506 Bensberg/Köln
Jäger, U.	Kernforschungszentrum Karlsruhe, Karlsruhe
Jung, H.	Institut für Kernphysik der Universität Kiel, Kiel
Kriváň, V.	Max-Planck-Institut für Metallforschung, Laboratorium für Reinststoffe, Katharinenstrasse 17, D-707 Schwäbisch Gmünd
Krüger, K.	Arbeitsgemeinschaft Versuchsreaktor, 517 Jülich
Lieser, K.H.	Technische Hochschule Darmstadt, Darmstadt
Mattes, M. (Miss)	Institut für Kernenergetik der Universität Stuttgart, Pfaffenwaldring 31, 7000 Stuttgart 80
Meixner, C.	Kernforschungsanlage Jülich, 517 Jülich
Münzel, H.	Kernforschungsanlage Karlsruhe, Karlsruhe
Neuert, H.	Institut für Experimentalphysik, Universität Hamburg, 2 Hamburg 36
Priesmeyer, H.G.	Institut für Reine & Angewandte Kernphysik der Universität Kiel, 2054 Geesthacht
Ross, E.	Technische Hochschule Darmstadt, Fachbereich Anorganische Chemie und Kernchemie, Hochschulstrasse 4, D-61 Darmstadt
Scherer, W.	Kernforschungsanlage Jülich, 517 Jülich
Stiller, P.	Institut für Kernenergetik der Universität Stuttgart, Pfaffenwaldring 31, 7000 Stuttgart 80
Wagemann, R.	Arbeitsgemeinschaft Versuchsreaktor, 517 Jülich

Wagner, S.
Physikalisch-Technische Bundesanstalt,
Bundesallee 100,
D-33 Braunschweig

Wahl, D.J.
Arbeitsgemeinschaft Versuchsreaktor,
517 Jülich

Weitkamp, C.
Institut für Angewandte Kernphysik,
Postfach 3640,
5700 Karlsruhe

Wolf, R.
Institute of Clinical Radiology,
Langenbeckstrasse 1,
D-65 Mainz

GREECE

Dritsa, S. (Miss)
Nuclear Research Center "Democritos",
Aghia Paraskevi Attikis,
Athens

Mavroyannakis, E.
Nuclear Research Center "Democritos",
Aghia Paraskevi Attikis,
Athens

HUNGARY

Berényi, D.
Institute of Nuclear Research,
Bem-tér 18/C,
Debrecen

Csikai, G.
Institute of Experimental Physics,
Kossuth University,
Bem-tér 18/A,
Debrecen

Gado, J.
Central Research Institute for Physics,
P.O.B. 49,
Budapest 114

Nagy, M.
Nuclear Reactor at Technical University of Budapest,
Műegyetem Rakpart 9,
1111 Budapest

Szatmáry, Z.
Central Research Institute for Physics,
P.O. B. 49,
Budapest 114

Vorsatz, B.
Central Research Institute for Physics,
P.O. B. 49,
Budapest 114

INDIA

Eswaran, M.A.
Nuclear Physics Division,
Bhabha Atomic Research Centre,
Bombay 400085

IRAN

Pazirandeh, A.	Tehran University Nuclear Centre, P.O. Box 2989, Tehran

IRAQ

Mousa, M.	Karkh 2/4/25, Al Mansoun City, Baghdad

ISRAEL

Rothenstein, W.	Technion, Israel Institute of Technology, Haifa
Yaari, A. (Mrs)	Racah Institute of Physics, The Hebrew University of Jerusalem, Jerusalem

ITALY

Bonalumi, R.A.	C.I.S.E., Cas. Post.3986, I-20100 Milano
Cecchini, G.P.	Progettazioni Meccaniche Nucleari, L. 60 S. Giuseppe 3, 16121 Genova
Farinelli, U.	Reactor Physics and Calculations, Casaccia Nuclear Centre, 00060 Santa Maria di Galeria, Roma
Kind, A.	Laboratori Nazionali dell'INFN, Legnaro, Padova
Marinelli, F.	Ministerio Industria, v. Molise 2, Roma
Moschini, G.	Laboratori Nazionali dell'INFN, Legnaro, Padova
Sandrelli, G.	ENEL-CRTN

JAPAN

Fuketa, T.	Japan Atomic Energy Research Institute, Tokai-Mura, Naka-Gun, Ibaraki-Ken
Murakami, Y.	Department of Chemistry, Faculty of Science, No.1-2, Fukasawa, Setaguya, Tokyo

NETHERLANDS

Gruppelaar, H.	Reactor Centrum Nederland, Petten N.H.
Wapstra, A.	Instituut voor Kernphysisch Onderzoek, Oosterring dijk 18, Amsterdam
Zijp, W.L.	Reactor Centrum Nederland, Petten N.H.

NORWAY

Steinnes, E.	Institutt for Atomenergi, P.O. Box 40, 2007 Kjeller

POLAND

Czubek, J.A.	Institute of Nuclear Physics, Ul. Radzikowskiego 152, Krakow
Kuchowicz, B.	University of Warsaw, Department of Radiochemistry and Radiation Chemistry, Zwirki Wigary 101, Warsaw 22
Marcinkowski, A.	Institute of Nuclear Research, Świerk k/Otwocka
Sujkowski, Z.	Institute for Nuclear Research, Świerk k/Otwocka

PORTUGAL

Martinho, E.	Laboratório de Física e Engenharia Nucleares, Sacavém

SOUTH AFRICA

Steyn, J.	National Physical Research Laboratory, P.O. Box 395, Pretoria

SWEDEN

Holmqvist, K.E.B.	AB Atomenergi, Studsvik, 61101 Nyköping

Persson, R.B.R.	University of Lund, Radiation Physics Department, Lasarettet, S-22185 Lund
Strömberg, N.L.G.	Research Institute of National Defense, Stockholm

SWITZERLAND

Bédert, W.	Nestec, La Tour de Peilz, Vaud
Brunner, J.	Eidgenössisches Institut für Reaktorforschung, CH 5303 Würenlingen

UNION OF SOVIET SOCIALIST REPUBLICS

Belyaev, F.	Institute of Experimental and Theoretical Physics, Moscow
Chukreev, F.	Kurchatov Atomic Energy Institute, Moscow
Kondurov, I.	Leningrad Nuclear Physics Institute of the Academy of Science of the USSR, Leningrad
Yankov, G.	Kurchatov Atomic Energy Institute, Moscow

UNITED KINGDOM

Crouch, E.	Atomic Energy Research Establishment, Harwell, Didcot, Berks
Dennis, J.	National Radiological Protection Board, London
Edens, D.	C.E.G.B., Berkeley Nuclear Laboratories, Berkeley, Glos
Ferguson, A.	Atomic Energy Research Establishment, Harwell, Didcot, Berks
Harbison, S.	Royal Naval College, Greenwich, London SE 10
James, B.T.	Institute of Naval Medicine, Alverstoke, Gosport, Hants
Moore, J.	UK Authority Safety and Reliability Directorate, Wigshaw Lane, Culcheth, Warrington, Lancs
Spyrou, N.	Physics Department, University of Surrey, Guildford, Surrey

UNITED STATES OF AMERICA

Angelo, J.A.	University of Arizona, Tucson, Ariz. 85721
Clark, B.	Martin-Marietta Aerospace, Dept. 1610, P.O. Box 179, Denver, Colo. 80201
Dillman, T.	Ohio Wesleyan University, Delaware, Ohio 43015
Dyer, F.	Oak Ridge National Laboratory, Oak Ridge, Tenn. 37830
Fluharty, R.	Los Alamos Scientific Laboratory, P.O. Box 1663, Los Alamos, N.Mex. 87544
Gewantman, L.H.	National Bureau of Standards, Washington, D.C. 20234
Gurewitsch, A.	General Electric Co., 1 Riner Rd., Schenectooly, N.Y.
Harris, R.	Los Alamos Scientific Laboratory, P.O. Box 1663, Los Alamos, N.Mex. 87544
Havens, W.W., Jr.	Columbia University, 520 West 120 St., New York, N.Y. 10027
Heath, R.	National Reactor Test Station, USAEC, Washington, D.C. 20545
Horen, D.J.	Oak Ridge National Laboratory, Oak Ridge, Tenn. 37830
Kolstad, G.A.	US Atomic Energy Commission, Washington, D.C. 20545
Lederer, M.	University of California, Lawrence Berkeley Laboratory, Berkeley, Calif. 94720
Markow, B.	USAMC Stiteur, IG Farben, Room 740, Frankfurt/Main, Federal Republic of Germany
McNally, J.R.	Oak Ridge National Laboratory, Oak Ridge, Tenn. 37830
Ozer, O.	Brookhaven National Laboratory, Upton, N.Y. 11973

Rousso, S.	US Atomic Energy Commission, Germantown, Md.
Schneider, R.T.	University of Florida, Department of Nuclear Engineering Sciences, Gainesville, Fla.
Stubbins, W.F.	University of Cincinnati, Cincinnati, Ohio 45221
Thom, K.	US Atomic Energy Commission, NASA, Washington, D.C. 20545
Thorpe, M.M.	Los Alamos Scientific Laboratory, P.O. Box 1663, Los Alamos, N.Mex. 87544
Way, K. (Miss)	Duke University, Durham, N.C. 27706
Weinberg, A.M.	Oak Ridge National Laboratory, P.O. Box X, Oak Ridge, Tenn. 37830
Wolkenhauer, W.C.	Battelle Northwest, Battelle Boulevard, Richland, Wash.
Zirkind, P.	Picatinny Arsenal, Dover, N.J. 07801

YUGOSLAVIA

Maksić, R.	Federal Administration for Inter. Scient. Educ. Cult. and Technical Operation, Kosančićev Venac 29, Beograd
Tomas, P.	Institut "R. Bošković", Zagreb

ORGANIZATIONS

CCE (Commission of European Communities)

Aten, A.H.W.	Bureau central de mesures nucléaires, Steenweg near Retie, Geel, Belgium
Bambynek, W.	Bureau central de mesures nucléaires, B-2440 Geel, Belgium
Birkhoff, G.	C.C.R. EURATOM, Ispra, Italy
Casini, G.	C.C.R. EURATOM, 21020 Ispra (Varese), Italy
Koch, L.	Institut des Transuraniens d'EURATOM, Karlsruhe, Federal Republic of Germany

Nicks, R.	C.C.R. Ispra, Italy
Rief, H.	C.C.R. Ispra, Italy
Rustichelli, F.	C.C.R. Ispra, Italy
Spernol, A.	Bureau central de mesures nucléaires, Retieseweg, B 2440 Geel, Belgium
Vernimb, C.	Center for Information and Documentation of the CCE, Luxemburg

FORATOM

Grinberg, B.	Centre d'études nucléaires de Saclay, B.P. n°2, 91 Gif-sur-Yvette, France (see also France)

IAEA (International Atomic Energy Agency)

Chernilin, Y.F.	Department of Technical Operations, IAEA, Kärntner Ring 11, 1011 Vienna, Austria
Finkelstein, A.	Department of Research and Isotopes, IAEA, Kärntner Ring 11, 1011 Vienna, Austria
Górski, L.	Division of Research and Laboratories, IAEA, Kärntner Ring 11, 1011 Vienna, Austria
Richman, D.M.	Division of Research and Laboratories, IAEA, Kärntner Ring 11, 1011 Vienna, Austria

JINR (Joint Institute of Nuclear Research)

Nagy, A.Z.	Joint Institute of Nuclear Research, Dubna, USSR
Sodnom, N.	Joint Institute of Nuclear Research, Dubna, USSR

NEA (Nuclear Energy Agency)

Fröhner, F.	Centre de compilation de données neutroniques, 91190 Gif-sur-Yvette, France
Rosén, J.	OECD/NEA, 38 Blvd. Suchet, Paris 16e, France
Royen, J.	OECD/NEA, 38 Blvd. Suchet, Paris 16e, France

AUTHOR INDEX

Roman numerals are volume numbers. Arabic numerals underlined refer to the first page of a paper. Other Arabic numerals refer to a contribution to the discussions.

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