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Editor: D.B. Syme Nuclear Physics Division AERE Harwell, Oxfordshire April 1979

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## United Kingdom Atomic Energy Authority HARWELL

U.K. NUCLEAR DATA PROGRESS REPORT FOR THE PERIOD JANUARY-DECEMBER 1978

Editor D.B. Syme

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Nuclear Physics Division, AERE Harwell, Oxfordshire

April 1979

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#### PREFACE

This report is prepared at the request of the United Kingdom Nuclear Data Committee and covers the period from January to December, 1978.

Contributions on "Chemical Nuclear Data" are gathered by the Chemical Nuclear Data Committee and grouped under that heading. This report includes work from various collaborations between laboratories of Harwell, Dounreay, Winfrith, Windscale, Berkeley (CEGB), MOD Aldermaston, Imperial College and Manchester University.

Other nuclear data is presented by laboratory. There are contributions this year from the Harwell and Winfrith laboratories of the UKAEA, the National Physical Laboratory, the CEGB Berkeley Nuclear Laboratories, the University of Aston in Birmingham, the Birmingham 'Radiation Centre, the University of Birmingham and the University of Edinburgh.

Contributions to the report on nuclear data topics are welcome from all sources and we extend an invitation to researchers in other laboratories of industry, government, the universities and polytechnics to use this channel of communication.

Where the work is clearly relevant to requests in WRENDA 76/77 (INDC(SEC)-53/URSF) request numbers are given after the title of the contribution.

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#### COMMITTEE ACTIVITIES

#### United Kingdom Nuclear Data Committee (Chairman: Dr. J.E. Lynn)

The UKNDC met once during the period of this report, and continued in its role of coordinating work on nuclear data in the UK and providing the formal link with other national and international committees and organisations. Summary reports from the sub-committees which deal with detailed nuclear data needs are given below.

The highlight of the year was the International Conference on Neutron Physics and Nuclear Data for Reactors and other Applied Purposes which was held at Harwell from 25 to 29 September 1978. This inaugurated the series of triennial European conferences which will alternate with similar conferences in the USA and the USSR to provide the world community interested in nuclear data with annual meetings of a general nature. There were approximately 250 participants from many countries including those in the Warsaw Pact; the largest delegations came from the USA, France, the Federal Republic of Germany, the UK, and the Commission of the European Communities. There were 29 invited papers presented in plenary sessions and 109 contributed papers most of which were delivered in two parallel sessions.

Our thanks are due to the Programme Committee and their helpers for a successful conference, for participants made many favourable comments on the variety and interest of the presentations and the efficient organisation of the meeting.

The Research Division of the Central Electricity Generating Board acted as the host for the twelfth Nuclear Data Forum which was held on December 11th at the CEGB Headquarters in London. There were two themes this year: nuclear data files and reactor radiation dosimetry. A review paper was given on each theme and the authors have kindly allowed us to reproduce their presentations in this report. A full and interesting programme was completed with 15 contributed papers. The meeting was very well supported, there being 97 registrations and more proposed contributions than could be presented in the time available.

#### Neutron Sub-Committee (Chairman: Dr. J.E. Lynn)

The Committee did not meet during the period covered by this report. Nuclear Incineration Sub-Committee (Chairman: Dr. M.G. Sowerby)

The Nuclear Incineration Sub-Committee has not met during the past year as little work of interest is being performed in the U.K. When there is renewed interest in the nuclear incineration option for waste disposal, the Sub-Committee will be reactivated.

#### Fusion Sub Committee (Chairman: Dr. C.A. Uttley)

There was one meeting of the fusion sub committee during this period. One item of importance to nuclear data requirements for fusion blankets was a report on the calculations examining the influence of nuclear data uncertainties on heating rate estimates carried out by Birmingham University. Work in the USA along similar lines has been applied to tritium breeding, so that a clearer picture is now available of the accuracies needed on a number of important reaction cross sections.

## Chemical Sub-Committee (Chairman: Mr. J.G. Cunninghame)

The Chemical Sub-Committee of the UKNDC, is made up of measurers, evaluators and users of chemical nuclear data and is able to advise on measurements and to consider in detail reports of work on the compilation and evaluation of data in this field. The preparation of the CNDC nuclear data file is under the control of the Data File Sub-Committee whose chairman is Mr. B.S.J. Davies (CEGB, BNL). The main committee held two meetings and the sub-committee three meetings in 1978. The request list published early in 1977 is now being revised.

Work is in progress on very nearly all items on the list but often on a long time scale and with little prospect of early completion. Data File work has now reached the stage where data is available to the user and the status of the file is indicated in the relevant part of this report.

#### Bio-Medical Sub-Committee (Chairman: Mr. J.A. Dennis)

The Bio-Medical Sub-Committee did not meet during 1978. An article on available sources of nuclear and atomic data for bio-medical applications prepared by the Committee has been published in the February 1979 issue of Physics in Medicine and Biology.

#### NUCLEAR DATA. ITS EVALUATION AND DISSEMINATION

الدواح وجاولا B S J Davies Lecture given to the UK Nuclear Data Forum, CEGB Headquarters, Sudbury House, London, on the 11th December 1978 ٠×.,

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It is probably reasonable to assume that virtually everyone here today is a user of nuclear data. A number of us will use such data in vast quantities perhaps for designing reactor shielding or evaluating decay heat production from irradiated fuel. At the other extreme, some will need just a single number every now-and-then; perhaps the half-life of some particular nuclide or a thermal neutron cross-section. Maybe some others make use of standard computer programs where data is already built in, so that their status as nuclear data users is not so apparent. Even those doing fundamental measurements and those evaluating measurements need nuclear data for their work; you need such data to generate more of it. So it is clear that nuclear data usage is very widespread; the universities need it for fundamental research, it is needed in medicine, in many industrial situations and especially in the nuclear power industry by designers, operators, people concerned with health and safety and by researchers both basic and applied. Less obviously, it is also used by designers of coal cutting machines and geologists drilling bore-holes for oil exploration and so on. So we have all these people with a very broad spectrum of interests whose data requirements vary from a single number to very large computer files containing hundreds of thousands of numbers. This brings us directly to the heart of the problem about which I propose to talk. As I implied in the title, the problem really falls into two parts, namely evaluation and communication. Consider, for example, a researcher who desperately needs to know the best possible value of the half-life of  $^{137}$ Cs. If he spends a week searching the literature he will find maybe a couple of dozen values measured by various people. So he only has two problems, one is finding a spare week to do the search, the other is assessing all the measurements and extracting a best value. It will probably not please him to think that maybe half-a-dozen other people have recently been through the same exercise, because he has no way of knowing who they are. So this is where evaluation and dissemination play their roles, because evaluation is the job of finding and extracting the measurements, discarding the bad ones, distilling the rest to produce a best value and finally letting the world know what that value is.

In a recent article<sup>(4)</sup> in the journal 'Science', Professor Stockmayer of Dartmouth College, Hanover, New Hampshire, discussed the problems arising from the uncritical acceptance of bad scientific information. I quote:

'A particularly pernicious aspect of this problem involves numerical data, which are essential in all branches of science and technology and are often needed to arrive at valid operational decisions. Unfortunately, the scientific literature contains many

erroneous values. Few scientists or engin-

eers seem to have given much thought to the

magnitude of the problem, and some probably

regard every numerical entry in a handbook

as revealed truth. Yet anyone who has had

and searched out a dozen or more reports.

to seek a particular number in the literature

only to end up with a set of widely disparate



#### Fig. 1 Information flowpath for nuclear data

values, comes to realize that a substantial intellectual effort and a considerable background in the field are needed to arrive at reliable figures.'

Professor Stockmayer goes on to state that the costs of search and critical evaluation are small: less than 2% of that for the original research and about one quarter of the cost of initial publication. The benefits, however, are large, although it is often difficult to translate them into monetary units.

So what is needed is a flowpath for information from the researcher who makes the measurements to the user. The main features are shown in Fig 1.

Occasionally evaluators will be in direct contact with users but more usually the evaluated data will be published in some way or other and the user will get his information via this indirect means. Often it will happen that the evaluator will provide the data in a form such that the user cannot directly make use of it; in this case the dissemination stage will need to include some further processing.

Thus far I have spoken in general terms. I now propose to concern myself mainly with nuclear decay data, which serves as an excellent example of the problem where, hopefully, a solution has been found. The term nuclear decay data encompasses a very large quantity of information. So, how does all this evaluation get done? How is it organised? Well, until recently, it really was not properly organised at all and, in some respects, this is perhaps still so. Up till a few years ago, evaluation was carried out at a number of independent centres. This resulted in a number of publications from Nuclear Data Sheets and the various editions of the Table of Isotopes to an assortment of wall charts and a number of publications on specialised topics such as internal conversion coefficients, atomic mass





evaluations, and gamma rays in increasing order of energy. Recently the use of magnetic computer tape has grown rapidly and the current system relies heavily on this medium, largely replacing written documents.

The manner in which the evaluation effort has come to be organised is largely a product of the sheer volume of data which has to be handled. The first Table of Isotopes was published in 1940<sup>(1)</sup>, it was

17 pages long. Since that time, the quantity of information has grown explosively as is, shown in figure 2; the 7th edition will contain about 1200 pages covering about 2000 nuclides and, incidently, will be the last in the series. The most recent issue of the ENSDF contains 6320 data sets listed on over 4 million computer card images.

This rising tide of evaluation work was, until recently, handled largely by the Nuclear Data Project at ORNL. By about 1975 it was becoming clear that the whole thing was becoming too large for any one group to handle. This realisation lead to a meeting on Non-neutron Data Evaluation at Geel in early 1976 at which the Americans sought help from the international community. At a further meeting in Vienna shortly afterwards an agreement on international co-operation was reached. The general form of the structure



# Fig. 3 Structure for international co-operation on evaluation of non-neutron nuclear data

great deal of evaluation effort is also carried out. The international effort is largely within the box marked 'A-chain evaluators'. The main distribution centres are BNL, CAJAD (Moscow Institute of Atomic Energy), ZAED (Karlsruhe) and IAEA (Vienna). There are also a number of secondary centres, that of main interest to the UK being CCDN,

which was set up is shown in figure 3. The operation is centred at BNL (Brookhaven National Laboratory) which is acting

as a communications centre. All the basic computer files are housed and updated at ORNL, where, as I shall show shortly, a Gif-sur-Yvette, France.

The division of evaluation effort is shown in figure 4 (from ref 3). It is still mainly American but with contributions from 7 other countries. I have tried here to show the division of effort by drawing a single straight line to represent mass numbers from 1 to 250 and then chopping it in to 10 sections as shown. I have then marked the mass number ranges allocated to the various countries and the particular centres within these countries. The abbreviations which I had to include are: UP/University of Pennsylvania

NDP/Nuclear Data Project (Oak Ridge National Laboratory)

#### USSR US/UP 25 1 : NED/UTRECHT 50 26 UK/ DARESBURY + KUW / ISR US/NDP 51-75 FRG / ZAED 1100 76 US / NDP + SWD / LUND JAP/ JAERI 101 125 USSR US/NNDC US/LBL 126 150 US/INEL US/LBL 151 175 US/LBL 176 200 US/NDP 201 225 ISSP US/NDP 250 226 NDF

EVALUATION ALLOCATIONS

Fig. 4 Division of evaluation effort for non-neutron nuclear data between participant countries. The abbreviations are explained in the text

ISR/Kuwait Institute for Scientific Research ZAED/Fachinformationszentrum Energie, Physik, Mathematik, Karlsruhe JAERI/Jap. Atomic Energy Research Institute, Tokai-Mura NNDC/Nat. Nuc. Data Centre, BNL LEL/Lawrence Berkeley Lab., Univ. California INEL/Idaho Nat. Engineering Lab. Idaho Falls

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The UK contribution comes from Dr. Twin at Daresbury who is working in conjunction

with the Kuwait Institute.

The quantity of actual effort committed to doing this work is shown in Table 1 broken . ..

down by country. ્ય પ

· . . .

1.1

#### Table 1

#### Evaluation Assignments

Nation	No. of Chains	Manpower Commitment (PMY)
USA USSR Holland UK Germany (FR) Japan Sweden Kuwait	175 14 24 16 20 12	14 4 2/3 <sup>3</sup> 3 1 <sup>1</sup> 2 1



Fig. 5 Distribution of time of last evaluation for various mass chains

This, again, underlines the fact that the bulk of the work is still to be done in the United States, where 57% of the effort is being applied. Alternatively, if one does a quick count of the number of nuclides involved and divides by each nation's committed effort, one finds values ranging from 23 nuclides/man in Russia to 216 nuclides/man in Holland.

The international system is based on a number of large computer files. The first of these is the Nuclear Structure Reference File which contains keyworded bibliographic references. These are used both for publication of the 'Recent Reference' section of the journal Nuclear Data Sheets and for transmission to evaluators. Based on author produced keyworded abstracts (Nucl. Phys. and Phys. Rev. C) which were merged with NSR in 1973. Secondly, there is a set of Working Files which contain all relevant experimental and theoretical information for use by evaluators. The main working file is kept at ORNL, but others exist at the various evaluation centres. Thirdly there is the Evaluated Nuclear Structure Data File which is the basic file for distributing evaluated information. Lastly there is a variety of derived files such as the ENDF/B series of files and a number of printed publications of such things as gamma energies from fission products in increasing order of energy and tables of evaluation Atomic Masses and Spins and Moments.

This international system is aimed at carrying out evaluations of the decay properties of all known nuclides on a four year cycle. This is not as remote a possibility as one might imagine. There are still a few mass chains which have not been evaluated for about 10 years, but these are very much the exception as is shown in figure 5. This refers to the situation as it was in August this year. The mean age of the data at that time was 3.4 years compared with a target value of 2 years which the 4 year cycle requires. So, hopefully, all this international effort will result in the putting together of very large sets of evaluated data. It is, however, clear that this is not going to be of immediate help to our man sitting in his office wanting to know the half-life of  $^{137}$ Cs. Which brings us to the second question of how all this data is to be disseminated. The first stage is to distribute a copy of the latest ENSDF tape to the various data centres every 6 months. From here, copies will be sent to individidual laboratories on request. In parallel with this, a number of different hard copy routes are being used. The ENSDF file has been arranged so that the journal Nuclear Data Sheets may be printed directly from it, which is now being done. Next a new Handbook of Isotopes will be published in 1982; this will largely replace the Table of Isotopes but will be restricted to nuclides of practical importance. However, useful as these publications

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are, it has been shown that the most frequently consulted source of data is a wall chart of the nuclides. Accordingly a new General Electric chart has just been published and will be followed by another in 1981. There is also to be a new German wall chart from Karlsruhe in 1980. For those who need their data in larger quantities the routes for dissemination are not so clear. The latest version of the Evaluated Structure File is relatively easy to obtain, but extracting data from it in a usable form is not as easy as it may first appear. Computer programs to produce a readable listing and to convert the data into other formats (ENDF/B4 or B5, for example) exist in this country as well as the US and France, but running them is essentially a do-it-yourself job which is not so simple and straightforward as one might like. Apart from this, there is an ongoing effort in this country under the auspices of the Chemical Nuclear Data Committee which is aimed at regular updates of several specialised files of decay data, specialised that is, in the sense of containing data on specified classes of nuclide. These files are currently kept at Harwell, Winfrith and the CEGB computer centre and copies have been sent to other establishments. Hence at this level it comes down to a matter of obtaining data by private communication with the people concerned.

A useful bibliography of compilations and evaluations has recently been published by the International Atomic Energy Agency<sup>(2)</sup>.

I would like to conclude with a further brief quotation from Professor Stockmayer: 'Data evaluation is an unglamorous activity, unlikely to win Nobel prizes for its practitioners. Moreover, the very modesty of its cost tends to obscure its importance. It is thus not surprising to learn in the committee's report that current US activity is about one-third to one-half of that required to keep pace. If this situation is to change, a greater awareness of the need for and importance of data evaluation on the part of the scientific community seems essential.'

1. J.J Livingood, G.T. Seaborg, Reviews of Modern Physics, 12 (1940) 30

2. A. Lorenz, IAEA Report INDC (NDS)-98/LN, (1978)

3. A. Lorenz, IAEA Report INDC (NDS)-92/LN, (1978)

4. W.H. Stockmayer, Science 201, No.4 (1978).

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SOME REMARKS ON FAST REACTOR DOSIMETRY

J.E. Sanders

U.K.A.E.A., Reactor Physics Division, A.E.E., Winfrith. Lecture given at the Nuclear Data Forum, C.E.G.B. Headquarters, Sudbury House, London on 11 December, 1978

Reactor Dosimetry is concerned with the characterisation (spectrum, intensity, timedependence) of both in-core and external radiation fields with particular reference to their influence on the physical properties of materials. It ranges from the monitoring of basic irradiation experiments in MTR's to the long term surveillance of operating power plant and thus has close affinities with both core physics and shielding. Indeed in the UK, dosimetry has normally been dealt with under these headings rather than recognised as a separate activity. On the international scene, both Euratom and the IAEA have long-established working groups dealing with dosimetry matters and biennial Symposia are organised jointly by the EWGRD and the American Society for Testing and Materials (ASTM) - see References 1 and 2. ENDF/B includes a special Dosimetry File and Version 5, due for release shortly, is likely to be adopted as a reference for many detector cross-sections. Current interest is focussed on two aspects:-

- (i) the radiation-embrittlement of LWR pressure vessels. Some older American reactors may be faced with premature shut-down or long outage time for annealing, and improved standards and surveillance procedures are being called for by operators and regulatory bodies (see eg. Reference 3);
- (ii) the neutron damage to the components of sodium-cooled fast reactors, leading to phenomena such as void-swelling and irradiation-induced creep having a major influence on design and operation. Behaviour of fast reactor materials is investigated primarily by irradiation programmes in facilities such as PFR and, in due course, FFTF, where the stringent conditions in power reactors can be closely represented.

The present remarks are restricted to the dosimetry of in-core irradiations in fast reactors, in particular PFR. These will play a key role in evaluating fuel, clad, wrapper, and absorber materials for future commercial systems. PFR experiments involve both macroscopic studies (including bowing and dilation) on complete fuel elements, and smallerscale investigations using special 'demountable sub-assemblies' (DMSA) in which a number of thermally-independent clusters containing materials under test are enclosed within a normal hexagonal wrapper.

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The neutron interactions of relevance to materials behaviour have widely-differing energy dependence, and Fig. 1 shows the spectral response for some important cases. A knowledge of the spectrum at the irradiation position is thus an essential factor in accurate dosimetry. In earlier work damage to structural materials was often correlated against a simple parameter such as fluence above 0.1 or 1 MeV, but nowadays it is customary to adopt a more sophisticated approach based on cascade theory, the model of Norgett, Robinson and Torrens<sup>(8)</sup> being generally accepted for iron and other constitutents of reactor steels. Fig 1 includes the energy distribution of the damage calculated in this way with the FD5 displacement cross-sections. Helium production by (n,alpha) reactions is a possible cause of high-temperature embrittlement as well as influencing the incubation of void-swelling. Most of these reactions in structural materials have effective thresholds in the several-MeV range, and poorly-known cross-sections.





Target accuracies (one sigma) for the estimation of DMSA heater pin power (3% relative to core total), gamma-energy deposition (15%), fuel burnup (3%), displacement dose (10%), helium production (15 appm), and absorber heating and burnup (10%) have been indicated by the sponsors of PFR experiments. Note that gamma radiation is of importance in the present context because of its contribution to sample heating rather than as a source of material damage. No continuous monitoring of DMSA clusters is provided and temperatures, which need to be known to  $\pm 10^{\circ}$ C, are based essentially on calculation.

The first question to be asked is whether these accuracies can be achieved using the very extensive calculational and databanking framework that has been developed for PFR,

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assuming some means of absolute normalisation. Access to the core for detailed measurements is (as in all power reactors) extremely limited, and the main burden of validation rests on zero-power experiments in Zebra, which have included fairly close simulations of typical PFR loadings. The scope of this validation, covering neutron spectra, reaction-rate distributions in both normal fuel and singularities (such as control-rods), and gamma-ray energy deposition, has been summarised (Reference 4). It is concluded that in the majority



Fig. 2 The spectrum at the centre of Zebra Core 11 as measured by proton-recoil, 200-metre time-of-flight, and a doublescintillator spectrometer, and comparison with FGL5-MURAL calculation
of cases, the target accuracies are attainable by calculation using existing methods or
practicable refinements of them. As one example, Fig. 2 shows the neutron energy spectrum
in Core 11 (very similar to that in PFR) as measured by a combination of time-of-flight,
proton-recoil and scintillation spectrometers; the difference between calculation and
experiment amounts to a 2% change in the spectrum-averaged iron displacement cross-section.
As a second, Fig: 3 indicates the close agreement between predicted and measured
<sup>239</sup>Pu
fission-rate distributions in one version of the PFR mockup. Similarly for gamma-energy

-19-



Fig. 3 Comparison of predicted and measured channel power in PFR Mockup, Zebra Assembly 13. Values of C/E-1 in units of  $10^{-3}$  are indicated

deposition, Zebra work using thermoluminescent dosimeters has tended to confirm the adequacy of the photon source data currently in use (and better data is available), while the geometry of the DMSA may be accurately represented by Monte Carlo methods.

The neutron and gamma-ray fluxes can be established on an absolute scale by normalising the calculations to the total the heat output of the reactor as measured by a thermal balance technique, for which an accuracy of 4% is claimed. This involves computations on 3-D models of the core, taking into account day-to-day variations in power level, flux perturbations due to movements of control-rods

and possible re-locations and rotations if the irradiation is extended over more than one reactor run. Uncertainties in these procedures must be allowed for A retrospective check, albeit on a limited number of samples, is available from fuel burnup analysis by means of stable neodymium fission products.

The alternative (and traditional) approach to dosimetry is the use of activation detectors located near sample positions to provide a 'direct' measurement of fluence and a basis for normalising calculated spectra independent of the complexities of the thermal balance/calculation route. While this type of measurement is in principle straightforward, there are fairly severe constraints in practical application and very few detectors satisfy the criteria for energy-dependence, half-life of induced activity, absence of secondary reactions in capture products, and compatability with reactor environment. Dosimeter design must combine secure location during irradiation with ease of subsequent retrieval and measurement. Materials must be of accurately-defined composition and stable under prolonged exposure at temperatures of up to  $\sim 650^{\circ}$ C. Standards for absolute counting must be established. Development work in these areas in support of dosimetry of EBR2 and FFTF experiments has been extensively reported (see eg.Reference 5).

The total (energy-integrated) fluence is conveniently monitored by <sup>239</sup>Pu fission with its fairly constant cross-section over the range of interest, a long-lived fission

-20-

product (eg.  $^{137}$ Cs) being used for detection by non-destructive gamma-ray spectrometry. Additionally, some monitor of the high energy component is desirable to confirm the spectrally-dependent perturbations caused by control-rod movements. The  $^{58}$ Ni(n,p)  $^{58}$ Co (71 days) and  $^{54}$ Fe(n,p)  $^{54}$ Mn(312 days) reactions are obvious candidates; both respond predominantly (80%) to neutrons in the 2.5 - 7.5 MeV range. The cross-section for the  $^{58}$ Ni reaction is known to about 5% from threshold (values of a few microbarns) upwards (see eg. Reference 6), but the  $^{58}$ Co half-life is only marginally long enough for integrating over PFR irradiations (of  $\sim$  60 days). The  $^{54}$ Fe reaction is preferable from this aspect but its cross-section is somewhat less well determined. Fission reactions in  $^{237}$ Np and  $^{238}$ U, while much used as threshold detectors in low flux experiments, are less suitable for high power reactor applications because of uncertainties associated with the build-up and subsequent fission of  $^{238}$ Pu and  $^{239}$ Pu.

A reaction which has been much discussed for damage monitoring in power reactors is the inelastic excitation of the 29 keV niobium isomer,  $^{93}$ Nb (n,n')  $^{93m}$ Nb, which has a long half-life ( $\sim$  15 years) and a cross-section believed, from integral comparisons, to be similar to that of the analogous  $^{103}$ Rh (n,n')  $^{103}$ Rh reaction. The latter is extensively applied to zero-power and shielding experiments since it has a spectral response bearing some resemblance (although by no means identical) to that for iron displacements (see Fig. 4), but the short half-life of  $^{103}$ Rh (56 minutes) renders it unsuitable for long-term monitoring.



Fig. 4 Comparison of rhodium activation and iron displacement spectra in a fast reactor core

Methods for counting the soft radiations from <sup>93m</sup>Nb have been developed at Winfrith and trial irradiations have been successfully carried out in both PFR and SGHWR (Reference 7). The main obstacle to its adoption is the poor nuclear data, no differential cross-section measurements yet having been made in the region of interest.

In summary, the highly-developed calculational framework for fast reactor neutronics appears capable of meeting most requirements for dosimetry of PFR materials-irradiation experiments, assuming that an adequate procedure for absolute normalisation is established. Added confidence can be provided by the careful application of in-core fluence monitors although only a few suitable reactions have been identified. Some measurements of the  $^{93}$ Nb (n,n')  $^{93m}$ Nb cross-section would be valuable in this context. Other nuclear data needs for dosimetry include a further improvement of the  $^{54}$ Fe(n,p) $^{54}$ Mn cross-section and confirmation of the fission yields used for burnup measurements and  $^{239}$ Pu fluence monitors. Better information on many of the (n,alpha) reactions responsible for gas production in structural materials is also required for correlating damage phenomena.

- Proceedings of the First ASTM-EURATOM Symposium on Reactor Dosimetry (Petten, 1975). EUR 5667 e/f (2 vols)
- Proceedings of the Second ASTM-EURATOM Symposium on Reactor Dosimetry (Palo Alto, 1977). NUREG/CP - 0004 (3 vols)
- 3. W.N. McElroy et al, "Standardisation of Dosimetry and Damage Analysis Work for LWR, FBR and MFR Development Programs". Reference 2, Vol <u>1</u>, 17
- 4. J.E. Sanders, "The Zero-Power Basis of Fast Reactor Dosimetry". AEEW R 1183
- 5. E.P. Lipincott, J.A. Ulseth "High Flux-Fluence Measurement in Fast Reactors" Reference 2, Vol <u>1</u>, 271
- 6. D.L. Smith, J.W. Meadows, NSE 58, 314 (1975)
- 7. W.H. Taylor "Studies of the <sup>93</sup>Nb(nn')<sup>93m</sup>Nb Reaction". Reference 2, Vol 2, 831
- 8. M.J. Norgett, M.T. Robinson and J.M. Torrens, IAEA Meeting on Radiation Damage Units, Seattle (1972)

# CINDA LISTINGS <u>Ginni -----</u>

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ELEMENT			ENE	RGY	DO	CUMEN	TATION	ſ			
S A	QUANTITY	TYPE	MIN.	MAX.	REF	VOL	PAGE	DATE	LAB	COMMENTS	
Li	7	Ν, ΝαΤ	EXPT- PROG	4.7+6	1.4+7	UKNDC	P94	54	4/79	HAR	SWINHOE+ activation relative to $^{27}$ Al (n, $\alpha$ )
S	32	DIFF ELASTIC	EXPT- PROG	2.0+6	3.0+6	UKNDC	P94	97	4/79	BIR	JANICKI+ graphs given com pared with optical model
C1	35	N-PROTON	EVAL	-	-	UKNDC	P94	87	4/79	WIN	STORY data file prepared
Ċa	40	DIFF- ELASTIC	EXPT- PROG	2.0+6	3.0+6	UKNDC	P94	97	4/79	BIR	JANICKI+ graphs given com pared with optical model
Ar	40	N, GAMMA	EVAL	-	_	UKNDC	P94	87	4/79	WIN	STORY conversion of ENDL FLLB 7148
ΊĹ	47	N, PROTON	EXPT- PROG	4.7+6	1.4+7	UKNDC	P94	56	4/79	HAR	SWINHOE+ activation relative to $27A1(n,\alpha)$
Ti	48	N, PROTON	EXPT PROG	4.7+6	1.4+7	UKNDC	P94	56	4/79	HAR	SWINHOE+ activation relative to $2^7$ Al(n, $\alpha$ )
Cr 1	NAT	N,ALPHA	EXPT- PROG	+6	+7	UKNDC	P94	57	4/79	HAR	COOKSON+ preliminary measurement started
Cr	50	TOTAL XSEC	EXPT- PROG	+2	+5	UKNDC	P94	42	4/79	HAR	BOWEN+ measurement com- plete analysis in progres
Cr	· 50	N,GAMMA	E AL	-	-	UKNDC	P94	87	4/79	WIN	STORY data file prepared
Cr	52	TOTAL XSEC	EXPT- PROG	+2	+5	UKNDC	P94	42	4/79	HAR	BOWEN+ measurement com- plete analysis in progres
Cr	53	TOTAL XSEC	EXPT- PROG	+2	+5	UKNDC	P94	42	4/79	HAR	BOWEN+ measurement com- plete analysis in progres
Mn	55	N,GAMMA	EXPT- PROG	2.5-2	-	UKNDC	P94	88	4/79	WIN	TAYLOR obtained from check of detector system
Fe 1	NAT ,	TOTAL XSEC	EXPT- PROG	+2	+5	UKNDC	P94	42	4/79	HAR	BOWEN+ measurements com- plete analysis in progres
Fe 1	NAT	N,ALPHA	EXPT- PROG	+6	+7	UKNDC	P94	57	4/79	HAR	COOKSON+ preliminary measurements started.
Fel	NAT	N,GAMMA	EXPT- PROG	2.0+2	5.0+4	UKNDC	P94	36	4/79	HAR	GAYTHER+ capture yield data
Fe l	NAT	DIFF- ELASTIC	EXPT- PROG	2.9+6	-	UKNDC	P94	111	4/79	EDG	ANNAND+ polarisation and differential XSECT
Fe	56	TOTAL XSEC	EXPT- PROG	+2	+5	UKNDC	P94	42	4/79	HAR	BOWEN+ measurement com- plete analysis in progres
Fe	56	RES- PARAM	EXPT- PROG	1.1 3		UKNDC	P94	36	4/79	HAR	GAYTHER+ parameters of th 1.1 keV
Fe	57	TOTAL XSEC	EXPT- PROG	+2	+5	UKNDC	P94	42	4/79	HAR	BOWEN+ measurement com- plete analysis in progres

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ELE	FLEMENT			ENE	RGY	L	OCUME	NTATIO	N		
S	A	QUANTITY	TYPE	MIN.	MAX.	REF	VOL	PAGE	DATE	LAB	COMMENTS
Ni	NAT	TOTAL XSEC	EXPT- PROG	+2	+5	UKNDC	P94	. 42	4/79	HAR	BOWEN+ measurement com- plete analysis in progress
Ni	NAT	N,ALPHA	EXPT- PROG	+6	·+7	UKNDC	P94	57	4/79	HAR	COOKSON+ preliminary measurement started
Ni	58	RES- PARAM	EXPT- PROG	1.0+4	6.4+5	UKNDC	P94	42	4/79	HAR	SYME+ shape fit to trans- mission data
Ni	58	N, PROTON	EXPT- PROG	4.7+6	1.4+7	UKNDC	Р94	56	4/79	HAR	SWINHOE+ activation relative to $2^{7}A1$ (n, $\alpha$ )
Cu	NAT	DIFF÷ ELASTIC	EXPT- PROG	2.9+6	-	UKNDC	P94	111	4/79	EDG	ANNAND+ polarization and differential XSECT
Cu	NAT	DIFF- ELASTIC	EXPT- PROG	1.4+7		UKNDC	P94	110	4/79	EDG	BEGUM+ polarization and differential XSECT
NЪ	93	N,2N	EXPT- PROG	1.4+7		UKNDC	P94	91	4/79	NPL	RYVES+ work in progress
Rh	103	IN- ELASTIC	EXPT- PROG	FAST		UKNDC	P94	88	4/79	WIN	TAYLOR+ production of Rh-103 isomer
Ag	107	N,GAMMA	EXPT- PROG	MAX W		UKNDC	P94	88	4/79	NPL	RYVES+ production of Ag 108 isomer
I	127	DIFF- ELASTIC	EXPT- PROG	2.9+6		UKNDC	P94	111	4/79	EDG	ANNAND+ polarization and differential XSECT
Та	181	N,2N	EXPT- PROG	1.4+7	1	UKNDC	P94	91	4/79	NPL	RYVES+ work in progress
Ta	181	n, gamma	EXPT- PROG	2.5-2		UKNDC	P94	. 88	4/.79	WIN	TAYLOR+ obtained from check of detector system.
W	NAT	DIFF- ELASTIC	EXPT- PROG	2.9+6		UKNDC	P94	111	4/79	EDG	ANNAND+ work in progress
Au	197	N,2N	EXPT- PROG	1.4+7		UKNDC	P94	91	4/79	NPL	RYVES+ work in progress
Au	197	N,GAMMA	EXPT- PROG	2.4+2	2.0+3	UKNDC	P94	33	4/79	HAR	GAYTHER+ data used in resonance analysis
Au	<sup>`</sup> 197	RES PARAM	EXPT- PROG	2.4+2	2.0+3	UKNDC	P94	33	4/79	HAR	GAYTHER+ shape analysis of capture data
Hg	NAT	DIFF- ELASTIC	EXPT- PROG	2.9+6		UKNDC	P94	111	4/79	EDG	ANNAND+ polarization and differential XSECT
Τ1	NAT	DIFF- ELASTIC :	EXPT- PROG	2.9+6		UKNDC	P94	111	4/79	EDG	ANNAND+ work in progress
₽Ъ	NAT	DIFF- ELASTIC	EXPT- PROG	2.9+6		UKNDC	P94	111	4/79	EDG	ANNAND+ polarization and differential XSECT

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ELEN	MENT			ENE	RGY	T	OCUME	NTATIO	N		, ,
S	A	QUANTITY	TYPE	MIN.	MAX.	REF	VOL	PAGE	DATE	-LAB	COMMENTS
РЪ	NAT	DIFF- ELASTIC	EXPT- PROG	2.9+6		UKNDC	P94	. 111	4/79	EDG	ANNAND+ work in progress
РЪ	NAT	DIFF- ELASTIC	EXPT- PROG	1.4+7		UKNDC	P94	110	4/79	EDG	BECUM+ polarization work in progress
Bi	209	DIFF- ELASTIC	EXPT- PROG	2.9+6		UKNDC	P94	. 1,11.	4/79	EDG	ANNAND+ work in progress
Th	232	N, Fission	EXPT- PROG	TR	UP	UKNDC	P94	48	4/79	HAR	SYME+ measurement com- plete analysis in progres
U	NAT	DIFF- ELASTIC	EXPT- PROG	2.9+6		UKNDC	P94	-111	4/79	EDG	ANNAND+ work in progress
U	235	N,T	EXPT- PROG	PILE		UKNDC	P94	75 ,	4/79	HAR	MCKEAN+ tritium yield fro thermal fission
U	235	N,T	EXPT- PROG	FISS		UKNDC	P94.	75	4/79	HAR	MCKEAN+ tritium yield from fast fission
U	235	N, FISSION	EXPT- PROG	2.5-2		UKNDC	P94	88	4/79	WIN	TAYLOR+ obtained from check of detector system
U	235	FISS- YIELD	EXPT- PROG	FAST		UKNDC	P94	76	4/79	HAR	CUNINGHAME+ study effects of spectrum changes on yields
U .	235	SPEC. FISS.N	EXPT- PROG	9.4+5	6.0+6	UKNDC	P94	102	4/79	BIR	OWEN+ spectrum of delayed neutrons at given incident neutron energies
U	235	SPEC. FISS.N	EXPT- PROG	1.0+5		UKNDC	P94	48	4/79	HAR	ADAMS+analysis of data taken at STUDSVIK.
v	238	N, 2N	EXPT- PROG	1.4+7		UKNDC	P94	91	4/79	NPL	RYVES+ work in progress
U	238	FISS- YIELD	EXPT- PROG	9.0+5	1.7+6	UKNDC	P94	76	4/79	HAR	CUNINGHAME+ fission field of Mo-99
U	238	FISS- YIELD	EXPT- PROG	FAST		UKNDC	P94	76	4/79	HAR	CUNINGHAME+ study effects of spectrum changes on yields
Pu	239	Ν,Τ	EXPT- PROG	PILE		UKNDC	P94	75	4/79	HAR	MCKEAN+ tritium yield from thermal fission
Pu	239	Ν,Τ	EXPT- PROG	FAST .		UKNDC	P94	75	4/79	HAR	MCKEAN+ tritium yield from fast fission
Pu	239	N FISSION	EXPT- PROG	2.5-2		UKNDC	P94	88	4/79	WIN	TAYLOR+ obtained from check of detector system
Pu	239	FISS- YIELD	EXPT- PROG	FAST		UKNDC	P94	76	4/79	HAR	CUNINGHAME study effects of spectrum changes on yields
Am	241	N, GAMMA	EXPT- PROG	FAST		UKNDC	P94	82	4/79	HAR	GLOVER+ integral for fast reactor
Am ·	241	ALL	EVAL			UKNDC	P94	28	4/79	HAR.	LYNN+ T.B.P. aș AERE- R8528
Am	243	N, GAMMA	EXPT- PROG	FAST		UKNDC	P94	82	4/79	HAR	GLOVER+ products of Cm-24 for fast reactor spectrum

## 1. NUCLEAR PHYSICS DIVISION, AERE, HARWELL

(Division head: Dr. B. Rose\*)

#### Introduction

Nuclear data measurements in Nuclear Physics Division are diverse and are performed on a variety of sources. Since the results obtained from the various Harwell machines are not easily classified according to the energy of the charged beams, individual research items are as before labelled with a single letter indicating on which machine the experiments were performed. These labels are as follows:

Cockcroft-Walton Generator	A
3 MV pulsed Van de Graaff Generator IBIS	В
6 MV Van de Graaff Generator	С
14 MeV Tandem Generator	D
45 MeV Electron Linac	Ε
Variable Energy Cyclotron	G
Synchrocyclotron	Н

In the contents pages there is a cross reference to the accelerator where a measurement was made (when this applies).

The material for this contribution is taken from the chapter on Nuclear Data and Technology for Nuclear Power in the 1978 Nuclear Physics Division Progress Report AERE PR/NP 26 and is organised by subject material in the order: low energy neutrons and fission to high energy neutrons and fusion.

\* Acting division head in 1978: Dr. J.E. Lynn

#### 

1.1 The new machine for the electron linac laboratory (J. E. Lynn, M. S. Coates, P. P. Thomas and B. P. Clear)

It has not been possible to hold to the schedule outlined in PR/NP 25, p 9 and the revised estimate for the start of experimental operation with the new linac is now the early summer of 1979.

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Although all the new experimental buildings on the linac site were finished at the expected time the linac test programme carried out at the R.D.L. works at Swindon has taken longer than anticipated. It is now nearly complete, with all four modulator units successfully tried, and only the last of the 8 accelerator sections remains to be beam tested. Three of the modulators and five of the accelerator sections have been delivered to the Harwell site and installation is well under way. The remaining equipment will follow when the work tests are finished during January 1979. The final stages of installation and commissioning trials on the fully assembled machine will be carried out during the early months of 1979.

Essentially all of the magnets and quadrupole elements for electron beam handling are in position and ready for use with the exception of the pulse magnets needed for simultaneous (multiplexed) target operation. Conventional steady field magnets have been installed at the beam switching positions for the trial phase in the interest of simplifving beam transport tests. Some technical difficulties have been encountered, however, with the pulse magnets themselves. The field switching performance needed is proving difficult to achieve and the problems may not be solved in time for the units to be installed for the initial stages of experimental operation.

Technical difficulties in another area have resulted in delays in the manufacture of the tantalum targets for neutron production (see PR/NP 25, p 9). Cracking occurred in one of the faces of the beryllium containment vessel of the fast neutron target during the final stage of manufacture. The cause of this is now thought to be understood and it is hoped that the finished assembly will be delivered to Harwell during January 1979. Difficulties with the condensed matter target were all relatively minor, and delivery of this assembly is imminent.

Targets containing uranium are being produced as spares for both the condensed matter and the fast neutron cells. Metallurgy Division have overcome many of the problems (see PR/NP 25, p 9) connected with the production of Zircaloy clad uranium plates which

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will be used therefore in the spare condensed matter target. Difficulties still remain, however, in using tantalum as a cladding material and the spare fast neutron target will be made up from uranium plates inside helium filled tantalum cans. The helium is needed on heat transfer grounds. This target will only be able to dissipate 5 kW of electron beam power compared with 30 kW for an ideal target but most of the high resolution work using electron pulse widths in the 5-10 ns range can be carried out without restrictions at this lower power level<sup>(1)</sup>.

 M. S. Coates, P. P. Thomas, B. P. Clear, R. N. Sinclair and J. E. Lynn, A new electron linear accelerator at A.E.R.E., Harwell, paper presented at the IV National Soviet Conference on Neutron Physics, Kiev, USSR, April 1977

# 1.2 Evaluation of differential nuclear data for <sup>241</sup>Am (J. E. Lynn, B. H. Patrick, <u>M. G. Sowerby and E. M. Bowey)</u>

The evaluation of differential nuclear data for <sup>241</sup>Am has been completed and the file is now in the U.K. Nuclear Data Library. Documentation of the work exists in draft form and will shortly be sent for publication as report AERE - R 8528, with the following abstract.

'An evaluation of differential neutron data has been carried out for  $^{241}$ Am, the result of which has been incorporated into the U.K. Nuclear Data Library as DFN 1009A. The evaluation covers the energy range from  $10^{-5}$  eV to 15 MeV and includes the total, capture, fission, elastic and inelastic scattering, (n,2n) and (n,3n) cross-sections,  $\bar{\nu}$  and the fission neutron spectrum. Also discussed are the half-life and decay of  $^{241}$ Am, the branching of the radiative capture cross-section to form the ground and isomeric states of  $^{242}$ Am and resonance integrals. Where possible the evaluation is based on measured data but much use has been made of nuclear reaction theory and systematic behaviour of the nuclear parameters of the actinides to calculate the required data where no measurements exist. Uncertainties have been assigned at representative energies throughout the range. Some of the results of the evaluation are compared with data from other evaluations.'

#### 1.3E Capture cross-sections of structural materials [Relevant to request numbers: 184-193, 212, 589, 1383, 1384]

Preliminary results of neutron capture cross-section measurements made with a large liquid scintillator on the structural materials iron, nickel and chromium have been

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described in previous progress reports. During the past year much of the effort has been devoted to improving the methods of analysing the raw capture yield data and developing the computer program used for this purpose. The program REFIT<sup>(4)</sup>, which has been adapted to handle capture data and which is described elsewhere (p 23) in this report, is now being used exclusively to obtain the final neutron resonance parameters from the measurements. Described below are the calculation of two important properties of the large liquid scintillator required in the analysis and some examples of resonance parameters obtained.

### 1.3.1 Detector efficiency (R. B. Thom and B. W. Thomas)

In the previous report (PR/NP 25, p 26) a method of determining the efficiency of the large liquid scintillator for capture events was described which made use of the shape of the observed pulse amplitude spectra. The method has now been developed using a more mathematically precise characterisation of the amplitude spectrum. We call this characterisation parameter the fractional energy,  $E_{\rm F}$ , which is given by:

$$E_{\mathbf{F}} = \sum_{\mathbf{i}=\mathbf{L}}^{\mathbf{U}} \mathbf{A}_{\mathbf{i}} E_{\mathbf{i}} / \sum_{\mathbf{i}=\mathbf{L}}^{\mathbf{U}} \mathbf{A}_{\mathbf{i}}$$

where  $A_i$  is the number of events in channel i corresponding to gamma-ray energy  $E_i$ , and L and U are the lower and upper amplitude channel limits used in the data analysis. A wide variety of amplitude spectrum types, generated by the gamma-ray Monte Carlo code GAMOC (PR/NP 25, p 26) have been investigated and a correlation is found to exist between the fractional energy and the required efficiency. One such correlation curve for an excitation level energy of 8 MeV and lower amplitude limit of 3 MeV is shown in Fig. 1. The plotted points are well represented by a polynomial function of the type:-

$$\varepsilon_{\rm F} = \varepsilon_0 + \frac{3}{\sum_{\rm N=1}^{\rm \Sigma} \frac{\varepsilon_{\rm N}}{({\rm R}_{\rm F} - {\rm R}_{\rm O})^{\rm N}}} \qquad ({\rm R}_{\rm F} > {\rm R}_{\rm O}$$

where  $\varepsilon_{\rm F}$  is the efficiency,  $\varepsilon_{0,1,2,3}$  and  ${\rm R}_0$  are constants and  ${\rm R}_{\rm F}$  is the fractional energy  ${\rm E}_{\rm F}$  divided by excitation energy. The points marked Fe and Au on the figure apply to spectra calculated by GAMOC from cascade schemes typical of a medium mass and a heavy mass nucleus. The manner in which these schemes were generated is described in the

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Fig. 1 Correlation observed between the fractional energy and the detector efficiency for 30 pulse amplitude spectrum types calculated by the gamma-ray Monte-Carlo code GAMOC<sup>(1)</sup>. The full curve is a mathematical function (see text) which is fitted to the data points

previous report. The successful application of this method to these complex spectra of real nuclei is taken as an indication of its . validity.

Another approach to the problem of allowing for the energy dependence of the efficiency makes use of a pulse amplitude weighting technique first suggested by H. Maier-Leibnitz<sup>(1)</sup>. A channel weighting factor,  $W_i$ , is sought which produces a detector response of unity:

$$\begin{array}{c}
 U \\
 \Sigma \\
 i=L
 \end{array}
 \begin{array}{c}
 W \\
 ij
 \end{array}
 \begin{array}{c}
 u \\
 i=L
 \end{array}$$

where, as before, L and U are the pulse amplitude channel limits and A<sub>ij</sub> is the number of events in the i<sup>th</sup> channel for the j<sup>th</sup> spectrum type. Using the same set of amplitude spectrum types as before, a set of weighting factors, W<sub>i</sub>, was derived. These were applied to the Fe and Au spectra and

both responses were found to be within 2% of unity. This method, however, suffers from the disadvantage of being difficult to apply when admixtures of spectra with different isotopes are encountered.

1.3.2 Neutron sensitivity of the large liquid scintillator (R. B. Thom)

An essential property of a device whose principle of operation depends on the detection of gamma-rays emitted promptly following neutron capture, is that it should have a low probability of registering those events caused by neutrons scattered, rather than captured, by the sample and subsequently absorbed by the materials of the detector and its environment. This is especially important in the measurement of capture crosssections of medium nuclei where neutron scattering is typically three orders of magnitude

<sup>+</sup>Imperial College of Science and Technology

more probable than neutron capture. In such measurements using the time-of-flight method, one differentiates between prompt neutron events which occur within the timing resolution of the system and those detected later in time, usually after some degree of moderation in the detecting medium. The latter component contributes to a background which can be determined experimentally. The prompt component, however, is usually of small magnitude and is not clearly distinguishable from legitimate capture. Care was taken at the outset to construct the large liquid scintillator from materials of low capture cross-section and little or no resonance structure below a few hundred keV neutron energy (2). Nonetheless. when a sample of pure graphite was placed in the tank and exposed to a neutron beam which had passed through an aluminium background filter, slight but identifiable dips were present in the time-of-flight spectrum at the 'black resonance' energies in aluminium. Since capture in the graphite sample is negligible this indicates that a prompt background component was present. To substantiate these experimental observations and to understand . more fully the response of the detection system to neutrons, the neutron transport code MORSE<sup>(3)</sup>, originating from the Radiation Shielding Information Center at Oak Ridge, was adapted to meet the requirements of the problem\*. Based on 100 group cross-sections in ANISN format, the code applies the Monte Carlo method to the tracing of individual neutrons which start from a source energy group and whose histories terminate either upon absorption or when the particle escapes from the system. The individual absorptions are then summed and stored in appropriately chosen time and energy bins for each geometric region. The response of the system to gamma-rays from a particular nucleus is then determined with the gamma-ray Monte Carlo code GAMOC. This type of analysis was performed for several neutron energies and the resulting curve of neutron sensitivity (i.e. probability of detection per neutron scattered divided by an assumed average capture probability) for the pulse amplitude range used in most of the capture data analysis is indicated in Fig. 2.

The neutron sensitivity curve decreases smoothly with increasing neutron energy as expected owing to the absence of structure in the cross-sections of the tank materials. The main contribution to the sensitivity arises from capture in the 30 cm long, 1.2 cm thick  ${}^{10}$ B sleeve which surrounds the sample at the centre of the tank. This sleeve was

\*The help and guidance of Mr. J. G. Needham of Materials Physics Division in the application of MORSE is gratefully acknowledged.

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Fig. 2 Neutron sensitivity of the Harwell large liquid scintillator as calculated by the neutron transport code MORSE(4), compared with published calculations<sup>(5)</sup> for two other detection systems recently engaged in capture cross-section measurements

originally installed to protect the capture sample from neutrons returning after moderation in the scintillant. Although the principal absorption mechanism in <sup>10</sup>B is the (n, $\alpha$ ) reaction, an estimated one in eight thousand events will be a neutron capture with the subsequent release of ~ 11.5 MeV in gamma-ray energy. This fraction of neutron captures is the ratio of the (n, $\gamma$ ) to (n, $\alpha$ ) cross-sections at thermal energy as given in BNL-325 (third edition) and is subject to an uncertainty of 40%. It was assumed that this ratio remains constant for neutron energies up to around 400 keV where sizable resonances start to contribute to the <sup>10</sup>B cross-section. The only other non-negligible contribution to the neutron sensitivity arises from the Be tube which passes through the tank itself, but this is only significant at higher energies where the sensitivity is small. From the graphite sample experiments mentioned above a figure of 10<sup>-4</sup>, with an estimated error of 50%, was deduced for the neutron sensitivity in the 25-100 keV region. This is somewhat higher but not in disagreement with the calculated sensitivity. Fig. 2 shows that the Harwell detector compares favourably with two of the more important detection systems recently engaged on similar capture cross-section programmes.

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### 1.3.3 Resonance parameters of gold (D. B. Gayther, M. C. Moxon, B. W. Thomas, R. B. Thom and J. B. Brisland) [Relevant to Wrenda request number: 589]

Capture cross-sections measured with the large liquid scintillator are normalized to the average capture cross-section of gold in the energy region 25 to 35 keV (assuming a value of 589 mb). The measurements obtained with the standard gold sample, which provide the normalization, have recently been analyzed with the shape analysis program REFIT<sup>(4)</sup> (see also p 23 of this report) to derive resonance parameters in the energy region from ~ 240 eV to ~ 2 keV. The parameters determined so far are in good agreement with previously published data which were mostly derived from transmission measurements and hence, unlike the capture data, do not depend on absolute normalization. This result provides a valuable confirmation of the measurement of the shape of the neutron spectrum incident on the sample and verifies assumptions concerning the efficiency of the large liquid scintillator for detecting capture events. For those resonances with neutron widths greater than 100 meV, the radiation width can be accurately determined, and in the energy region below 1 keV its average value is found to be 125 meV.

Figure 3 shows examples of fits to the gold capture yield data. In Fig. 3(a) which covers the energy region from 1840 to 1930 eV, the present measurements are not capable of resolving all the resonances given in the third edition of BNL-325. Nevertheless, by determining only the resonance energies and the smaller of the two partial widths, reasonable fits to the data are obtained. In the well resolved resonance at 240 eV (Fig. 3(b)) it was found possible to determine the resonance energy and both neutron and radiation partial widths from capture data on one sample to statistical accuracies approaching those of previously published parameters using multi-sample data. Provisional resonance parameters found by least squares fits to the data of Fig. 3 are given in Table 1.

At the lower energies, clusters of data points are often found to lie above the calculated curve, as for example in the energy regions 236.5 to 237.5 eV and 244 to 245 eV of Fig. 3(b). Although the deviation of individual points from the curve may not be statistically significant, fits to some of the clusters produce significant neutron widths.

<sup>†</sup>Imperial College of Science and Technology



Fig. 3(a) Least squares fit to capture yield data for gold covering the energy region from ~ 1840 to 1930 eV



Fig. 3(b) Least squares fit to the capture yield data in the energy region around the 240 eV resonance. (Note the change from linear to logarithmic scales at the first decade of the ordinate)
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		•			
Resonance energy (eV)	Neutron width (meV)	Radiation width (meV)	x <sup>2</sup>	ND*	$\chi^2/df^+$
$1913.1 \pm 0.1$ $\{1913.6 \pm 0.7\}$	(1840)	123.6 + 7.9	)		
$1887.1 \pm 1.1 \\ \{1893.3 \pm 0.7\}$	$3.1 \pm 1.6 \\ \{6.1 \pm 1.1\}$	(124)	)		
$1883.0 \pm 0.1$ {1883.6 ± 0.7}	(170)	155.7 <u>+</u> 14.9	) 27.8	30	1.46
$1860.2 \pm 0.2 \\ \{1860.0 \pm 0.7\}$	80.6 <u>+</u> 13.0 {86.4 <u>+</u> 18.7}	(124)	) ) )		
$1856.3 \pm 0.3$ $\{1856.3 \pm 0.7\}$	(1040)	137.8 <u>+</u> 20.0	)		
$240.86 \pm 0.003 \\ \{240.5 \pm 0.5\}$	77.05 <u>+</u> 0.95 {72 <u>+</u> 4}	$122.8 \pm 3.3$ {100 ± 15}	· 100 <b>.</b> 0		1.03

### Provisional resonance parameters in gold determined from-

least squares fit to data shown in Fig. 3

partial widths in parenthesis () are unchanged in the fit the quoted uncertainties are from the least squares fit only parameters in parenthesis {} are taken from the 3rd Edition of BNL-325 \*number of data points

<sup>+</sup>chi-squares divided by the degrees of freedom, i.e. number of data points minus the number of parameters

### 1.3.4 Resonance parameters of iron (D. B. Gayther, M. C. Moxon, B. W. Thomas, R. B. Thom<sup>+</sup> and J. E. Jolly)

The capture yield data obtained with a 2 mm sample of natural iron have been analysed with REFIT<sup>(4)</sup> to obtain first values for the parameters of the main resonances up to a neutron energy of 50 keV. Better values for the resonance parameters are now being determined from a simultaneous shape fit to all the data obtained with the three sample thicknesses. The resonance parameters in  $^{56}$ Fe at 1.15 keV have important implications for the Doppler effect in reactor systems; the simultaneous fit for this resonance is shown in Fig. 4 and the parameters are listed in Table 2. Experimental uncertainties in the parameters are given in the table. It was found that a change of 10% in the Doppler width had a negligible effect on the partial widths obtained with simultaneous analysis, but such a change had a significant effect on the parameters obtained from the analysis of data from a single sample.

## Table 2

# Resonance parameters obtained for the 1.15 keV resonance in For

from a simultaneous shape analysis fit to capture yield data

obtained with three samples

	•		
•			

	(1)	(2)	(3)	(4)	(5)
Resonance energy (eV)	1152.69	<u>+</u> 0,007	<u>+</u> 0.0	<u>+</u> 0.0	<u>+</u> 1.2
Neutron width (meV)	73.93	<u>+</u> 0.45	<u>+</u> 12.9	<u>+</u> 0.0	0.0
Radiation width (meV)	577.2	<u>+</u> 5.9	+ 8.9	<u>+</u> 2.0	0.0

- (1) the values of the parameters determined in a least squares fit to measurements on three samples with five sets of data covering a neutron energy range from 912 to 1530 eV
- (2) the uncertainty obtained from the least squares fit
- (3) the uncertainty in the parameters associated with a 15% uncertainty in the normalisation of the original data
- (4) the uncertainty associated with the resolution function. The pulse width and target decay were determined from fits to narrow resonances in the energy region between 5 keV and 50 keV, and errors in other parameters associated with the resolution function were assumed to have a negligible effect on the resonance parameters
- (5) the uncertainty of  $\pm$  0.05 m in the length of the flight path produces an error of  $\pm$  1.2 eV in the resonance energy and only a negligible effect on the other parameters

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- H. Maier-Leibnitz, private communication to R. L. Macklin and J. H. Gibbons (Phys. Rev. <u>159</u> (1967) 1009)
- (2) D. B. Gayther, Report AERE R 8082 (1975)
- (3) MORSE-CG, 'General Purpose Monte Carlo Multigroup Neutron and Gamma-Ray Transport Code with Combinatorial Geometry', RSIC Computer Code Collection-203
- (4) "Neutron Data of Structural Materials for Fast Reactors", Proc. of a Specialists' Meeting, CBNM, Geel, December 1977, K. H. Bockhoff (Editor) Pergamon Press, Oxford, 1978

# 1.4 <u>REFIT - A least squares fitting program for resonance analysis (M. C. Moxon and D. A. J. Endacott)</u>

The program REFIT<sup>(1)</sup> has been modified during the past year to allow its use for the simultaneous analysis of neutron capture yield and transmission data. Multi-level R-matrix resonance parameters are adjusted so that the calculated transmission and/or capture yield agree with the observed data within the limit of the errors. Fits to several sample thicknesses of different isotopic composition can be carried out at the same time. The nuclear cross-section for each spin of each nucleus is calculated from initial parameters using an R-matrix multi-level formalism. The cross-section is then Doppler broadened assuming an ideal gas model with a given effective temperature. The transmission for a given sample is then calculated from the cross-sections and given abundances for the nuclei. When capture data are present, the capture yield includes the effects of multiple collision of the neutrons in the sample. The theoretical curves are folded into the resolution function and compared with the observed data. The Harwell computer library minimising routine VAO2A is then used to deduce the parameters from the best fit to the data. One of the main problems in extracting resonance parameters from neutron capture data is the large amount of computing time required to calculate the capture yield due to neutrons which are initially scattered and then captured on subsequent collisions. In REFIT, various simplifying assumptions are made in the steps taken to calculate the contribution to the capture yield from neutrons which have scattered more than once in the sample. As a result resonance parameters can be extracted using significantly less computer time than with nominally more rigorous programs, without any significant sacrifice in accuracy. The procedures followed in REFIT are now outlined.

The capture yield from multiple interactions of neutrons in the sample is divided into two parts. The first is due to neutrons initially scattered and then captured on the second interaction, and the second is the sum of capture events occurring in

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subsequent interactions.

The escape probability for neutrons initially scattered is calculated as a function of the scattering angle. This calculation takes into account the effects of finite sample size and Doppler effects. The fractions of neutrons that are either captured or scattered again are calculated from the number of neutrons that interact for a second time in the sample. Up to this stage the treatment is that used in conventional Monte Carlo programs and is essentially exact.



Fig. 5 Comparison of calculation of the capture yield for a resonance in <sup>56</sup>Fe at 27.7 keV <u>assuming</u> a neutron width of 1.5 keV and a radiation width of 1.5 eV. The sample diameter was 41.8 mm with a thickness of 13 mm (0.1 atoms/barn)

For the next part of the calculation it is assumed that neutrons which scatter a second time in the sample (1) do so isotropically in the laboratory system, (2) are distributed uniformly throughout the sample and (3) have an energy distribution covering the range allowed by elastic scatter. The probability of escape from the sample is then calculated from equation 6, page 23 of reference 2 using the appropriate cross-section, and hence the fractions of neutrons captured and scattered in the sample at this stage are

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found. The process is repeated on this and following generations of scattered neutrons until the fraction of neutrons remaining in the sample is less than 1% of the capture yield. The total capture yield is found by summing the capture contributions (including of course the primary capture yield) arising at successive stages of the full calculation.

A resonance in <sup>56</sup>Fe has been used to compare calculations carried out with three computer codes on samples up to a thickness of 0.1 atoms/barn. The resonance was assumed to have an energy of 27.7 keV, a neutron width of 1.5 keV and a radiation width of 1.5 eV. Figure 5 shows results obtained with REFIT, the Harwell Monte Carlo program, and the Monte Carlo program used at Lucas Heights, New South Wales, Australia by Allen et al<sup>(3)</sup> for a sample of thickness 13 mm (0.1 atoms/barn) and 41.8 mm in diameter. All three calculations are in good agreement even for this thickest sample where multiple scattering effects in the region above the resonance energy are very large. Table 3 gives a comparison of the areas obtained by summing the calculated data over the energy range 20 to 35 keV.

Program	Sample radius (mm)	Neutron beam radius (mm)	S th (mm)	ample ickness (atoms/b)	Capture area between 20 and 25 keV (b-éV)
Lucas Heights Monte Carlo	20.9	20.9	13	0.1	200.9
Harwell Monte Carlo	π	11	<b>17</b> .1		198.9 <u>+</u> ~ 1.0
REFIÍ	tt	11	, t f	".	196.5 <u>+</u> ~ 6.0
Harwell Monte Carlo	$2.09 \times 10^6$	2.09 x $10^6$	11		261.2 <u>+</u> ~ 1.5
REFIT	1 1 1 1 1 1 1 1 1 1 1 1 1 1 1 1 1 1 1			11 II	263.2 + ~ 5
1	· · · · · · · · · · · · · · · · · · ·				

	Table	3		
			• ,	

Calculated capture areas

Resonance parameters <u>assumed</u> in all the calculations  $E_R = 27.7 \text{ keV}$ 

 $\Gamma_{n} = 1.5 \text{ keV}$  $\Gamma_{\gamma} = 1.5 \text{ eV}$ 

The uncertainties on the Harwell Monte Carlo program are derived from the statistical spread in the calculation and in REFIT from estimates of the uncertainties in the various approximations made in the calculation. The computer times required to calculate the curves shown in Fig. 5 are 40 seconds for REFIT, 123 seconds for the Lucas Heights Monte Carlo program and 303 seconds for the Harwell Monte Carlo program.

Figure 6 shows the REFIT calculation for the thickest sample carried out assuming different sample and neutron beam diameters. Evidently the capture yield depends on these parameters as well as the sample thickness. This suggests that in the analysis of capture data, especially thick samples, effects due to sample size and non-uniform illumination should be taken into account.



Fig. 6 Calculation using REFIT with the same parameters as in Fig. 5 with a sample diameter of (a)  $4.18 \times 10^6$  mm and (b) 41.8 mm and a thickness of 13 mm

The program is used in the analysis of the neutron capture data for structural materials obtained with both the large liquid scintillators and the Moxon-Rae detector (see p 18 of this report). Analysis of the capture measurements carried out on the

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gold sample used as the standard for the measurements on the structural materials has already started and covers the neutron energy range from 200 eV to 2000 eV.

 M. C. Moxon, REFIT A least squares fitting program for resonance analysis of neutron transmission data, Proc. Conf. on Neutron Data of Structural Materials for Fast Reactors, Geel, December 1977 Published as "Neutron Data of Structural Materials for Fast Reactors", K. H. Bockhoff (Editor), Pergamon Press, Oxford (1978)

(3) B. J. Allen et al, private communication (1978)

1.5.H Neutron transmission measurements on the synchrocyclotron (P. H. Bowen, A. D. Gadd, G. D. Jones, D. B. Syme and I. L. Watkins) [Relevant to request numbers: 154-8, 165-7, 208, 212-14, 222, 256, 261]

Twenty transmission measurements on structural materials have been carried out by neutron time-of-flight experiments during the year. The basic parameters for each of these measurements are given in Table I. It will be seen that the samples measured include separated isotopes of Cr and Fe. These were obtained in a joint loan to Harwell and CBNM from ORNL. Measurements were also made using samples of natural iron and nickel in the presence of a cobalt filter designed to provide an in-situ measurement of the background. Similarly a sodium fluoride filter was used during two of the measurements, on  ${}^{50}\text{Cr}_20_3$  and  ${}^{52}\text{Cr}_20_3$ . Since September the data are being processed in a new way; now the transmission for each component run is calculated and these are combined to derive the final value of transmission and its internal error together with an external error deduced from the variation from run to run. From run number 17 onwards the data have been taken with the Camac incrementing store (PR/NP 25, p 156). This has removed the count rate limitation imposed when data were taken by means of a computer programme with spectrum sorting in core and storage on disc.

# 1.6.H Resonance parameters of <sup>58</sup>Ni from analysis of transmission data (D. B. Syme and P. H. Bowen) [Relevant to request numbers: 256, 233-8]

#### 1.6.1 Introduction

In support of the programme to determine the capture cross sections of the major structural materials used in fast reactors, many neutron transmission data have been obtained on the synchrocyclotron<sup>(1)</sup>. Progress in analysis of these data for resonance parameters in the case of the nickel isotopes was described in PR/NP 25, p 36. For the main isotope, <sup>58</sup>Ni, the upper limit of our analysis has since been extended from 300 keV

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<sup>(2)</sup> K. M. Case, F. De Hoffmann and G. Placzek, Introduction to the theory of neutron diffusion, Los Alamos Scientific Laboratory (1953)

Sequence number	Sample material	Thickness atom/barn	Path.(1) (m)	Channel width ( ns)	Channel structure(2)	Run number	Pulses <sup>(3)</sup> (10 <sup>5</sup> )
36	<sup>53</sup> Cr <sub>2</sub> 0 <sub>3</sub>	0.1189	50	5	a	7298	189.4
37	<sup>52</sup> Cr <sub>2</sub> 0 <sub>3</sub>	0.1189	100	2.5	ь	8199	405.3
38 5	<sup>50</sup> Cr <sub>2</sub> 0 <sub>3</sub>	0.07773	100	2.5	b.	8299	537.5
39	<sup>50</sup> Cr <sub>2</sub> 0 <sub>3</sub> (NaF)	0.07773	100.	2 <b>.</b> 5 ′	Ъ	8294	147.8
40 <sup>.</sup>	<sup>52</sup> Cr <sub>2</sub> 0 <sub>3</sub> (NaF)	0.1189	100	2.5	Ъ	8293	169.1
41	<sup>50</sup> Cr <sub>2</sub> 0 <sub>3</sub>	0.03178	100	2.5	Ъ.	8298	300.7
42	<sup>52</sup> Cr <sub>2</sub> 0 <sub>3</sub>	0.03145	100	2.5	ъ	8297	195.1
43	<sup>52</sup> Cr <sub>2</sub> 0 <sub>3</sub>	0.03145	50	5	с	8296	147.1
44	<sup>50</sup> Cr <sub>2</sub> 0 <sub>3</sub>	0.03178	5 <u>0</u>	5	С	8295	204.4
45	<sup>57</sup> <sub>Fe2</sub> <sup>0</sup> 3	0.02977	50	5	с	8999	(4)
46	<sup>57</sup> Fe2 <sup>0</sup> 3	0.02977	50	5	с	8998	(4)
47	<sup>57</sup> Fe2 <sup>0</sup> 3	0.02977	50	5	с	8099	(4)
48	2mmFe(Co)	0.01721 <sup>(5)</sup>	50	5	с	8065	(4)
49	6mmFe(Co)	0.05164 <sup>(5)</sup>	50	5	с	8117	. (4)
50	6mmNi(NaF)	0.5480 <sup>(5)</sup>	50	5	с	8149	(4)
51	3mmNi(NaF)	0.2740 <sup>(5)</sup>	50	5	с	8150	(4)
52 .	<sup>56</sup> Fe2 <sup>0</sup> 3	0.03032	50	5	с	8199	(4)
53	<sup>56</sup> Fe <sub>2</sub> 0 <sub>3</sub> (A1Mn)	0.03032	50	5	с	8198	(4)
54	<sup>53</sup> Cr <sub>2</sub> 0 <sub>3</sub>	0.08877	50	5	с	. 8399	(4)
55	<sup>53</sup> Cr <sub>2</sub> <sup>0</sup> 3	0.004902	50	5	с	8398	(4)

Table 1 List of transmission measurements made in 1978

Notes

All 50m runs are taken with a <sup>6</sup>Li-glass detector. All 100m runs are taken with 1. an NellO detector

2. The channel structure implies a doubling of channel width at the following channels

- (a) 13000, 23000
- (b) no doubling (c) '16384, 2476
- 3. The number of pulses is for the sample IN run only. The number of pulses in the sample OUT run is adjusted roughly to equalize the total number of counts in the two spectra. Experiments at 100m are performed at 800pps; those at 50m are performed at 400pps

Processing for these runs is not complete and the run numbers given are proposed 4.

5. Approximate thicknesses



FINAL FIT

0.678E 03

0.654E 03 0.666E 03



to 1000 keV, in order to increase the statistical sample of individual resonances and the accuracy of average parameters. This work was reported at the September Harwell conference<sup>(2)</sup>, and some of the more salient points are given below.

## 1.6.2 Shape analysis for resonance parameters

Resonance parameters were obtained by shape analysis <sup>(3)</sup> for 62 l = 0 and 228 l > 0resonances below 1000 keV. The accuracy diminishes above 650 keV due to computational problems associated with the increasing number of visible levels, their increasing overlap and the need to treat a wide energy range because of resonance-resonance interference. The analyses of these were carried out mainly to reduce edge (distant level) effects for the 41 l = 0 resonances observed in the 10-640 keV range. Fits to the 290-375 keV and 594-678 keV regions are shown in Figs. 7 and 8. The clear pattern of s-wave resonances of Fig. 7 have given way to a complicated picture of overlapping and interfering components in Fig. 8 where the quality of fit is also limited by uncertainties in the resonance parameters for regions of even higher energy.

#### 1.6.3 Resonance parameter distributions and average values

The observed spacing graph for s-wave levels (Fig. 9) is linear in first order to





about 800 keV. The strength function plot (Fig. 10) is linear to about 900 keV. The apparent structure corresponds to the larger resonances and is not significant under statistical tests for intermediate structure. the distribution of reduced widths in the 0-640 keV range (Fig. 11) follows the expected Porter Thomas<sup>(4)</sup> form (curve) with an observed mean of 4.31 eV and about 5 missed levels of low width.

With corrections for missing levels, our average parameters are compared with those of BNL  $325^{(5)}$  and Frohner's recent review<sup>(6)</sup> in Table 4. Our mean spacing in the 0-640 keV range is smaller than in the 0-418 keV range where it agrees better with Frohner's value. This is a manifestation of



DISTRIBUTION OF REDUCED WIDTHS



Fig. 10 Strength function graph for l = 0 resonances observed in 58 Ni transmission data



the slight increase we observed in the density of levels from 0-600 keV as expected over such a wide energy range. The gradual fall in density above this energy is probably due to increasing losses of small levels. The strength functions are all in satisfactory agreement.

#### Table 4

	<u> </u>	Carl State State State		<u> </u>	and the second second
	Source	Energy rangè	No. of levels observed	So (10 <sup>4</sup> )	Do (keV)
·.·	Present work	0 - 640	41	2.9 <u>+</u> 0.6	14.6 <u>+</u> 0.4
,	Present work	0 - 418	23	$3.1 \pm 0.9$	15.5 <u>+</u> 0.7
	Frohner <sup>[11]</sup>	0 - 400	20	3.2 <u>+</u> 1.0	16.7 <u>+</u> 1.6
	BNL 325 <sup>[5]</sup>	0 ~ 600	31	3.1 <u>+</u> 0.8	(20)
		f =			

 $\frac{58}{\text{Ni:}}$   $\ell = 0$  Statistical resonance parameters.

1.6.4 Future work

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The present analysis of <sup>58</sup>Ni will be completed soon and that of the natural data will continue. Development of the computer code to handle larger arrays and of special interactive programming to speed job preparation will be necessary to cope with the vast amount of data to be analysed in the isotopes of the structural materials.

- G. D. James, D. B. Syme, P. H. Bowen, A. D. Gadd and I. L. Watkins, "Neutron Data of Structural Materials for Fast Reactors", K. H. Bockhoff (Editor) (Pergamon Press, Oxford, 1978)
- (2) D. B. Syme and P. H. Bowen, in "Neutron Physics and Nuclear Data", (NEA, Paris, 1979) 319
- (3) M. C. Moxon, in "Neutron Data of Structural Materials for Fast Reactors", ibid.
- (4) C. E. Porter and R. G. Thomas, "Fluctuations of Nuclear Reaction Widths" Phys. Rev. Vol 104, No. 2 (1976) 483
- (5) S. F. Mughabghab and D. I. Garber, "Neutron Cross Sections Volume 1, Resonance Parameters", BNL325, Third Edition, Vol 1, p28 [EANDC (US) 183/L, INDC (USA)-58/L]
- (6) F. H. Fröhner, in "Neutron Data of Structural Materials for Fast Reactors", ibid.

1.7.H

Structure in the <sup>232</sup>Th neutron fission excitation function by observations of fission neutrons (D. B. Syme, G. D. James and M. C. Cooke<sup>+</sup>) [Relevant to request numbers 617, 618, 620, 621]

Recent subtreshold fission cross-section data have indicated structures characteristic of rotational and vibrational states in  $^{232}$ Th and  $^{231}$ Pa $^{(1,2)}$  which may be associated with shallow third minima in the region of the second saddle point of the fission barrier for the light actinides $^{(3)}$ . We have attempted to confirm the existence of the fine structure in  $^{232}$ Th by the alternative technique of fission neutron detection. This allows large samples and hence long flight paths and good resolution in a neutron time of flight spectrometer, although it is not the variation with energy of  $\sigma_{f}$  the fission crosssection which is observed, but of the product  $\sigma_{f}.\overline{\nu}$ , where  $\overline{\nu}$  is the number of neutrons released per fission. We have completed an experiment of this kind on the Harwell Synchrocyclotron using a natural thorium disc target at 97.48m from the pulsed source and resolution of 0.1ns m<sup>-1</sup>.

The data analysis is still in progress. At this stage we can confirm the broader structures previously observed but can not immediately verify all the previously claimed fine structure.

- (1) J. Blons, C. Mazur and D. Paya, Phys. Rev. Lett., 35 (1975) 1749
- (2) A. Sicre et al, "Physics and Chemistry of Fission", Rochester (1973) (IAEA, Vienna, 1974) Vol I, p71
- (3) P. Moller and J. R. Nix, ibid. p103

# 1.8 <u>Measurement of the fast fission neutron spectrum of <sup>235</sup>U at 0.1 MeV incident neutron</u> <u>energy (J. M. Adams, L. Eriksson\* and B. Trostell\*)</u>

The completion of the analyses of the experimental data obtained at Studsvik (PR/NP 25, p 43) has been delayed due to the lack of available effort. The fast fission neutron time-of-flight spectrum obtained for one of the two identical detectors employed for these measurements is shown in Fig. 12. A specially modified version of the Monte Carlo multiple scattering correction code, MAGGIE<sup>(1)</sup>, has been employed to correct the data for the finite size of the <sup>235</sup>U sample used. This has resulted in the magnitude of

Queen Mary College, University of London Present address: Plèssey Telecommunications Ltd., Edge Lane, Liverpool

\*AB Atomenergi, Studsvik, Sweden



the correction not being as great below ~ 1 MeV as that predicted by a simple first order analytical approach<sup>(2)</sup>. However, it has to be pointed out that this analytical approach is really only valid in situations where the overall correction is small which is not the case below ~ 1 MeV. Although a Watt description provides a better representation of the fast fission neutron spectrum as compared to a Maxwell description, there do still remain 'fluctuations' in the data when plotted as the ratio of  $N(E)_{EXPT}/N(E)_{FIT}$  as a function of E, the fission neutron energy, where N(E) is the number of fission neutrons per unit energy interval. In an attempt to obtain a better mathematical representation, further fits to the data are in progress using a description which includes a simple 'evaporation spectrum' term to account for the 'scission' neutron component.

J. B. Parker, J. H. Towle, D. Sams and P. G. Jones, Nucl. Instr. and Methods, <u>14</u> (1961) 1 and J. B. Parker, J. H. Towle, D. Sams, W. B. Gilboy, A. D. Purnell and H. J. Stevens, Nucl. Instr. and Methods, <u>30</u> (1964) 77

(2) M. M. Islam and H. H. Knitter, Nuc. Sci. Eng. 50 (1973) 108

#### 1.9 <u>A fission chamber for the intercomparison of fast neutron flux density measurements</u> (D. B. Gayther)

A discussion group at the International Specialists' Meeting on Neutron Standards held at the National Bureau of Standards in March 1977<sup>(1)</sup> proposed that the international comparison of flux density measurements organized by the Bureau International des Poids et Mesures should introduce cross-section measurements for use as transfer methods. This would allow flux measurements on the monoenergetic sources (such as Van de Graaffs) and the "white" spectrum sources (such as linacs) to be compared. Suitable neutron energies which were suggested for the intercomparison were 144 keV, 565 keV, 2.5 MeV, 5.5 MeV, 14 MeV and 25 MeV, the lowest of which would enable reactor filtered beams to be included. The <sup>235</sup>U(n,f) cross-section was considered to be the most suitable for the purpose, and it was later suggested that the <sup>238</sup>U(n,f) cross-section could be included. at the higher energies without much extra effort by making a back-to-back <sup>235</sup>U/<sup>238</sup>U fission chamber. Nuclear Physics Division, AERE, undertook to provide the detector and the following is a brief description of a proposed design.

The detector will consist of an ionization chamber of conventional design operated with a continuous gas flow of argon plus 10% methane. This choice was dictated by the need for stability of performance and ease of operation. A timing resolution of 5-10 ns is readily achieved which is adequate for use on linacs and will allow some timing

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discrimination of unwanted events on pulsed monoenergetic sources.

"If an accurate determination of the fission cross-section is to be made the detection efficiency for fission events should ideally approach 100%; corrections for such effects as loss of fragments in the fissile deposit and scattering of incident neutrons from the chamber structure would be small and calculable. To achieve realistic counting rates on existing linacs in the required energy range it is necessary to use a multiplate chamber with foils of moderate thickness. The above corrections will not therefore be entirely trivial. The chamber must consequently be regarded primarily as a transfer instrument for the intercomparison of neutron flux measurements. However, in designing the detector and specifying its mode of operation, the need to make accurate determinations of the efficiency correction factors has been borne in mind and it is hoped that it will be possible to derive the fission cross-section with some precision from measurements taken with the chamber.

Two separate chambers will be constructed, one containing <sup>235</sup>U foils and the other containing <sup>238</sup>U foils. This arrangement is preferred to a simple back-to-back chamber since with the multiplate system it reduces the effects of scattering in the foil backings and it also increases the fissile content for the given chamber length.

Each chamber will contain five platinum plates, each painted over a diameter of 76 mm on both sides with the fissile material to a thickness of ~ 5 µg/mm<sup>2</sup>. The foils are being prepared by Mrs. K. M. Glover of the Chemistry Division, AERE. Painting was considered to be the only available technique capable of providing durable deposits of the required thickness and diameter. Platinum was chosen for the backing material in preference to aluminium which is often used for cross-section measurements, mainly because structure in its scattering cross-section at the lower neutron energies under consideration is not as pronounced. Six tantalum collector plates will complete the electrode assembly which will be held together by small diameter nylon rods and spacers. The assembly will be mounted concentrically in the cylindrical body of the chamber with lightweight support structs. The body will be made of stainless steel and will contain thin end windows to minimize scattering of the incident neutrons. The detailed design and construction of the chambers will be supervised by Dr. J. W. Leake of the Instrumentation and Applied Physics Division, AERE.

A consequence of the proposed design when used in "open" (uncollimated) geometry is a small contribution to the observed counting rate from neutrons scattered by the chamber

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walls, end plates, etc. A design in which this effect would be negligible was considered to be impracticable since the chamber must be sufficiently robust and compact for its task. A valid intercomparison of flux measuring techniques on the monoenergetic and white neutron sources will therefore only be possible if, in each case, an allowance for the wall contribution is made correctly. In using the chambers each laboratory will accordingly be expected to either.

- (a) effectively eliminate the wall contribution by collimation. The neutron beam required would be of uniform intensity across the foil sub-assembly and would not strike the main body of the chamber. This condition will always apply to the white sources. It could also be applied on the monoenergetic sources since enough fissile material is present in the chambers for them to be used at a distance of several metres from the target.
- or (b) use open geometry but determine the wall contribution experimentally; for example, by using a shadow bar which acts as the exact conjugate of the above collimator.

It is hoped that the chambers will be ready for use by mid-1979.

(1) Proc. of the International Specialists' Symposium on Neutron Standards and Applications, Gaithersburg 1977, NBS Special Publication 493 (1977)

# 1.10.H Measurement of fast neutrons using nuclear track detectors (A. V. Hague\* and C. B. Besant\*)

The properties of solid state track detectors for neutron measurements in high gamma radiation fields are being investigated. The programme consists of 1) Designing a neutron detector based on the tracks from recoil protons,  $(n,\alpha)$  reactions or fission fragments; 2) Calculating the response of detectors to neutrons in the energy range 0 to 17 MeV; 3) Verification of the calculated response by measurements using monoenergetic neutrons; 4) Measuring the effect of a high gamma dose on the detector performance; 5) The application of automatic track detection techniques.

A measurement of recoil protons from neutrons in the energy range 0.1 to 17 MeV has been carried out for cellulose nitrate. The measurements were made using the IBIS accelerator and the Harwell Long Counter as an absolute measurement of neutron flux.

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Fig. 13 Sensitivity of cellulose nitrate as a track detector for recoil protons from incident neutrons as a function of neutron energy. The curve is an eye fit through the points

The results shown in Fig. 13 have been interpreted in terms of a simple model based on the (n,p) scattering cross section and a detection efficiency for protons which increases with energy. The agreement between experiment and theory is not yet satisfactory and has lead to the realisation of a number of fundamental differences between neutron and charged particle detection in track recorders. These differences are 1) Neutron induced tracks originate uniformly from within the body of the material. 2) Neutron induced tracks are not readily observed on the incident face because recoil protons are directed into the bulk of the material. 3) In contrast with charged particle tracks, a chemical etch of the back surface of the material will reveal tracks which are caused by charged particles leaving the material. The characteristics of such tracks differ because the energy loss of the emergent particle in the material decreases as the etch penetrates into the material. The emerging recoil protons from the back surface will have a range of different energies and directions, even for monoenergetic neutrons.

Experiments on fission fragment tracks in Makrofol from (n,f) in  $^{238}$ U have to be

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carried out and the effect of gamma rays on track recognition has been measured for doses up to 24 M rads. Automatic track counting techniques, developed at Imperial College, are being applied to neutron induced track measurement and should help to overcome the subjective nature of human decisions on the number of tracks seen in an etched foil.

# 1.11.A,D <u>Measurements of MeV neutron cross-sections by activation (M. T. Swinhoe\* and C. A. Uttley)</u>

Neutron cross-sections relevant to reactor dosimetry and fusion reactor data requirements have been measured between 4.7 and 14.1 MeV using the Tandem and Cockcroft-Walton accelerators. The irradiations on the Tandem were carried out using the  $D(d,n)^{3}$ He reaction with a 10 cm deuterium gas cell filled to 2 atm. The maximum neutron energy for the measurement of a cross-section was limited by the condition that the correction to the induced activity from break-up neutrons was less than 10%. Absolute cross sections were measured on the Tandem generator using the NE 213 liquid scintillator which was previously calibrated in terms of absolute efficiency from 1.5 to 25 MeV neutron energy by the associated particle method (PR/NP 25, p 43). The cross-sections measured on the Cockcroft-Walton acclerator at 14.1 MeV were relative to the  ${}^{27}$ Al(n, $\alpha$ )<sup>24</sup>Na cross-section of 122 <u>+</u> 2 mb. The history of each irradiation on the Cockcroft-Walton accelerator was monitored by observing the alpha particles emitted from the titanium tritide target, and the samples were contained in a 1 mm thick cadmium can for all the measurements.

As reported previously (PR/NP 25, p 44) the principal measurement was the 'Li(n,n'at) cross-section in which the induced tritium activity was determined by  $\beta$  counting. The lithium samples chosen for the measurement were 1 gm compacted discs of LiOH (99.99% <sup>7</sup>Li) enclosed in a thin case of the same material to compensate for loss of energetic tritons. This compound was preferrred to Li<sub>2</sub>CO<sub>3</sub> because no binder is required. Chemical analyses have shown a negligible concentration of monohydrate in the samples. It has also been established that the labile fraction of tritium stored in LiOH is 99.3% <sup>(1)</sup>. However the effect of this and other possible causes of loss of tritium during the preparation of the irradiated samples for  $\beta$  counting can be determined by a thermal reactor irradiation to measure the 2200 m sec<sup>-1</sup> value of the <sup>6</sup>Li(n, at) cross-section. For this purpose samples of LiOH containing 0.6% Li<sup>6</sup> have been prepared gravimetrically.

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Fig. 14 The cross-section for the <sup>7</sup>Li(n,n'at) reaction. The black points are the (various) previous data (obtained from the NEA data bank, Saclay). The open squares are the present measurements.

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The measured cross-sections between 4.7 and 14.1 MeV are shown in Fig. 14 and compared with other previous measurements and the ENDF/B IV evaluation. Only a few of the earlier data were direct measurements of tritium activity. The present data are well represented by the dashed curve which is 26% lower than the ENDF/B IV evaluation. A measurement of the 2200 m sec<sup>-1</sup> value of the <sup>6</sup>Li(n, $\alpha$ t) cross-section by cadmium difference has been carried out on the GLEEP reactor and a preliminary value of 900 ± 50 barns has been obtained. Further work is in progress to investigate the effect of flux depression and self shielding and on the mass spectrometry of the samples.

Thin foils of aluminium and natural titanium were also irradiated concurrently with the lithium samples at most energies. The values of the Al(n, $\alpha$ ) and the <sup>47</sup>Ti(n,p) and <sup>48</sup>Ti(n,p) cross-sections are given in Table 5 which includes a measurement of <sup>58</sup>Ni(n,p) relative to Al(n, $\alpha$ ) at 14.1 MeV. The high cross-section for <sup>4</sup>Ti(n,p)<sup>47</sup>Sc at 14.1 MeV of 220 <u>+</u>5 mb compared with the accepted value of 120 mb for enriched <sup>47</sup>Ti samples is explained by the contribution from <sup>48</sup>Ti(n,np)<sup>47</sup>Sc. The natural isotopic abundances of <sup>48</sup>Ti and <sup>47</sup>Ti are 73.96% and 7.29% respectively and the observed value of 220 mb corresponds to a cross-section for <sup>48</sup>Ti(n,np)<sup>47</sup>Sc of 9.9 <u>+</u>0.5 mb in agreement with the value of 9 <u>+</u>2 mb obtained by Qaim and Molla<sup>(2)</sup> at 14.7 MeV. The activities induced in the metal foil samples in these irradiations were measured by the Actinide Analysis Group in Chemistry Division and the tritium activity by the Low-level Tritium Measurements Laboratory in Industrial Physics Group.

Neutron	1	Reaction Type								
Energy MeV	A1(n,α)	47 <sub>Ti(n,p)</sub>	<sup>48</sup> Ti(n,p)	<sup>58</sup> Ni(n,p)						
14.1	(122 + 2)	220 <u>+</u> 5	63 <u>+</u> 2	382 <u>+</u> 15						
14.0	124.1 + 0.7		68 <u>+</u> 2.6							
11.8	117.0 + 3.7	:	53.7 <u>+</u> 2.2	· .						
9.8	88.4 + 2.2	144.5 + 4.2	33.2 + 1.0							
7.5	25.4 <u>+</u> 0.6	117.0 <u>+</u> 3.5	10.9 <u>+</u> 0.5							
5.8		83.1 + 3.9	0.71 + 0.14							

Table	5
Table	

Threshold	reaction	cross-se	ctions

(1) Hiroshi Kudo, Kichizo Tanaka and Hitoshi Amano. Journal Inorg. Nucl. Chemistry <u>49</u> (1978) 363

(2) S. M. Qaim and N. I. Molla, Proc. of the 9th Symposium on Fusion Technology, Pergamon Press p 589(1976)

## 1.12.A (n,α) cross-sections (J. A. Cookson, M. Langton\* and C. Wise\*) [Relevant to request numbers: 1369-71, 1394-96, 1417-19]

We aim to measure  $(n, \alpha)$  cross-sections for Fe, Ni and Cr, which are components of stainless steel, at energies of interest for fusion and fast reactors. The initial phase



Fig. 15 The energy spectrum of the alpha particles emitted from the natural Ni(n,α) reaction with 14.7 MeV neutrons, after background subtraction. was reported last year (PR/NP 25, p 45). Since then considerable progress has been made using a detector telescope consisting of 4 proportional counters and a CsI scintillator. Preliminary angular and energy distributions of the alphas from natural Ni(n, $\alpha$ ) with 14.7 MeV neutrons from the Cockcroft-Walton accelerator have been obtained (Fig. 15). When the 14.7 MeV data on all three of the elements of interest is complete, measurements at lower neutron energy will be started using the tandem accelerator.

#### 1.13.D (α,n) yields from light elements (D. West and A. C. Sherwood) [Relevant to request numbers 19,53,59,1525, 1526, 1528]

The large polythene moderator for measuring neutron yields independently of their energy (PR/NP 25, p 46) was installed at the Tandem generator together with its electronics in January. Fig. 16 shows a photograph of the moderator in place on its trolley. A first run took place in February and data taking runs have been carried out at monthly intervals since then. Apart from a few initial tests, concerning the sensitivity of the system to (a) position of the target along the axis of the detector and (b) misalignment of the beam with respect to the axis of the detector, measurements started at once. The conducting targets Al, Mg, C, Si and Fe were first measured.

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Fig. 16 Shielded polythene moderator (1 m right cylinder) with nine counter holes, shown withdrawn from the long Faraday cup which supports the target. In the foreground on the left is the Rutherford scattering chamber for beam energy measurement

1.13.1 Method

Some limitations and necessary changes in procedure soon became apparent. These were:

- The need for a theodolite permanently set on the axis of the detector and beam pipe for aligning the beam.
- 2) A single energy measurement using Rutherford scattering from a thin Au foil could be used for neutron yield measurements with several targets in succession on the same day provided the beam intensity was not altered. Even so, check inspection measurements of the Au scattered α-particle peak position were needed after each yield measurement (approximately one hour's duration). As expected from preliminary trials the beam energy could not be reproduced on successive days to better than 10 keV. Over a period of three months the observed mean departure from nominal energy was

only - 6.6  $\pm$  2.5 keV but the standard deviation of any single setting of the energy was 10.7 keV, the accuracy of measurement being 1-2 keV.

Methods were developed of measuring the thickness of Au and the dead layer on the Si detector by inclining it to the direction of incident particles and it was also found necessary to determine the thickness of the Au foil scatterer and its C backing. This was done by interposing the foil between a  $^{244}$ Cm  $\alpha$  particle source and the detector and measuring the reduction in energy of the  $\alpha$  particles. An absolute determination of the intensity of the Au-scattered peak (using the Tandem  $\alpha$  particles) then enabled both the C and Au thicknesses to be deduced. The growth of carbon deposited on the foil from pump oil could be measured and allowed for. It amounted to about 10 µgm/cm<sup>2</sup> for a foil used throughout three runs.

- 3) With a Si target some irreproducibility of yield was first observed which was thought to have been due to the effects of channelling of  $\alpha$ -particles with the single crystal target used. As similar effects may occur with rolled metal targets the procedure was adopted of inclining all targets with their normals at 6<sup>°</sup> to the beam axis.
- 4) The neutron counting system was satisfactory and stable with a very low background. Prior to each machine run, a single set of neutron calibration runs was taken using the calibrated Am-Be neutron source. Neutron counting rates from the light element targets were kept below values at which counting losses were significant. This may well not be possible with the more prolific neutron producers (Be, B, Li and F). We are therefore considering a change from the type 2126 discriminators to type 2133 which have a well defined deadtime. An increase of deadtime from its present mean value of 2.7 µsec to 10 µsec will result.
- 5) No yield measurements were possible for  $\alpha$  particle energies below 3.6 MeV where stable operation of the Tandem became impossible.

#### 1.13.2 Results

The yield measurements we have made to date are plotted as individual values in Fig. 17. They include data taken for the insulating material UO<sub>2</sub> which presented no special problems in practice. The continuous lines in the figure are previously available data cited and/or derived from thin target data by Liskien and Paulsen<sup>(1)</sup>. Our plotted values of yield are not yet corrected for the neutron leakage from the system during calibration with the Am-Be source. A preliminary value for this leakage derived from two computer models is 0.5%. Neutron absorption with the calibration source both in the

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Fig. 17 Thick target yields from a-particles. Previous data are shown as the continuous lines

source itself, which is expected to amount to 0.2%, and in the beam pipe will be allowed for when details of measurements of gold foils irradiated in the moderator are received from N.P.L. The plotted values are also not corrected for any neutron producing i impurities discussed below.

The measured yield values agree well with previous determinations especially for Al and C. The data for Si and UO<sub>2</sub> on the other hand show considerable divergences from previous data. Work on the materials mentioned is now essentially finished. Other targets available in convenient solid form are Be, B, UC and stainless steel. There remain targets such as Na or Li, which require special handling arrangements, and the gaseous targets D, N, O and F which will require considerable target development work. There is also interest in studying the yields of compounds of elements whose individual neutron yields have been measured. UC is one such case.

#### 1.13.3 Neutron producing impurities in the targets

All the targets used above, which were of high nominal purity, have been examined by studying the  $\gamma$ -ray spectra under  $\alpha$ -particle bombardment. The  $\gamma$  rays arise predominantly from ( $\alpha$ ,n), ( $\alpha$ ,p) or ( $\alpha$ , $\alpha$ ') reactions and characterise the target material<sup>(2)</sup>. The analysis of the large amount of data collected has not been completed. Some definite results have however emerged already.

- (a) A stainless steel target of reactor grade material which gave a neutron yield very much higher than that from Fe was shown to contain an appreciable quantity of Al identified from its  $\gamma$ -ray spectrum. It was shown to be a surface contamination introduced during rolling particular samples and was removed by grinding off a surface layer several thousandths of an inch thick. Another constituent of reactor grade stainless steel, Si, was detected and corresponded in amount to the chemical analysis, 0.34% by weight. This would dominate the rather small neutron production at energies below about 6 MeV.
- (b) The UO<sub>2</sub> sample gave identifiable  $\gamma$ -ray lines from  $\alpha$ -induced reactions in C, Al and Si. If confirmed, these would contribute the following percentage neutron yields for 7.5 MeV  $\alpha$ -particles: respectively, 0.7%, 0.9% and 0.5%. The quantities of material present are very much smaller than the percentages of neutrons produced from them but are roughly consistent with the specified impurity levels for reactor grade material.
- (c) With iron the situation is complicated by masking of the three most prominent oxygen

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 $\gamma$ -ray lines at 0.3505 MeV, 1.3951 MeV and 1.9822 MeV and the two most prominent silicon lines at 1.2661 MeV and 1.7789 MeV by radiations due to Fe itself. However by using other  $\gamma$  radiations namely the 1.1200 MeV line from  $0^{18}(\alpha,n) \text{Ne}^{21}$ , the 2.2338 MeV line from  $\text{Si}^{28}(\alpha,p)p^{31}$  and the 6.1304 MeV line from  $\text{C}^{13}(\alpha,n)0^{16}$  the following neutron contamination from the Fe target is to be expected at 5 MeV:

> from Oxygen  $\leq 1.6 \times 10^{-10}$  neutrons/ $\alpha$ from Silicon  $\leq 3.1 \times 10^{-12}$  neutrons/ $\alpha$ from Carbon  $\leq 2.9 \times 10^{-11}$  neutrons/ $\alpha$

These values are to be compared with the measured yield which is in the region of  $10^{-10}$  neutrons/ $\alpha$  at 5 MeV. The rapid variation of the yield from Fe with energy soon makes these possible contaminations unimportant at higher energies.

 (d) A uranium metal target, used in unravelling the γ ray spectrum from UO<sub>2</sub>, itself had a considerable and unexpected oxygen content. It is to be re-examined after grinding off the surface layers.

- (1) H. Liskien and A. Paulsen, Atomkernenergie, 30 Lfg 1 (1977) 59
- (2) E. W. Lees and D. Lindley, Annals of Nuclear Energy, 5 (1978) 133

1.14 The calculation of thick material  $(\alpha, n)$  yields (B. W. Hooton and A. R. Talbot) There are many situations involving plutonium or other actinides where the yield of neutrons from  $\alpha$  activity is required. The neutron yield can be calculated by integration of the differential cross-section as the alpha particle loses energy. This is a cumbersome calculation requiring a detailed knowledge of the differential cross-section and an approximate method which is simple to use would be useful, particularly since the  $\alpha$ activity can be present in a wide variety of host materials.

The yield from N(I,J) atoms of a nucleus I in a material J is given by

$$Y(I,J) = N(I,J) \int_{0}^{E_{m}} L^{-1}(J,E) \sigma(I,E) dE$$
 (1)

where  $E_m$  is the maximum energy of the  $\alpha$  particle, L (J,E) is the specific energy loss for the material and  $\sigma(I,E)$  is the ( $\alpha$ ,n) cross-section. The integrand depends on J but if L(J,E) is a slowly varying function of energy then the yield can be approximated by,

$$Y(I,J) = N(I,J).L^{-1}(J,E) \int_{0}^{E_{m}} \sigma(I,E) dE$$
 (2)

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The integral is now a constant, for a given value of  $E_m$ . The ratio of yields from nuclei of type I in two materials is given by

 $\frac{\underline{Y}(\underline{I},\underline{J})}{\underline{Y}(\underline{I},\underline{K})} = \frac{\underline{N}(\underline{I},\underline{J})}{\underline{N}(\underline{I},\underline{K})} \frac{\underline{L}^{-1}(\underline{J},\underline{E})}{\underline{L}^{-1}(\underline{K},\underline{E})}$ 

Calculations on the validity of this approximation have been carried out using a constant value of L(J) equal to the specific energy loss at  $0.8E_m$ . The results for a variety of materials containing oxygen are given in Table 6. The absolute values agree with Liskien<sup>(1)</sup> for 0<sub>2</sub> but are about 17% higher for UO<sub>2</sub>. This difference can largely be attributed to differences between the values of L(J,E) used by Liskien and those used in the present calculation<sup>(2)</sup>. The results show a good agreement in absolute yield at 5.0 MeV and that the ratio of yields can be predicted to within 3%. The approximation can be used to predict the yield Y(I,J) in any material relative to the experimental value in one standard material.

		Table	<u> </u>				
Neutron yields	from	oxygen	per	10 <sup>8</sup>	alpha	particles	

Yields are calculated according to equations (1) and (2)

۰.	$E_m = 5.0 \text{ MeV}$				м.	$E_{\rm m} = 4.0  {\rm MeV}$			
	Ec	1. (1)	Appro	<b>x.</b> Eq. (2)	Eq	Eq. (1)		. Eq. (2)	
	Y	<sup>Y</sup> / <sub>Y</sub> (U0 <sub>2</sub> )	Y	Y/Y(U02)	Y	<sup>Y</sup> / <sub>Y</sub> (U0 <sub>2</sub> )	Y	Y/Y(U02)	
• 0 <sub>2</sub>	3.77	2.83	3.78	2.84	1.40	2.70	1.33	2.64	
н <sub>2</sub> 0	2.50	1.88	2.51	1.89	0.925	1.78	0.877	1.74	
U0 <sub>2</sub>	1.33	1.0	1.33	1.0	0.519	1.0	0.504	1.0	
UO2(NO3)6H20	2.34	1.76	2.35	1.77	0.878	1.69	0.834	1.65	

(1) H. Liskien and A. Paulsen, Atomkernenergie 30 (1977) p59

(2) L. C. Northcliffe and R. F. Schilling, Nucl. Data Tables 7 (1970)

#### 1.15.E Neutron production from $(\alpha, n)$ reactions in actinide oxides (E. W. Lees)

As described in PR/NP 25, pp 50-55, the neutron yield from  $(\alpha,n)$  reactions on <sup>17</sup>0 and <sup>18</sup>0 in a 12 g <sup>241</sup>AmO<sub>2</sub> sample has been measured to be  $(2.78 \pm 0.40) \times 10^3$  n/s/g <sup>241</sup>Am; this work is now published <sup>(1)</sup>. The relatively poor accuracy of the measurement (~ 15%) was due to the presence of a small F impurity in the sample which contributed approximately half of the emitted neutrons from the <sup>19</sup>F( $\alpha,n$ ) reaction. The presence of the F impurity was deduced by studying the gamma-ray spectrum of the 12 g AmO<sub>2</sub> sample and comparing it to those resulting from alpha-reactions on light elements; the gamma-ray spectra were obtained using a 80 cm<sup>3</sup> Ge(Li) detector. The number of neutrons from <sup>19</sup>F( $\alpha,n$ ) reactions  $\cdot$  was estimated by relating the gamma-ray and neutron intensities of the 12 g AmO<sub>2</sub> sample and comparing the obtained using a 40 cm<sup>3</sup> Ge(Li) detector. The number of neutrons from the 12 g AmO<sub>2</sub> sample and comparing the gamma-ray and neutron intensities of the 12 g AmO<sub>2</sub> sample and commercial Am/F sources whose neutron output had been previously calibrated. However, this correction decreased the original 4% accuracy in the knowledge of the total neutron output of the sample.

Clearly, it would be desirable to repeat the measurement with a "pure"  $^{241}$ AmO<sub>2</sub> sample and work this year has concentrated on finding such a sample. Three samples have been investigated and the gamma-ray spectra from 2 of these are shown in Fig. 18. The 0.5 g sample (Fig.18a) contains F in an even greater concentration than that found for the original 12 g sample. The 3 g sample spectrum illustrated in Fig. 18(b) is the only one investigated to date which appears to show little or no F contamination; in addition to the  $^{60}$ Co and  $^{40}$ K room background lines, there is evidence for the presence of a  $^{154}$ Eu impurity in this sample.

At present, the 3 g sample has been returned to TRC Amersham for repacking in a standard X2 source container whose geometry and construction is well specified. It is hoped to perform the neutron yield measurements during 1979. It is also planned to make measurements of the neutron yield from a fuel pin prior to irradiation in order to check the theoretical estimates.

(1) E. W. Lees and D. Lindley, Ann. of Nucl. Energy 5 (1978) 133

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Fig. 18 The gamma-ray spectra from 0.5 g and 3 g  $^{241}$ AmO2 samples are shown in (a) and (b) respectively. Note the presence of reaction gamma-rays from both F and 17,180 in the 0.5 g sample and the dominance of the gamma-ray spectrum from the 3 g sample by the  $^{154}$ Eu impurity

# 1.16 A model for studying coincidence systems used in plutonium assay (B. W. Hooton and E. W. Lees)

Coincidence systems used in detectors for plutonium assay have been studied by a Monte Carlo model which assigns a time coordinate to each neutron (PR/NP 25, p 60). This model has been developed further by including the effect of an extended dead time in the neutron discriminator. The system now contains two dead times in series: an extended dead time in the discriminator  $T_1$ , and a non-extended scaler dead time T when the model is applied to the Variable Dead-time Counter (VDC). The effect of an extended dead time is to modify the time distribution of random neutrons into a non-Poisson form by forbidding pulse separations of value less than  $T_1$ . It is useful to define an effective deadtime for the system in terms of that required to satisfy the usual equation for counting losses under a Poisson distribution situation, namely

$$T_{e} = 1/C_{o} - 1/C$$

where  $C_{o}$  is the gross count rate generated by the discriminator and C is the measured rate in the scaler with dead time T.

The effect of two dead times in series has been derived using the theory of Muller<sup>(1)</sup> and it can be shown that the effective dead time is given by

$$\mathbf{T}_{\mathbf{A}} = \mathbf{T} - \mathbf{T}_{\mathbf{1}}$$

This relationship has been verified using the model.

The model has been used to simulate VDC experiments with input parameters specified to state a) the amount of spontaneous fission from  $^{240}$ Pu, b) the intensity of random ( $\alpha$ ,n) neutrons and c) an amount of fission multiplication. The fission multiplication parameter was determined by a separate neutron Monte Carlo code MONK and the model parameters were intended to reproduce the Euratom VDC developed at ISPRA. The detection efficiency was taken as 0.062 and the neutron die-away characteristics as a rise time of 2.5 µs and an exponential decay of 33 µs. The results are given in table 7 in terms of two parameters intended to measure fission intensity. The parameter X is defined as

$$\mathbf{x} = \mathbf{C}_{0} - \frac{\mathbf{C}}{1 - \mathbf{T}_{0}\mathbf{C}}$$

and is clearly shown to be inadequate, for the value of X for 100g and no multiplication is clearly less than the proportionality value of 285. It is interesting to note that this weakness is disguised when multiplication is present to the extent that the value

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#### Table 7

		· , ·	
· .	1g <sup>240</sup> Pu	100g <sup>240</sup> Pu	100g <sup>240</sup> Pu With multiplication
X	2.85 + 0.04	232 <u>+</u> 24	264 <u>+</u> 20
Y	2.86 <u>+</u> 0.04	289 <u>+</u> 24	334 <u>+</u> 22

Calculated variation of parameters X and Y (see text) supposedly

proportional to fission intensity, versus mass of <sup>240</sup>Pu

rises to 264. The true value with multiplication should be 332 showing that the parameter Y is a much better representation of the true situation. The parameter Y has been defined by Stanners  $^{(2)}(3)$  and includes more detailed consideration of counting losses. The Y values given in table 7 were obtained using a value of 23 µs for the Stanners parameter q. This value is related to the die away time constant of 33 µs in a manner which is not yet fully understood and it has been recommended by Stanners as the empirical value of q which gives the best agreement with measured data. The model will continue to be applied to the VDC and will be developed to examine the shift register techniques for measuring pulse spacings.

(1) J. W. Muller, Nucl. Inst. and Methods 112 (1973) 47

(2) W. Stanners, Directorate of Euratom Safeguards, Luxembourg. (Private communication)
(3) E. W. Lees and B. W. Hooton, AERE-R 9168 (1978)

1.17 Calculations of neutron multiplication and its effect on the response of neutron coincidence counters (E. W. Lees and B. W. Hooton)

We have continued our calculations of the effects of neutron multiplication on the response of Neutron Coincidence Counters (NCC) used in Safeguards work. A report on such calculations specifically for the Euratom Variable Dead-time Counter (VDC) has been published <sup>(1)</sup>.

Figs. 19 and 20 are taken from ref. 1. Fig. 19 illustrates the calculated response (Y) of the VDC as a function of  $^{240}$ Pu mass for Pu0<sub>2</sub> containing approximately 20%  $^{240}$ Pu arranged in spherical geometry; clearly for the same nominal mass of  $^{240}$ Pu one obtains results dependent on the density of the sample and always in excess of the zero

multiplication response where the response is proportional to the mass of <sup>240</sup>Pu present. As one would expect, isotopic and chemical compositions, geometrical arrangements and density are important criteria in determining the extent of neutron multiplication.



Fig. 19 Theoretical estimate of Y including fast fission from spontaneous fission and  $(\alpha, n)$  reaction neutrons as a function of <sup>240</sup>Pu mass; the two curves correspond to two choices of material density (see text for further details)

Fig. 20 Comparison of experimental data for mixed oxide samples ( $PuO_2/UO_2 = 30/70$ ) of large mass with the present calculations. The Y value per gram of  $^{240}Pu$  has been plotted to accentuate the effects of neutron multiplication (see text for further details)

Fig. 20 illustrates a comparison of the present calculations with experimental measurements on mixed  $Pu0_2/U0_2$  samples. The detector response (Y) per unit gram of  $^{240}Pu$  has been plotted to illustrate more clearly the effects of neutron multiplication since the former should be a constant in the absence of neutron multiplication. Although the theoretical estimate is too high in absolute magnitude, it does reproduce the observed trend of the results. The discrepancy is probably due to uncertainties in the packing density and geometry and also possible due to a choice of the material density being higher than the actual value.

Indeed, the major problem encountered to date in comparing experiment and theory has been the lack of detailed knowledge regarding the geometry and density of the doubly sealed samples. At present, a batch of samples whose geometry and density are well defined is being prepared and we hope to carry out a series of collaborative experiments on these with NMACT. Additionally, the theoretical calculations of neutron multiplication will be adapted to permit the theoretical response of the shift register system of analysis<sup>(2)</sup> to be predicted. Again, these predictions will be compared directly to the results obtained with the well defined standards.

- (1) E. W. Lees and B. W. Hooton, AERE R 9168 (1978)
- (2) K. Böhnel, KFK 2203 Karlsruhe (1975)

(3) R. Berg et al., EUR 5158e (1974)

1.18 N.M.A.C.T. portable neutron coincidence counter (PNCC) (E. W. Lees and F. J. G. Rogers (NMACT))

NMACT have now received their own PNCC the design of which is based on the Euratom instrument<sup>(1)</sup>. NCC instruments are used in Safeguards work for the passive, nondestructive assay of Pu mass by measurement of the <sup>240</sup>Pu content; the other Pu isotopic ratios can be determined by gamma-ray spectroscopy<sup>(5)</sup>. The basic layout of such a detector is shown in Fig. 21 and consists of 6 units (each containing three <sup>3</sup>He tubes) arranged in a hexagon around a well in which the sample to be assayed is placed. The analysis method illustrated in Fig. 21 is of the Variable Dead Time form<sup>(2)</sup> and is based on the fact that since the neutrons from spontaneous fission are correlated in time, then after the usual dead time corrections are performed, one is left with a residual proportional to the number of correlated neutrons, i.e. proportional to the number of spontaneous fission events.

$$M_{240} \propto b - \frac{c}{1 - cd}$$

where b is the true count rate and c is the count rate measured in a scaler with nonextending dead time d. Other analysis systems have been employed  $^{(3,4)}$ , but all utilise the fact that spontaneous fission usually results in 2 or more neutrons being released simultaneously, i.e. they all essentially measure the neutron multiplicity during the fission event.

It is important to understand fully the response of any instrument used in Safeguards work in order to assess its limitations and also to have confidence regarding its deployment in the 'field'. Consequently, we have been calculating the theoretical response of

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Fig. 21 Block diagram of a NCC detector with a Variable Dead-time form of analysis

NCC at Harwell (this report, pp 51 and 52). However, it is necessary to have accurate knowledge of certain physical properties of the instrument in order to proceed with the theoretical calculations and a measurement programme was undertaken on the NMACT PNCC. The most important properties measured are listed below:

1.18.1 Intrinsic system dead time

The minimum resolving time between successive neutron pulses was determined by two independent techniques to be 4.8  $\mu$ s for each six sub-sections of the detector. Since the six are processed independently (see Fig. 21), then the overall effective dead time is 0.8  $\mu$ s. This value is much longer than one would prefer and is due to the high pressure <sup>3</sup>He filling in the detectors; the use of  ${}^{10}\text{BF}_3$  counters would reduce this dead time by about a factor of eight but at the expense of detection efficiency.

1.18.2 Efficiency as a function of neutron energy

The efficiency has been measured at different locations using calibrated  $^{241}Am(\alpha,n)$  sources; the response is non-linear and rapidly increases at low neutron energies (Table <sup>8</sup>). This implies that any spectrum shifts due to sample materials will distort the detector response from the 'bare'  $^{240}$ Pu source calibration.
#### Table 8

# NMACT PNCC efficiency as a function of neutron energy The larger errors associated with the measurement on 25th July, 1978

Source	Average Neutron Energy (MeV)	Efficiency (%)				
		25th July, 1978 location A	3rd November, 1978 location B			
Am/Li	0.45	8.68 <u>+</u> 0.19	8.32 <u>+</u> 0.04			
Am/F	1.25	6.84 <u>+</u> 0.10	6.73 <u>+</u> 0.03			
Am/B	2.7	5.21 + 0.11	5.19 <u>+</u> 0.05			
Am/Be	4.2	4.94 <u>+</u> 0.17	4.87 + 0.05			

are due to the much larger neutron background at location A

#### 1.18.3 Die away time

The neutron capture probability distribution as a function of time after the fission event was measured using the detection of a  $^{252}$ Cf fission fragment as the START pulse for a time to amplitude converter (TAC); the detected neutrons were used as the STOP inputs.





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The response obtained (Fig. 2) does not follow any single exponential behaviour until after 25  $\mu$ s. The value of 39  $\mu$ s obtained from the exponential behaviour of the curve is longer than the 34  $\mu$ s that one would obtain with a single-shot TAC which is incapable of dealing with more than one neutron per spontaneous fission event.

### 1.18.4 <u>Time distribution between neutrons from the same fission event</u>

This was again measured using a  $^{252}$ Cf source and TAC, but the analysis is as yet incomplete.

#### 1.18.5 Reproducible response to Pu0, standards

This was measured over a period of months, during which time the PNCC had been dismantled and reassembled, in order to assess its long term stability and reproducibility. As is shown in Table 9, this appears to be satisfactory.

#### Table 9

#### NMACT PNCC consistency checks using Pu0, standards;

·. ·				and the second
• • •	240 Pu mass		Ŷ	
	(g)	31st.August, 1978	7th September, 1978	16th November, 1978
	9.0	30.2 <u>+</u> 0.8	31.1	29.3
	18.0	59.3 <u>+</u> 2.1	60.7	
	27.0	89.5 <u>+</u> 2.5		89.2
	36.1	119 <u>+</u> 4	129	
	45.1	157 <u>+</u> 6		147
	54.1	183 <u>+</u> 6	191	
	63.0	214 <u>+</u> 8		209'
, • 1	72.3	257 <u>+</u> 9	249	
	81.2	273 <u>+</u> 12		267
•	90.4	316 <u>+</u> 12	323	· · · · ·
	99.4	337 <u>+</u> 13		333
	108.5	402 + 18	, 380	
	117.5	404 <u>+</u> 20	431	399

#### Y is the response of the instrument

A joint exercise with the IAEA to compare the practical usage of the NMACT NCC and their own NCC has just been completed and the results are awaited with interest. Finally, the extension of the pulse processing to the shift register method<sup>(3)</sup> has not proved possible due to malfunctions of the associated electronics and to the inherent pulse rate limitations of the existing design. It is hoped to correct this in the near future.

- (1) R. Berg et al, EUR 5158e (1974)
- (2) E. W. Lees and B. W. Hooton, AERE R 9168 (1978)
- (3) K. Böhnel, KFK 2203 Karlsruhe (1975)
- (4) E. J. Dowdy et al, IAEA-SM-231/69 (1978)
- (5) M. F. Banham, AERE R 8737 (1977)

#### 1.19 Fuel cycle studies (M. G. Sowerby)

A number of paper studies on topics involved with the nuclear fuel cycle have been carried out during the past year in response to requests for information from a number of sources. Most of the work has been related to one or more of the following topics:

- (i) the amounts of different nuclei produced in and discharged from reactors and their associated activities
- (ii) alternative fuel cycles
- (iii) non-proliferation

A paper entitled "Non-proliferation and Nuclear Data" was presented at the International Conference on "Neutron Physics and Nuclear Data for Reactors and Other Applied Purposes", Harwell, September 1978. The abstract was: "A review is made of the problem of the proliferation of nuclear weapons with particular emphasis on proliferation and nuclear power. Some indications of the nuclear data requirements associated with methods of reducing proliferation risks are presented."

1.20 Status of the fission product inventory code FISPIN at Harwell (B. W. Thomas and D. A. J. Endacott)

Since the previous report (PR/NP 25, p 71) considerable effort has been made to generalise and streamline the reading of data libraries from a range of input devices (i.e. cards, disc, tape). Some difficulties have arisen due to the existing structure of the code but versatility has been improved and limitations can be specified.

Some minor restructuring of the code has taken place and a few remaining errors have

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been eliminated. Modifications have been made so that the code can now be used to read library data from tape in the ENDF/B IV form.

A range of data libraries for different reactor types is now available and these can be loaded in card image form using the Panvalet library system. These will be available as data sets on a direct access device (disc) in the near future.

1.21 Assessment of activation and transmutation in fusion reactor first wall and structural materials (O. N. Jarvis)

In the previous report (PR/NP 25, p 72) the adaptation of the fission reactor code ORIGEN<sup>(1)</sup> to fusion reactor applications was described and the sources of data suitable for inclusion in a nuclear data library to be accessed by ORIGEN were listed. The past year has seen the completion of a nuclear data library which can be used for general studies with some confidence. There will, nevertheless, be a continuing need to upgrade the library as new experimental measurements and evaluations become available. Also, a much improved and extended version of the ORIGEN code will become available shortly and this should be adopted as it will correct certain deficiencies which have become apparent in the present code.

A review paper on Nuclear Data Requirements for Transmutation and Activation of Reactor Wall and Structural Materials was prepared for an IAEA advisory group meeting held in Vienna during December 1978. This paper is a shortened account of a report<sup>(2)</sup> in which the sources of nuclear data for activation/transmutation calculations are detailed and the published literature on activation/transmutation assessments reviewed. Also the reliability of such calculations is assessed by reproducing (satisfactorily) certain extensive published studies and finally, the particular nuclear reactions which contribute significantly to the activation, transmutation and gas production in several potential structural materials are identified.

(1) M. J. Bell, ORNL-4628 (1973)

(2) O. N. Jarvis, AERE-R 9298 (to be published

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#### 2.1 INTRODUCTION

Chemical nuclear data experiments and evaluations are coordinated by the Chemical Nuclear Data Committee (Chairman: J.G. Cuninghame). This committee, which is also the Chemical Sub-Committee of the UKNDC, is made up of measurers, evaluators and users of chemical nuclear data and is able to advise on measurements and to consider in detail reports of work on the compilation and evaluation of data in this field. The preparation of the CNDC nuclear data file is under the control of the Data File Sub-Committee (DFSC) whose chairman is Mr. B.S.J. Davies (CEGB, BNL). The main committee held two meetings and the Sub-Committee three meetings in 1978. The request list published early in 1977 is now being revised.

Work is in progress on very nearly all items on the list but often on a long time scale and with little prospect of early completion. Data File work has now reached the stage where data is available to the user and the status of the file is indicated in the table below.

The efforts of the committee to reduce the duplication work have been successful and the objective of effective international cooperation in the data field is being pursued.

This section brings together reports on the chemical nuclear data work being done at Harwell (AERE), Winfrith (AEEW), Aldermaston (AWRE), Dounreay (DNPDE), Windscale (BNFL) and Berkeley (CEGB) under the headings:

2.2 MEASUREMENTS

2.2.1 Fission Yields

2.2.2 Neutron Cross-Sections

2.2.3 Half-lives

2.3 EVALUATIONS AND COMPILATIONS

2.3.1 Data File Sub-Committee

2.3.2 Fission Yields

2.3.3 Related Compilation Studies

2.2 MEASUREMENTS

2.2.1 Fission yields

(1) <u>Tritium yields in thermal and fast fission (I.C. McKean and E.A.C. Crouch (AERE))</u> The method developed is being applied to samples of <sup>235</sup>U and <sup>239</sup>Pu in dilute nitric acid solution. These have been irradiated in polythene containers in the GLEEP

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reactor and after a suitable cooling period the tritium will be separated and measured.

For the fast fission measurements samples of pure metallic <sup>235</sup>U and <sup>239</sup>Pu have been prepared and sealed into containers made of Superpure Aluminium (99.9999% Al) ready for irradiation in ZEBRA. Blanks will be irradiated at the same time including some containers made from Aluminium of a composition equivalent to reactor grade Aluminium to check whether this easily available material will be suitable for containers in any further irradiations. The <sup>235</sup>U wire used has been produced by a method which involved contact with a heating medium of potassium, sodium and lithium carbonates as an intermediate step. Analysis for lithium contamination of the 1 mm diameter wire indicated a content of only 0.2 ppm lithium. It is expected that the irradiations in ZEBRA will take place in April 1979. Samples of <sup>240</sup>Pu and <sup>241</sup>Pu have been ordered from the USA for future irradiations.

#### (2) Fission yield measurement's at Dounreay (W. Davies and V.M. Sinclair (DNPDE))

Irradiation of the capsules of the PFR fission yield experiment commenced in June 1978 (the work for this experiment was outlined on pages 43-44 of UKNDC (75)p71 - the progress report for April '74 to March '75).

# (3) <u>Absolute fast fission yields of <sup>99</sup>Mo and other nuclides in <sup>238</sup>U fission</u> (J.G. Cuninghame and H.H. Willis (AERE))

The measurements made so far have been at 900 keV neutron energy (3 runs) and 1700 keV (2 runs). Further runs at 1300 and 2000 keV are planned but the work is at a halt due to staff shortage.

(4) Development of a method for the simultaneous measurement of fission yields of a large number of fission products from a low flux irradiation of nuclear fuel (J.G. Cuninghame and H.H. Willis (AERE))

The method depends on highly accurate  $\gamma$ -spectrometry of un-separated or only partially separated fission product sources using Ge(Li) detectors accurately calibrated for efficiency. Tests have now been successfully completed and we are using it in the measurements reported in the item below.

(5) <u>ZEBRA-BIZET</u> experiments to study the effect of change of reactor neutron spectrum of fission yields (J.G. Cuninghame and H.H. Willis (AERE))

Four irradiations have been made for each of two <sup>235</sup>U, two <sup>238</sup>U and <sup>239</sup>Pu metal beads of 100 mg weight. Two were in the inner core and two in the outer. One of the samples of each of the fissile materials was counted directly, while the other was dissolved and used to prepare purified samples of certain fission products present at very low yield viz. As, Ag, Cd, Sn, Sb, and Rare Earths.

Provided the total number of fissions in the target material is accurately known, absolute fission yields can be obtined from the  $\gamma$ -spectra by the method mentioned



Fig. 1 Fission yields for fission of <sup>235</sup>U irradiated in an inner core position of the Zebra-Bizet reactor

in the item above, but even if it is not, semi-absolute yields can be produced by normalisation of the whole yield curves to 200%. More irradiations still have to be performed and we are yet some way from being able to determine the number of fissions properly but preliminary results, exemplified by Fig. 1 are encouraging.

(6) Development of a version of the GAMANAL computer programme <sup>(1)</sup> for analysis of  $\gamma$ -spectra, primarily for fission yield studies (Jill A.B. Goodall (AERE))

Continued development of the programme now gives the following options additional to those mentioned in last years report (UKNDC (78)P88,p91).

- (a) Addition or subtraction of spectra from each other.
- (b) Output of the detailed calculations by which GAMANAL estimates the background and the fit to the peaks.
- (c) Use of a pulse generator peak to estimate live time.

Extensive testing of the programme leads us to believe that it now provides one of the most accurate means of analysis of  $\gamma$ -spectra available, is simple to use and is superior to either DIODE or SAMPO.

(7) <u>Development of software to allow a PDP-11 computer to perform as four independent</u>, automatic 4-K pulse height analysers (J.G. Cuninghame and J. Venn (AERE))

The original software (written by a commercial concern) has been extensively

modified to

ł,

- (a) Allow dumping of spectra on floppy discs as RT-11 files,
- (b) Convert the programme from "stand-alone" to an RT-11 file,
- (c) Alter the maths package and the routines for accurate off-line spectrum analysis
- (d) Introduce a user-constructed RT-11 file containing spectral data for on-line identification of nuclides giving rise to peaks which the programme has found.

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#### (8) Absolute calibration of X-ray and $\gamma$ -ray detectors (J.G. Cuninghame and H.H. Willis) (AERE))

We have fixed a standard geometry for liquid X- and Y-ray sources consisting of 6 ml of solution in a plastic vial. Using a series of such absolutely standardised sources we have prepared efficiency curves for three Ge(Li) and one Si(Li) detector at various distances. The Ge(Li) detectors are calibrated from 100 keV to 2 MeV and the Si(Li) detector from 5.9 to 136 keV.

# (9) Excitation functions and isomer ratios in the reactions <sup>93</sup>Nb (α, xn) (Jill A.B. Goodall (AERE), C.L. Branquinho, S.M.A. Hoffmann, G.W.A. Newton, V.J. Robinson, H.Y. Wang, I.S. Grant (Manchester University ))

Excitation functions and isomer ratios for 93, 94, 95, 96 Tc from the reactions 93 Nb ( $\alpha$ , xn) have been measured radiochemically for energies from 16 to 72 MeV produced by the Harwell VEC and the Manchester Linac. The excitation functions can be satisfactorily explained by a hybrid model which combines compound nucleus and pre-equilibrium processes, while the addition to this of the spin distribution arising from the initial interaction



Fig. 2 Example, for the (a,2n) reaction, of the comparison between experimental isomer ratios ( $\odot = VEC$ , x = Linac) and those calculated (hatched area) from the combined compound nucleus/pre-equilibrium model with added spin distribution

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gives a fit to the observed isomer ratios; this is illustrated by the example for the  $(\alpha, 2n)$  reaction shown as Fig. 2.



A paper on this work has been submitted to J. Inorg. Nucl. Chem.

Fig. 3 Excitation function for the reaction <sup>93</sup>Nb(a,p3n)<sup>93m</sup>Mo. The broken line shows the calculated to J. Inorg. Nucl. Chem. function

(10) Spallation cross-sections in the system <sup>93</sup>Nb (α, xn) (Jill A.B. Goodall (AERE), C.L. Branquinho, S.M.A. Hoffmann, G.W.A. Newton, V.J. Robinson, I.S. Grant (Manchester University))

Excitation functions were measured radiochemically for six radioactive products formed in reactions of  $9^{3}$ Nb with <sup>4</sup>He ions produced by the Harwell V.E.C. A fair fit to the data is found by using a model which combines compound nucleus and pre-equilibrium nuclear emission. Fig. 3 is an example, for the ( $\alpha$ , p3n) reaction. A paper on this work has been submitted to L Inorg Nucl Chem

(11) An investigation of angular momentum of primary fission fragments of <sup>208</sup>Po\* and <sup>221</sup>Ac\* by radiochemical measurement of isomer ratios (J.G. Cuninghame, Jill A.B. Goodall (AERE), J.E. Freeman, G.W.A. Newton, V.J. Robinson, J.L. Durrell, G.S. Foote, I.S. Grant, M. Rathle (Manchester University ))

We have measured by radio-chemical means isomer ratios of Br and Sb isotopes produced by bombardment of Pt, Pb and Bi targets with <sup>4</sup>He and <sup>12</sup>C beams from the Harwell V.E.C. and Manchester Linac and leading to <sup>208</sup>Po and <sup>221</sup>Ac compound nuclei. We have developed a model for the angular momentum distributions of the primary fragments assuming that the compound nucleus can be treated as a Fermi gas, calculated the isomer ratios from this model and compared the results with our measured ones. Fig. 4 shows this comparison and indicates reasonable agreement between the calculated and measured ratios, although the latter seemed to be systematically high. In this figure the label denotes the interacting nuclei and the observed fragment (ex C + Bi; <sup>126</sup>Sb). The dashed line for the C + Bi; <sup>126</sup>Sb case is the calculated curve if the change in fragment spin during de-excitation of the fragments is neglected. In the <sup>84</sup>Br cases the dashed lines are the calculated curves, assuming a value of 4 for the trapping efficiency parameter of the isomer for the fragments.

A paper on this work has been submitted to J. Phys.

(12) Mass distribution and charge dispersion in <sup>208</sup>Po\* fission measured by radiochemical means (Jill A.B. Goodall (AERE), G.W.A. Newton, J. Hemingway, V.J. Robinson (Manchester University))

The practical measurements on the V.E.C. are now complete and the data are being

assessed.

(13) On-line measurement of variation with energy and angular momentum of total kinetic energy and pre-neutron emission fragment masses in fission of <sup>208</sup>Po\* (J.G. Cuninghame (AERE), J.L. Durell, G.S. Foote, I.S. Grant (Manchester University ))

This work is now complete and is being written up for publication.

(14) On-line measurement of variation with energy and angular momentum of total kinetic energy and pre-neutron emission fission fragment masses in fission of highly fissile compound nuclei (J.G. Cuninghame (AERE), J.L. Durell, G.S. Foote, and I.S. Grant) (Manchester University))

This work is about to restart after being held up because of other commitments

- of the staff concerned.
  - (15) The measurement of v as a function of fission fragment mass and energy in fission of Po\* (J.G. Cuninghame (AERE) J.L. Durell, G.S. Foote, I.S. Grant (Manchester University))

This work is now complete and is being written up for publication

(16) The measurement of v as a function of fission fragment mass and energy in fission of highly fissile compound nuclei (J.G. Cuninghame (AERE) J.L. Durell, G.S. Foote, I.S. Grant (Manchester University))

This work is about to restart after being held up because of other commitments

of the staff concerned.

(17) <u>Measurement of elastic scattering and fission cross sections for fission of <sup>208</sup>Po\* (J.G. Cuninghame (AERE), J.L. Durell, G.S. Foote, I.S. Grant (Manchester University)).</u>

We have made measurements of these cross sections for the reactions of  ${}^{12}$ C on  ${}^{196}$ Pt and  ${}^{16}$ O on  ${}^{192}$ Os but we intend to make them also for  ${}^{4}$ He on  ${}^{204}$ Pb before writing

up the work.

We have now measured the excitation functions for the  $6 \rightarrow 4 \rightarrow 2 \rightarrow 0$  cascade of the even-even isotopes  $^{204}$ Po and  $^{202}$ Po for the reaction  $^{12}$ C +  $^{196}$ Pt in addition to the measurements reported in UKNDC (78)P88 for  $^{4}$ He +  $^{206}$ Pb. We intend to extend the measurements to include the reaction  $^{16}$ O +  $^{192}$ Os before writing up the work.

(1) R. Gunnick and I.B. Niday, UCRL 51061 Vols 1,2,3

<sup>(18)</sup> The competition between fission and neutron emissions in Po compound nuclei (J.G. Cuninghame (AERE), J.L. Durell, G.S. Foote, and I.S. Grant (Manchester University)).



Isomer ratio



Fig. 4 Comparison of measured isomer ratios with values calculated using the Fermi gas model developed during this work. For explanation of the dashed lines see the test. The label on each figure refers to the reacting nuclei then the fragment actually observed (ex C+Bi:<sup>126</sup>Sb)

#### 2.2.2 Neutron Cross Sections

(1) <u>Measurement of neutron cross-sections in PFR (E.A.C. Crouch, I.C. McKean, J.G. Cuninghame, H.H. Willis (AERE) with W. Davies, D.J. Lord, V.M. Sinclair (DNPDE) and N.R. Daly (AWRE))</u>

Some progress has been made in the past year on the work outlined on pages 53-55 of the UKNDC Progress Report for April 1974 to March 1975 - UKNDC (75)P71. Irradiation of some of the capsules containing the high-power experiment has been completed - these are now awaiting dissolutions and analysis. The sub-assemblies containing all the remaining experiments, except the low-power experiments, are still being irradiated.

(2) <u>DFR irradiation of <sup>242</sup>Pu (V.A. Proudler, W. Davies and V.M. Sinclair (DNPDE))</u>

There has been little progress during the year, owing to shortages of effort and facilities. Analysis of the dissolved irradiated  $^{242}$ Pu for isotopes of Pu, Am and Cm is still in hand.

(3) Integral capture cross-section of <sup>241</sup>Am and <sup>243</sup>Am in Zebra (Kathlene M. Glover, R.A.P. Wiltshire (AERE))

Work has continued on the measurement of the integral  $(n,\gamma)$  cross-section of <sup>243</sup>Am during this period. Three <sup>243</sup>Am samples were irradiated in aluminium capsules in the BIZET CFR type core in the ZEBRA reactor at Winfrith. The three samples were processed individually. In each case the sample capsule and contents were dissolved and <sup>244</sup>Cm produced in the irradiation was separated from the aluminium, fission products and americium by a combination of precipitation and ion exchange techniques. The <sup>244</sup>Cm content was determined by alpha spectrometry. The experimental work is now complete and the results await final evaluation.

This data is of world wide interest.

(4) Integral capture cross section of <sup>241</sup>Am and <sup>243</sup>Am in PFR (Kathlene M. Glover, and R.A.P. Wiltshire (AERE))

Samples are currently being irradiated in PFR.

2.2.3 Half-lives

(1) <u>Half life of <sup>239</sup>Pu (Kathlene M. Glover, M. King, R.A.P. Wiltshire, D. Brown (AERE)</u>)

We have nearly completed a final evaluation of the results on the redetermination of the half life by accurate specific activity measurements on samples of Cs<sub>2</sub>PuCl<sub>6</sub>, prepared from 99.1% enriched <sup>239</sup>Pu. This will be published as: 'A redetermination of the half life of plutonium 239 by specific activity measurements.' D. Brown et al, AERE-R 9270 (1978). (2) <u>Half life of <sup>237</sup>Np (Kathlene M. Glover, B. Whittaker, R.A.P. Wiltshire with</u> M.F. Banham, D. Brown, A.J. Fudge (AERE))

Preliminary work on the redetermination of the alpha half life of  $^{237}\mathrm{Np}$  by specific activity measurements on  $Cs_2NpO_2Cl_4$  is in progress. The material has been purified and samples have been provided for  $\gamma$  intensity measurements. Investigations of the effect of Cs on source uniformity and thickness are in progress.

# 2.3 EVALUATIONS AND COMPILATIONS

#### Data File Sub-Committee (B.S.J. Davies (CEGB), Kathlene M. Glover (AERE) M.F. James, A.L. Nichols (AEEW), J.R. Parkinson (BNFL), A. Tobias (CEGB), 2.3.1 D.G. Vallis (AWRE))

'A report by Nichols<sup>(1)</sup> describing the contents of the Chemical Nuclear Data Files has been published together with another <sup>(2)</sup> which lists the decay data for activation products of reactor materials. A paper describing the data files was presented to the International Conference on Neutron Physics and Nuclear Data for Reactor and Other Applied Purposes, Harwell, September 1978.

Table 1 -	UK Cher	nical N	uclear	Data	File
	Status	Table,	Novemb	er 19	978

Data	Present Status	File Development
1. F.P. Decay Data	Exists as UKFPDD/1. Derived from merging Tobias 1973 data with US ENDF/B4	Revised data for about 100 nuclides available. Final check- ing in decay heat calculations required.
2. Activation Products of Structural Materials Decay Data	Available in ENDF/B4 format for 91 nuclides.	None planned.
<ol> <li>Heavy Element and Actinide Decay Data</li> </ol>	None in file	Evaluation for 119 nuclides being done by Nichols (AEEW) in ENDF/B5 format. 68 complete so far.
4. Decay Data for Other Nuclides	None in file	No effort available
5. Fission Yields	Available in ENDF/B4 format, based on Crouch's second round, for 7 fissile nuclides. Data refers to fission product ground states only.	Further round of adjustment under way, based on revised criteria. Data for isomeric states of fission products being evaluated.
6. Delayed Neutrons	Tomlinson data are still recomm- ended	New evaluation by Crouch under way

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Nichols is currently evaluating decay data for actinides and heavy elements. Except for spontaneous fission data, the evaluation process has been completed for 68 nuclides out of a target 119. These data are in ENDF/B5 format in order to accommodate data related to spontaneous fission.

Revised data for about 100 fission product nuclides have been provided by Davies and Parkinson.

The status of this data file at the end of 1978 is shown in table one.

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(1) A.L. Nichols, AERE	R-8904	i a i	· .			
(2) A.L. Nichols, AERE	R-8903					
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#### 2.3.2 Fission yields

#### (1) Fission product yields: assessments and consistent sets (E.A.C. Crouch (AERE))

Although experimentally determined fission product chain and independent yields are reported in the literature at a steady rate, and the fission product library is steadily updated, there are still not enough experimental data available to give direct estimates of all the fission product yields of interest to reactor physicists. The method used to provide the necessary information on the basis of what is known consists of finding adjustments to chain yields (interpolated or extrapolated as necessary) and to independent yields deduced by estimating parameters from the known independent yields. The adjustments are derived by applications of physical conservation laws and are not based on any particular model. Using the results of this method very good agreement was obtained with experimental values for the after-heat in power reactors (1,2). However. further constraints have been applied by means of Dr. J.R. Reid's (of Computer Science and Systems Division, AERE) sparse matrix techniques and with his programming help. It is hoped to test these results soon. The method is now based on minimising  $\chi^2$  as described previously (UKNDC 78 P88,p99) with the added constraint that the sum of the yield of complementary elements must be equal.

The constraints do not include the effect of delayed neutron emissions nor that of isomeric yields. Delayed neutron emission is currently being studied and a paper on isomeric yield estimation has been submitted to the chemical Nuclear Data Committee for an opinion on the method of calculating isomeric yields in the absence of direct experimental results.

The U.K. Chemical Nuclear Data Files: An evaluated set of radioactive decay data for Reactor Applications. B.S.J. Davies, E.A.C. Crouch, M.F. James, A.L. Nichols, J.R. Parkinson, A. Tobias and D.G. Vallis

(2) Int. Conf. on Neutron Physics, Neutron Data for Reactors and other applied purposes. Harwell, September 1978

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# 2.3.3 Related compilation studies

(1) The CASCADE programme (G. Evangelides (Imperial College) and D.G. Vallis (AWRE))

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The computer programme CASCADE interprets evaluated decay schemes in terms of radiation data sets. The objective of this work is to expand and improve the code and to provide a data base for input to the code.

All the major modifications to the original version have now been completed and the programme is being tested in all its options.

The code can now handle any size problem provided the core limitation of the computer being used is not exceeded. This is made possible by the use of variable dimension format. If however the core required exceeds the core available the programme will automatically reduce the problem size according to preset parameters till it does fit the core available. Any nuclide requiring more core will be skipped and the next nuclide studied.

The programme can now fully evaluate proton and neutron emission and nuclides decaying by spontaneous fission. X-rays and anihilation radiation as well as conversion and Auger electrons can also be treated and results are now produced in the ENDF/B-IV type format.

An error in the calculation of the intensity of requested sets of coincident emissions has now been corrected. A subroutine is being developed to automatically calculate all the major combinations of coincidence emissions of the radiations emitted by any nuclide.

The input data base is provided for the CASCADE programme from NSDF/ENSDF compilations. The programme for this corrects for certain systematic errors in the original NSDF compilations and adds a header section to the data for easier evaluation for CASCADE input format.

The original CASCADE input format has been slightly modified. A daughter nuclide description has been added as well as a normalisation factor, thus enabling CASCADE to correctly evaluate intensities of nuclides with any number of decay modes. The secondary transition data can now be produced in a condensed format. That is if the values for the various emission possibilities are all zero then that transition can now be excluded except for the transition of the last level to the last but one level which

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denotes the end of the secondary transition data.

The internal conversion coefficients of Band et  $a1^{(1)}$  are being written to a direct access device in a similar manner to the Hager and Seltzer internal conversion coefficients (2) and will be obtained via the same subroutine in CASCADE which has already been modified . · 

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to do this.

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The SORT/MERGE routines have now been combined into one which will also convert the

old CASCADE radiation catalogues to the same format as the new ones which include both

end point and average energies or recoil energies in the case of alpha, proton and neutron والمنافع موجوا والمراجع emission. A couple of extra parameters have also been added to the catalogues.

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 I.M. Band, M.A. Listengarten and M.B. Trzhaskouskaya, Internal conversion coefficients for atomic numbers Z < 30, LIYA F-235 (in Russian) (1976), INDC(CCP)-92IN (1976),</li> ADNDT <u>18</u> (1976) 433

\* (a. 4-00 -(2) R.H. Hager & E.C. Seltzer, International conversion coefficient tables. I: NDT A4(1968) 1 and II: NDT A4 (1968)397

# 3. REACTOR PHYSICS DIVISION, AEE, WINFRITH (Division head: Dr. C.G. Campbell)

#### 3.1 Activation cross-section evaluations for the UKNDL (J.S. Story)

As part of a study of activation products in Advanced Gas-cooled Reactors, nuclear data files have been produced for the  ${}^{40}$ Ar(n, $\gamma$ ),  ${}^{50}$ Cr(n, $\gamma$ ) and  ${}^{35}$ Cl(n,p) reactions.  ${}^{40}$ Ar(n, $\gamma$ ). This is a straightforward conversion of the capture data in the ENDL file No.7148, which tabulates the cross-section at 72 points between 0.1 MeV and 20 MeV. These data were taken over without change, although a change in the interpolation rule between data points is involved; it is felt that the log x log interpolation rule used in the UKNDL is more realistic in this particular application. The cross-section of 0.66 barns at 0.025 eV is reasonably consistent with the thermal cross-section given in BNL-325 (1973); on the other hand the data file gives a relatively low resonance integral of 0.306 barns, as compared with 0.41  $\pm$  0.03 barns in BNL-325. For a more realistic file a careful study of the resonance parameter data would be necessary.

 $\sum_{i=1}^{50} Cr(\underline{n},\underline{\gamma})$ . Cross-section data for this reaction have been obtained directly from the graph shown in BNL-325 (1976) between 2.9 eV and 57 keV, at energies close enough for a good representation with the log x log interpolation rule. A cross-section of 15.9 barns at 0.0253 eV was included, and the file then extended to cover the range 0.1 MeV to 20 MeV by assuming a 1/v variation below 0.0253 eV and above 57 keV. It is proposed to merge the new files with the (n,2n), (n,p) and  $(n,\alpha)$  cross-sections already tabulated in DFN-936. <sup>35</sup>Cl(n,p). Cross-sections for this reaction were obtained directly from the graph in BNL-325 (1976), which covers the range 0.025 eV to 8 keV; at 0.0253eV the cross-section was taken as 0.49 barns. Data above 10 keV were obtained by tabulating the group crosssections from the RSIC data set (RW Roussin and DW Muir, ORNL DLC-33 (Nov. 1976)) at the mid lethargy point of each group. A fission spectrum averaged cross-section of 19mb was calculated from the file, whereas Calamand (IAEA technical report series No. 156, p 273) gives a value of 78 mb. It is felt that the latter value may include the (n,pn) reaction as well as (n,py). The new tabulation, from 0.1 MeV to 14.2 MeV will be added to the existing  $(n,\alpha)$  compilation in DFN-231.

3.2 AEE Counting Laboratory

# 3.2.1 $\beta$ -Power from fast neutron fission of <sup>239</sup>Pu and <sup>235</sup>U (W.H. Taylor, M.F. Murphy, <u>D.W. Sweet and M.R. March</u>)

The final results of the determinations of the  $\beta$ -power from the fragments from the fast neutron fission of <sup>239</sup>Pu and <sup>235</sup>U are given in AEEW R1212

#### 3.2.2 Thermal cross-sections (W.H. Taylor, M.F. Murphy and M.R. March)

Measurements primarily aimed at confirming the absolute detection efficiencies of  $\gamma$ -ray spectrometers also gave values of the thermal cross-section ( $\sigma_{2200m}$ .g) for the reactions  $^{239}$ Pu (n,f),  $^{235}$ U (n,f),  $^{55}$ Mn (n, $\gamma$ ),  $^{181}$ Ta (n, $\gamma$ ). These results showed discrepancies of only 1.5%, 1.2%, 0.8%, and 0.5% respectively with the presently preferred best values; for the two fission cross-sections these were selected by J.S. Story from the data given in the two papers by H.D. Lemmel<sup>(1,2)</sup>. This work is fully reported in RPD/WHT/P333.

3.2.3 Cross-section for <sup>103</sup>Rh (n,n')<sup>103m</sup>Rh (W.H. Taylor, M.F. Murphy and M.R. March)

The value of the cross-section for the 103 Rh (n,n')103 m Rh reaction has been tested by comparing the results of reaction rate determinations in Zebra with the values calculated using collision probability methods. The details of this comparison are given below.

· · ·	Reaction Rate Ratio	$\frac{Rh}{F_8}$	$\frac{F_8}{F_5}$	F <sub>9</sub> F <sub>5</sub>
	Calculated Value (C)	3.45	0.02763	1.045
	Experimental Value (E)	3.57 <u>+</u> 2.9%	0.02652 <u>+</u> 0.3%R 0.6%S	1.045 <u>+</u> 0.2%R 1.0%S
•	C/E	0.97	1.042	1.00

where  $F_8 = {}^{238}$ U (n,f),  $F_9 = {}^{239}$ Pu (n,f),  $F_5 = {}^{235}$ U (n,f) and Rh =  ${}^{103}$ Rh (n,n')  ${}^{103m}$ Rh. The good agreement obtained for the fission rate ratio indicates that the methods of calculation are adequate. The C/E value of 0.97 for Rh/F<sub>8</sub> is a confirmation that the cross-section data and the experimental method are accurate. The X-ray spectrometer was calibrated using a  ${}^{103}$ Pd -  ${}^{103m}$ Rh source which was calibrated by the I.A.E.A. using a  ${}^{4\pi e-X-ray}$  coincidence method which is essentially independant of the branching ratio for K-Xrays; and the DFN94 cross section data for Rh was used in the calculations (averaged with fission-spectrum weighting above 0.82 MeV and with  $\frac{1}{E}$  weighting from below 0.82 MeV down to the threshold for the reaction). It is relevant to note that the value for the total number of K X-rays produced per 100 disintegrations of  ${}^{103m}$ Rh of 6.76  $\pm$  0.05 obtained by the I.A.E.A.  ${}^{(3)}$ is in good agreement with the value of 6.97  $\pm$ 0.28 obtained by Butler and Santry  ${}^{(4)}$  when they measured the  ${}^{103}$ Rh (n,n') cross section.

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The presently accepted value for this parameter is 7.7 + 29 (5) -22 (5)

\_\_\_\_ 5 (1) H.D. Lemmel, Washington Conference of Nuclear cross-sections and Technology; NBS special publication 425, 1, 286 (1975) . . .

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(2) H.D. Lemmel, NBS special publication 493, 28 (1977)

(3) K.H. Czoek, I.A.E.A/RL/26 August (1974) •

- (4) D.C. Santry and J.P. Bulter, Can. J. Phys. <u>52</u> 1421 (1974)
- (5) A. McNair, private communication, December 1976

4. DIVISION OF RADIATION SCIENCE AND ACOUSTICS, NATIONAL PHYSICAL LABORATORY (Superintendent: Dr. W.A. Jennings)

Various methods of measuring neutron fluence rate are being developed, employed and intercompared as a means of improving the accuracy of the existing standards. 4.1 Neutrons in the 1-1000 eV energy range (E.J. Axton and A.G. Bardell)

As reported last year, no further progress has been made in the provision of a low energy neutron source based on the Van de Graaff accelerator as the machine has not been available for this work.

Measurements to determine the importance of the contribution of neutrons in this energy range to the total dose equivalent in a typical power station location have been completed. The measurements were made using polyethylene spheres with gold foils at the centre. The efficiency matrix for the spheres was determined by Monte Carlo calculations using ENDF/B4 neutron cross-section data, supported by calibrations at a few neutron energies. The same detectors were exposed in the slowing-down neutron spectrum from the NPL thermal column.

In order to determine the neutron spectrum the measurements were expressed in terms of Fredholm inequalities, viz:

 $B_{i} - \Delta B_{i} < \Sigma \Psi \quad \varepsilon' < B_{i} + \Delta B_{i}$ 

where  $B_i$  is the gold foil capture rate in the i-th sphere,  $\Psi_j$  is the neutron fluence rate in the j-th energy bin and  $\varepsilon_{ij}$  is the neutron capture rate in the gold foil of the i-th sphere for unit neutron fluence in the j-th energy bin. The 'tolerance'  $\Delta B_i$ is introduced to take account of uncertainties in the values of  $\varepsilon_{ij}$  and  $B_i$ . The fluence rates in bins containing the energies of the principal capture resonances of gold and manganese were determined from cadmium ratio measurements with gold and manganese foils, and various smoothing constraints were applied which led to unique and plausible solutions for the remaining thirty-four unknown energy bin fluence rates per unit lethargy. The results<sup>(1)</sup> demonstrated that the spectrum of the NPL thermal column and that produced by a 1/E-shaped spectrum terminating at about 6 MeV were equivalent in terms of doseequivalent. On the other hand, the neutron spectrum at the nuclear power station location is much softer; no more than one third of the dose equivalent can be attributed to neutrons of energy above 100 keV.

The need for neutron standards in the energy range below 100 keV is clearly demonstrated.

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(1) E.J. Axton and A.G. Bardell, "Neutron standards available at the National Physical Laboratory and the need for low energy standards", Proceedings of an Advisory group meeting on Neutron Reactor Dosimetry, I.A.E.A. Vienna, 13-17 Nov. 1978. To be 'published

#### 4.2 Intermediate energy neutrons (J.B. Hunt)

The measurement of neutron detection efficiency of the NPL standard long counter and of a De Pangher long counter at 920 keV, 727 KeV and 582 KeV, using the associated target activity technique has been completed and published <sup>(1)</sup>.

The efficiency of a De Pangher long counter has also been measured at 2 keV and 25 keV using the reactor filtered beams available at Physikalisch-Technische Bundesanstalt (P.T.B.), West Germany. The result at 25 keV appears to be 22% higher than that obtained using an absolutely calibrated SbBe ( $\gamma$ ,n) source. The possible reasons for this discrepancy are being investigated.

(1) J.B. Hunt and R.A. Mercer, Nucl. Instr. Meths. <u>156</u> (1978) 451

4.3 Fast Neutron Energies (T.B. Ryves, K.J. Zieba and P. Kolkowski)

The cross-section measurements for the reactions  ${}^{14}N(n,2n){}^{13}N$ ,  ${}^{19}F(n,2n){}^{18}F$ ,  ${}^{54}Fe(n,2n){}^{53}Fe$ ,  ${}^{27}Al(n,p){}^{27}Mg$  and  ${}^{27}Al(n,\alpha){}^{24}Na$  between 14.7 and 19.0 MeV have been completed and published <sup>(1)</sup>.

Further activation cross-sections are now being measured, including the (n,2n) reactions on Nb, Ta, Au, and  $^{238}$ U. An attempt will be made to measure the branching ratios in the decay of  $^{108m}$ Ta.

(1) T.B. Ryves, K.J. Zieba and P. Kolkowski, J. Phys. G. <u>4</u> (1978) 1783

# 4.4 Thermal neutron capture cross-section for the production of $^{108m}$ Ag(T.B. Ryves)

In reactor decomissioning calculations, the production of 127y  $^{108m}$ Ag from trace amounts of silver in the stainless steel structure of the reactor is very significant: The thermal (2200ms<sup>-1</sup>) neutron capture cross-section of  $^{107}$ Ag for the production of metastable  $^{108m}$ Ag has been measured as (0.366 ± 0.065)b, where the uncertainty at approximately 99% confidence limits is nearly all due to the half-life. The previously published value in BNL 325 third edition was (3.0 ± 1.5)b, which is an order of magnitude larger. In many practical instances, the operational life, T, of the reactor will be

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much less than the <sup>108m</sup>Ag half-life, and in these cases the estimated unsaturated activity build-up of <sup>108m</sup>Ag is proportional to  $\sigma_0 \{1-e^{-\lambda T}\} \simeq \sigma_0 \lambda T$  where  $\lambda$  is the <sup>105m</sup>Ag decay constant. Due to the correlation in our experiment, the product  $\sigma_0 \lambda$  has been measured far more accurately than the cross section, to about ± 5% uncertainty.

The ratios of the reduced resonance integrals to the  $2200 \text{ms}^{-1}$  neutron capture crosssection were also determined to be (3.0 ± 0.5) for  ${}^{108\text{m}}\text{Ag}$  and (15.5 ± 0.8) for  ${}^{110}\text{Ag}$ .

#### 4.5 Neutron source calibrations (E.J. Axton and A.G. Bardell)

Improvements in analytical techniques have demonstrated the presence of some hitherto undetected but significant impurities in the manganese sulphate bath solution. The main contaminants are small traces of lithium and cadmium. The solution is being replaced by higher grade material in time for the measurements which will form part of the forthcoming international comparison of neutron emission ratesorganised by the International Bureau of Weights and Measures (B.I.P.M.) over the period 1978-1980. Eventually sources previously measured at NPL will be revalued upwards by an amount between 0.3 and 0.5%

#### 4.6' International comparison of neutron fluence rate

The international comparison of neutron flux density arranged by the International Bureau of Weights and Measures (starting in late 1974) is now formally closed, and a report of the results has been published <sup>(1)</sup>.

A further intercomparison was proposed in 1977 to take place from 1979 onwards, the interim two years being devoted to a study of transfer methods. Unlike the previous intercomparison, the 1979 exercise is planned to include "white spectrum linac" neutron sources as well as mono-energetic neutron from Van de Graaff accelerators. To this end a suitable fission chamber is being designed and built at A.E.R.E. Harwell.

1. V.D. Huynh, NBS Special Publication 493 (1977) 244

#### 4.7 <u>Photoneutron production in high energy electron and X-ray beams used for radiotherapy</u> (E.J. Axton and A.G. Bardell)

Measurements are in progress to measure the photoneutron contamination of the SL75/20 linear accelerator (made by the M.E.L. Equipment Co Ltd) at Addenbrookes Hospital, Cambridge, by means of the multi-sphere technique described earlier. The measurements should yield spectral information which would allow a more accurate assessment of the

neutron dose-equivalent received by a patient under-going treatment than was possible in the second value of the second value of the second value of the second value of the second seco

(1) E.J. Axton and A.G. Bardell, Phys. Med. Biol. <u>17</u> (1972) 293

4.8 <u>Reference standard for fast neutron dosimetry (V.E. Lewis, E.J. Axton, D.J.-Young,</u> I.W. Goodier and J.L. Makepeace)

Using the purpose built Dosimetry accelerator, collimated beams of 14.7 MeV neutrons have been produced with dose rates of up to  $70\mu$ Gy s<sup>-1</sup> at the calibration position. A duoplasmatron ion source capable of increasing this by a factor 4 is being tested. Calibration of the neutron fields with ionisation chambers and Geiger-Müller dosemeters is in progress. (1 Gy = 1 Gray = 100R: Editor)

The neutron sensitivities for different cavity ionisation chambers (of tissue equivalent plastic, graphite and magnesium) have been measured for energies from 13.6 to 14.7 MeV in the low gamma component d + T neutron field produced in a low-scatter environment using the SAMES accelerator.

A dual thermoluminescent phosphor system for the measurement of gamma dose components of mixed fields, not dependent on the use of another neutron sensitive dosemeter, has been tested and shows potential as a transfer device. The activation of small niobium ingots as a technique for the measurement of the primary component of collimated d + T neutron beams has been found very useful, and has potential as a transfer standard.

Thermally stimulated exo-electron (TSEE) dosimetry is being investigated as a method for measuring the neutron and gamma-ray components of a mixed radiation field.

4.9 <u>Nuclear decay scheme measurements ( P. Christmas, D. Smith, M.J. Woods, R.A. Mercer,</u> <u>P. Cross and S.P. Brown</u>)

A high pressure proportional counter has been constructed for incorporation into a  $4\pi\beta-\gamma$  coincidence system. A preliminary value has been obtained for  $P_k^{}\omega_k^{}$  for  $^{55}$ Fe, by separately measuring the K X-ray emission and the absolute disintegration rate. This method promises to give a final value considerably more accurate than any previously available.

4.10 W-value measurements (P. Christmas, M. Burke, D.J. Thomas and I.R. Brearley (Birmingham University))

At Birmingham preliminary results have been obtained for protons of energy between

0.75 and 2.5 MeV in argon, methane and nitrogen. The precision of these results compares favourably with that of earlier measurements elsewhere, despite some difficulties associated with some as yet unresolved systematic effects. The Birmingham work has ceased for the present due to the departure of Dr. Brearley, but the NPL programme will continue.

#### 5. CEGB RESEARCH DIVISION, BERKELEY NUCLEAR LABORATORIES (Section head: Dr. B.M. Wheatley)

#### 5.1 The inventories of actinide and fission product isotopes in spent nuclear fuels: results of calculations using RICE (J.H. Mairs and S. Nair)

The Reactor Inventory Code RICE has been used to make comprehensive calculations of the actinide and fission product arisings in irradiated fuels from Magnox, AGR, PWR and Fast Breeder Reactors. Fuel inventories for Magnox, AGR and CFR (LMFBR) type reactors have been generated using newly derived neutron cross-section data sets. The WIMS and ARGOSY reactor lattice codes were used for the evaluation of the cross-sections for Magnox and ACR reactors, respectively.

Studies have been made of the actinide and fission product activity contributions for spent magnox fuel cooled for both short and long (geological) periods and for the radioactive waste arisings with and without uranium and plutonium reprocessing. The dependence of the  $^{134}$ Cs/ $^{137}$ Cs activity ratio on irradiation conditions was studied in detail for the magnox reactor. The dependence of the actinide build-up on increasing irradiation was also studied, particularly the build-up of the americium and curium isotopes. Similar calculations for AGR and CFR have been compared with the results for the Magnox case. Additionally, for CFR, studies were made of the dependence of the actinide  $\alpha$  heating from the spent fuel on the shelf-life of the fresh fuel prior to irradiation in CFR. The dependence of the spent fuel actinide inventories on the source of plutonium used in the fresh fuel was also studied for plutonium originating from Magnox, AGR and equilibrium CFR cycles.

One PWR inventory has been listed but the HTR was not considered since this system is of marginal interest to the CEGB at present.

It is anticipated that a comprehensive inventory listing and discussion of the information mentioned above will appear as a CEGB report in the near future.

#### 5.2 <u>Magnox fuel inventories: experiment and calculation using a point source model</u> (S. Nair & J.R. Harvey)

The results of calculations of Magnox fuel inventories using the RICE code, based on a simple point source model, and associated Magnox reactor data set have been compared with experimental measurements for four samples of spent Magnox fuel spanning the burnup range 3000-9000 MWd/Te. We considered the actinide isotopes <sup>234,235,236,238</sup>U, <sup>238,239,240,241,242</sup>Pu, <sup>241,243</sup>Am and <sup>242,244</sup>Cm, and the fission product isotopes <sup>142,143,144,145,146,150</sup>Nd, <sup>95</sup>Zr, <sup>134,137</sup>Cs, <sup>144</sup>Ce and daughter <sup>144</sup>Pr. The neutron

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emissions from a further two samples were also measured and compared with RICE predictions.

The uranium isotopic compositions determined by calculation and experiment are in good accord. The plutonium isotopic compositions differ by less than 14%, while the <sup>241,243</sup>Am predictions agree with experiment, within the errors. The <sup>242,244</sup>Cm predictions and the neutron emission do not differ by more than 25% for the samples with burnup exceeding 7000 MWd/Te. However, larger discrepancies are observed for the sample with burnup around 3000 MWd/Te. Good agreement was observed between experiment and calculation for the fission products considered in the present study. The results of the comparison were considered to be such as to justify the use of the code RICE in providing source terms for environmental impact studies, for the isotopes considered in the present work.

The work has been written up as CEGB report RD/B/N4349. A companion report, AERE-R-9212, describes the experimental techniques used by Foster et al. to analyse the samples.

#### 5.3 A preliminary assessment of the waste disposal problem for magnox steels (S. Nair)

A methodology has been described which can be used to assess the relative radiological toxicities of activation products over geological timescales. A computer code, called STRUMP, has been written to assist in this assessment. The methodology has been applied to an evaluation of the radiological implications of the waste disposal problem for decommissioned fixed steel components in Magnox reactors. The study concluded that for timescales up to 400,000 years the radiotoxicity of the activated steels was dominated, to a varying extent, by the isotopes  $^{63}$ Ni and  $^{59}$ Ni. An attempt was made to estimate toxicity levels corresponding to the activity limit of  $10^{-11}$  Ci gm<sup>-1</sup> below which radioisotopes may be considered "stable". The study indicated that steels activated in conditions typical of those encountered by large mass components in the core mid-plane of the core restraint structure may be considered "stable" and thus disposed without restriction only after about a million years.

This work has been written up as CEGB report RD/B/N4291.

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# 6. THE UNIVERSITY OF ASTON IN BIRMINGHAM, DEPARTMENT OF PHYSICS

#### (Head of Department: Professor S.E. Hunt)

# 6.1 <u>A study of the elastic scattering of 2 to 3 MeV neutrons by <sup>40</sup>Ca and <sup>32</sup>S using</u> an associated particle time of flight spectrometer (M.C. Janicki & A.J. Cox)

#### (1) Summary

An associated particle time of flight spectrometer based on the  $D(d,n)^{3}$ He reaction has been developed using thin deuterated polythene targets. The competing  $D(d,p)^{3}T$ reaction was used to monitor the neutron output. The spectrometer has been used to study the elastic scattering of 2-3 MeV neutrons in  $^{40}$ Ca and  $^{32}$ S; in each case the differential elastic scattering cross-sections have been measured.

In the angular range  $20^{\circ} - 100^{\circ}$  in the laboratory frame of reference, the results have been compared with an optical model analysis. Values of  $\chi^2$  per point of 3.46 and 2.84 were obtained for  ${}^{40}$ C and  ${}^{32}$ S respectively.

#### (2) Introduction

In the present work an associated particle time of flight spectrometer is described which enables differential elastic scattering cross-sections for 2 to 3 MeV neutrons to be made. Neutrons were produced using the  $D(d,n)^3$ He reaction, the deuterons being accelerated in the 3 MeV dynamitron at the Joint Aston and Birmingham Universities' Radiation Centre. The deuterium target consisted of a thin film of deuterated polythene to reduce the intensity of scattered deuterons and prevent the detectors saturating. The spectrometer has been used to study the elastic scattering of 2-3 MeV neutrons by Ca and S in an angular range  $20^{\circ} - 100^{\circ}$ . These two elements were chosen as only limited published data was available and because the separation of the ground states and the first excited states enabled any inelastic contribution to be eliminated. In the case of calcium, elastic cross-sections had been measured at neutron energies of 2.83 MeV (Abramson et al)<sup>(1)</sup> and 3.29 MeV (Reber and Brandenburger)<sup>(2)</sup> but no optical model analysis had been performed for the former energy. For sulphur there was an absence of both published data on the differential cross-sections for neutron energies below 2.4 MeV and of an optical model analysis.

#### (3) Experimental details

Neutrons with energies of between 2 MeV and 3 MeV were produced via the  ${}^{2}$ H(d,n) ${}^{3}$ He reaction using 1 MeV deuterons incident on thin deuterated polythylene targets. The deuterons were accelerated by a 3 MeV Dynamitron and a maximum target thickness of 2µm was employed to reduce the intensity of deuterons scattered into the charged particle

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detector. The <sup>3</sup>He particles associated with neutron production and used to give the zero-time signal, were detected by a silicon surface barrier detector with a sensitive depth of 100µm. The detector was used in conjunction with a charge sensitive preamplifier.

The effective neutron energy was changed by altering the angles of the neutron and charged particle detectors with respect to the incident deuteron beam. The neutron beam associated with the detected <sup>3</sup>He particles was incident on to a flat plate scattering sample placed at a distance of 30cm from the deuterated polyethylene target. Scattered neutrons were detected by a detector placed at 50cm from the sample. The detector consisted of a cylinder of NE 102A 5cm thick, 10cm in diameter, mounted on to a 56 AVP photomultiplier tube via a conical-shaped perspex light-guide. The photomultiplier tube was screened from stray magnetic fields by a  $\mu$ -metal shield and from spurious neutron and other radiation by a concentric cylindrical shield of paraffin wax and boric oxide.

The calcium scattering sample was made by compressing natural calcium granules into a rectangular tin plated mild steel container of wall thickness 0.5mm. The enclosed sample measured 13cm by 18cm and was 5cm thick. An identical empty container was used for background measurements. The sulphur scattering sample was natural sulphur moulded into a slab 12.5cm x 9.5cm x 2.8cm. X-ray examinations showed an absence of voids in both the samples.

Time-of-flight spectra were obtained using a standard electronic gating system shown in Figure 1. Signals from the charged particle detector, after suitable amplification and shaping were used as the start pulses for the time-to-pulse height converter (NE 4670). The stop pulse was obtained from the neutron detector after suitable shaping and discrimination. The total number of neutrons, at an energy of 2.18 MeV, incident on to the scattering sample of sulphur was calculated from the number of <sup>3</sup>He particles detected. This was possible because for a neutron energy of 2.18 MeV the associated <sup>3</sup>He particles were well resolved from other charged particle groups and a simple electronic window across the <sup>3</sup>He energy range gave the number of neutrons incident on to the scattering sample. At a neutron energy of 2.99 MeV however, the associated <sup>3</sup>He particles were not resolved from deuterons elastically scattered from carbon in the target. To overcome this the protons produced in the competing  ${}^{2}H(d,p){}^{3}H$  reaction were counted and related to the yield of  ${}^{3}$ He particles by the reaction cross-sections. The output of the timeto-pulse-height converter was proportional to the time difference between the  $^3$ He particle signal and the neutron signal. The resulting coincidence resolution curves had a full width at maximum of 65ns and 31.4ns for neutron energies of 2.99 MeV and 2.18 MeV respectively.

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The efficiency of the neutron detector for 2.18 MeV and 2.99 MeV neutrons was measured using the direct neutron beam. The centre of the face of the neutron detector was positioned 30 cm from the target centre, at  $0^{\circ}$ , to completely encompass the defined cone of associated neutrons. The efficiency of the neutron detector was then calculated as the ratio of the total number of coincidences to the total number of neutrons incident on the detector, as measured by the charged particle detector.

#### (4) Experimental procedure & results

Each sample was placed 30cm from the centre of the deuterated polyethylene target at an angle of  $45^{\circ}$  to the incident beam direction and with its vertical axis perpendicular to the horizontal plane in which the neutrons were observed. With this geometry, time-offlight spectra were recorded for laboratory scattering angles in the range  $20^{\circ}$  to  $100^{\circ}$ . Background corrections were applied by making alternate measurements with the sample in position and then with the sample removed. In each case a normal error distribution was fitted to the elastic scattering peak.

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The differential elastic scattering cross-section  $\sigma(\theta)$  in the laboratory system is



given by

Fig. 2a Differential cross-section for elastic scattering from calcium. The full curve is the optical-model prediction corrected for compound elastic scattering; O Reber and Brandenburger; A Abramson et al

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where  $N(\theta)$  is the neutron detector count,  $\phi$ is the total number of neutrons incident on the sample, n is the number of nuclei/cm $^3$ to of scatterer,  $\Delta \ \Omega$  is the solid angle subtended by the neutron detector at the sample and  $\varepsilon(E_n)$  is the detector efficiency as a function of neutron energy,  $\Sigma(r_i x_i)$  is the weighted average sample thickness taking account of the neutron beam divergence in the sample calculated using the reaction kinetics, r, defines the ratio of the area under the neutron beam profile curve in a small angular interval to the total area under the curve and x, denotes the sample thickness in the i th angular interval. The differential elastic scattering cross-section results are shown in figures 2a and 2b,

The results were compared to the predictions of the optical model using the computer code RAROMP after first removing the compound elastic contribution to the data. The compound elastic cross-sections have been found by Wilmore and Hodgson<sup>(3).</sup>

to be fairly isotropic in the energy region

under consideration with values of 84mb/sr and 50 mb/sr which were subtracted from the experimental data for calcium and sulphur respectively. The differential cross-sections for both calcium and sulphur were then calculated using the optical model parameter values of Becchetti and Greenlees<sup>(4)</sup>.



Fig. 2b Differential cross-section for elastic scattering from sulphur. The full curve is the optical model prediction corrected for compound elastic scattering; O Holmqvist and Wiedling

The comparisons of the experimental data and theoretical predictions show that the predictions are acceptable giving a value of  $\chi^2/$ point of 3.46 for calcium and 2.84 for sulphur. In view of the experimental uncertainties and the fact that the parameters of Becchetti and Greenlees represent parameters that are applicable to a large range of nuclei, it is felt that these give useful optical model comparisons.

#### References

(1) D. Abramson et al; EANDC(E) -149 (1971)

- (2) J.D. Reber and J.B. Brandenburger, Phys. Rev., 163 (1967) 1077
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#### 7. BIRMINGHAM RADIATION CENTRE, UNIVERSITY OF BIRMINGHAM

#### (Director: Professor J. Walker)

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7.1 Delayed neutron energy spectra (J.G. Owen, D.R. Weaver and J. Walker ) [Relevant to requests: 765-6]

The delayed neutron energy spectrum following fast neutron induced fission of <sup>235</sup>U has been measured using a gridded <sup>3</sup>He-ionisation chamber of the Shalev type employing pulse rise-time analysis<sup>(1)</sup>. The uranium was in the form of a 104 g metallic cylinder, enriched to 96.54% <sup>235</sup>U, and kindly loaned by AERE, Harwell. Suitable primary neutron fluxes have been produced by bombardment of tritiated or deuterated targets by either a proton or deuteron beam produced by the Birmingham Dynamitron Accelerator. The beam was pulsed 0.8 seconds on, 1.0 seconds off to generate a near equilibrium distribution of the delayed neutron groups, with counting occurring for 0.8 seconds during the beam off period. Four primary neutron energies, 0.940, 1.440, 1.760 and 6.0 MeV, have been employed, and the results are illustrated in Figures 1 and 2.

Figure 1 shows the three lower energy fission cases, whilst in Figure 2 the 6.0 MeV spectrum is compared to that obtained at 1.760 MeV. Over the mid-energy range a typical uncertainty of  $\pm$  7% exists on each energy group, but this increases to between  $\pm$  15 and  $\pm$  20% on the high energy tail. Also shown in Figure 2 is the delayed neutron spectra following thermal fission predicted by SAPHIER et al<sup>(2)</sup>.

The overall shape of the spectra measured in this work remains reasonably constant, but the relative intensities of several peaks, or groups of peaks, does vary considerably between the different fission cases. It is believed that these variations reflect changes in the fission yields of the delayed neutron precursors with fission energy

A considerable fraction of the delayed neutrons are seen to have energies below 100 keV, in general agreement with the work of ECCLESTON and WOODRUFF<sup>(3)</sup> (Fig. 3), although in disagreement with the majority of previous work. Whilst the possibility of neutron contamination in these low energy groups as a result of scattering in the material surrounding the detector cannot completely be ruled out, it seems unlikely that such a correction will substantially change the shape of the spectra. Work is currently underway to assess the magnitude of this contribution.

Many delayed neutrons are also observed above 1 MeV. It is believed that this is the result of the inclusion of significant yields from the shorter-lived delayed neutron groups, coupled with the use of a detector able to respond to neutrons in this





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Fig. 2 Comparison of measured and predicted delayed neutron spectra

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. ' energy region. These conclusions seem to be at least partly verified by the work of EVANS and KRICK<sup>(4)</sup>.

- (1) J.G. Owen, J. Walker and D.R. Weaver, "Energy Spectra of Delayed Neutrons from Uranium Fission". Proc. Int. Conf. on Neutron Physics and Nuclear Data for Reactors and Other Applied Purposes, Harwell, Sept. 1978
- (2) D. Saphier, D. Ilberg, S. Shalev and S. Yiftah, "Evaluated Delayed Neutron" Spectra and their Importance in Reactor Calculations", Nucl. Sci. Eng. 62 (1977) 660
- (3) G.W. Eccleston and G.L. Woodruff, "Measured Near-Equilibrium Delayed Neutron Spectra Produced by Th, 235U, 238U and 239Pu", Nucl. Sci. Eng. 62 (1977) 636
- A.E. Evans and M.S. Krick, "Equilibrium Delayed Neutron Spectra from Fast Fission of  $^{235}$ U and  $^{239}$ Pu". Nucl. Sci. Eng. <u>62</u> (1977) 652 (4)



Fig. 3 Comparison of the delayed neutron spectrum measured in this work with that of Eccleston & Woodruff

8. UNIVERSITY OF BIRMINGHAM, DEPARTMENT OF PHYSICS (Professor of Applied Nuclear Science: J. Malker)

- 8. Experimental neutronics studies for CTR blankets (M.C. Scott, B.R. Guy, R. Koohi-Fayegh, J. Perkins and B.Y. Underwood)
  - (1) <u>Neutron yield determination from a (D,T) source</u>

The continuous flow vanadium bath measurements of the neutron yield per alpha particle detected for a  $T(d,n)^4$ He source with associated particle monitoring has been re-analysed and some new measurements made. In particular, the gamma background in the sodium iodide detector monitoring the build up of  $^{56}V$  activity has been studied in some detail to determine (a) the contribution from the  $^{16}O(n,p)^{16}N$  reaction in the vandyl sulphate solution, and (b) the machine induced prompt gamma ray background from neutron capture in the room walls and the sodium iodide shielding. The latter, (b) proved to be significant, whereas it has been negligible with lower energy and radioisotope neutron





sources. The final results are shown in figure 1, where it can be seen that there is now agreement within the experimental errors. In particular, the systematic component of the error, arising from data uncertainties in the vanadium bath analysis, is the same as the discrepancy between the vanadium bath measurement and the calculated neutron yield<sup>(1)</sup>.

(2) Anisotropic yield and energy dependent studies for the lithium fluoride integral assembly

Even with low energy deuterons (ie.  $\sim$  200 keV) there is a significant angular dependence of the emitted neutron energy and a neutron yield anisotropy. Since the use of

a one-dimensional transport code (e.g. the discrete ordinates,  $S_n$ , code ANISN and its derivatives ) is convenient and efficient for general studies, the way in which such a code could be run to represent the actual experimental conditions has been investigated <sup>(2)</sup>. An exact 3-dimensional representation of the LiF experimental assembly and the target characteristics was used with the Monte Carlo code MORSE, the neutron flux being determined as a function of position and angle with respect to the deuteron beam on the target. The source used for the ANISN derived code XSDRNPM (P<sub>7</sub>,S<sub>16</sub>) which was found to give good

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agreement with the MORSE calculation depending on the angle; for measurements along a radius at a particular angle the source spectrum required was that of the target in that direction, i.e. the effect of neutrons emitted at other angles could be neglected. This is, of course, due to the strong forward scattering at these energies. An example of such a comparison is shown in Fig. 2.

The effect of an NE213 scintillator in producing perturbation in our lithium fluoride assembly has also been investigated (3). For a 5 cm x 5 cm cylinder of scintillator it was shown that the difference between the true spectrum and that determined from the scintillator





was < 2% over the energy range from 0.5 to 14 MeV at a radius of 46 cm in our 125 cm diameter sphere of LiF. For the miniature detector used in most of our work (1.4 cm x 1.4 cm cylinder) the perturbation is therefore completely negligible.

## (3) Lithium proportional counter studies

With the possibility in mind of using it for tritium production measurements in integral assemblies, some preliminary studies have been conducted into the construction and characteristics of a lithium proportional counter. Because no suitable gaseous form of lithium existed and because  $(n,\alpha)$  and (n,p) reaction cross sections were high in many of the atoms forming lithium compounds, a natural lithium metal coating was used on the inside of a cylindrical proportional counter shell filled with argon.

The response of a thinly coated counter to thermal neutrons is shown in figure 3, where it can be seen that the triton peak from the  ${}^{6}Li(n,\alpha)T$  reaction is well resolved: in a two parameter display of the same data (with pulse rise time and energy deposited as the parameters concerned) the separation of the tritons and alpha particles is quite distinct in rise time too. Studies of the response to high energy neutrons, which is much more complex because of (i) non tritium producing reactions in  ${}^{6}Li$  and  ${}^{7}Li$ , and (ii) (n, $\alpha$ ) and (n,p) reactions in the walls, are still continuing, so that it is too early to say what the utility of the counter, if any, will be.



- N.L. Evans, I.R. Brearley and Malcolm C. Scott, Nucl. Inst. and Meth. (1979) (in press)
- B.Y. Underwood, Internal Report, Department of Physics, University of Birmingham (1978)
- 3. B.R. Guy, M.Sc. Project Report, University of Birmingham (1978)



9. UNIVERSITY OF EDINBURGH, DEPARTMENT OF PHYSICS

(Head of Department: Professor R.A. Cowley)

9.1 <u>A multiparameter investigation of the ternary fission of <sup>252</sup>Cf (D.E. Cumpstey and D.G. Vass)</u>

A multiparameter study of the ternary fission of  $^{252}$ Cf has been carried out using the xenon gas proportional scintillation detector for fission fragments whose development has been reported in earlier editions<sup>(1)</sup>. Analysis of the profile of a scintillation pulse from this detector provided information about the kinetic energy of the fragment emitted from the front surface of the  $^{252}$ Cf source and also about the orientation of its track in the xenon gas relative to the axis of the detector. The light charged particles accompanying fission were detected, after passing through the Ni backing of the  $^{252}$ Cf source and a layer of xenon gas, in a conventional  $\Delta E \times E$  telescope consisting of two semiconductor radiation detectors mounted on the axis of the system.



Fig. 1 Most probable energy versus angle of emission for <sup>4</sup>He

For each ternary event, the type of light charged particle (e.g. <sup>1</sup>H,<sup>3</sup>H,<sup>4</sup>He or <sup>6</sup>He) was determined, the total kinetic energy of the particle was calculated (with corrections for the energy losses in the Ni foil and xenon gas), the kinetic energy of the fission fragment was recorded and the angle between the direction of the light particle and the light fragment computed. The light and heavy fragment directions are not collinear because of the recoil momentum imparted by the light charged particle and so corrections were made, where necessary, to enable all angles to be referred to the light fragment direction.

Data collected continuously over a period of a month with frequent calibration checks have been analysed to provide the energy spectra of  ${}^{3}$ H and  ${}^{4}$ He particles at 19 angles

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of emission. A Gaussian curve was fitted to each of these spectra and the most probable energy and the width of the distribution determined in each case. The variation of the most probable energy with angle of emission for  ${}^{4}$ He is shown in figure 1, where previous measurements  ${}^{(2,3)}$  are also plotted. Gross energy distributions (over all angles) and gross yields (over all energies) as a function of angle have also been obtained.

- D.E. Cumpstey and D.G. Vass, U.K. Nuclear Data Progress Reports,
  U.K.N.D.C. (78) P88, (1978) 124 and U.K.N.D.C. (76) P86, (1977) 55
- (2) M.J. Fluss, S.B. Kaufmann, E.P. Steinberg, B.D. Wilkins, Phys. Rev. C7 (1973) 353
- (3) K. Tsuji, A. Katase, Y. Yoshida, T. Katayama, F. Toyofuku, H. Yamamoto, Physics and Chemistry of Fission II (Proc. Int. Sym., Rochester N.Y.), IAEA Vienna (1974).
  - 9.2 The Polarization and differential cross-section for the elastic scattering of 16 MeV neutrons (Amena Begum and R.B. Galloway) [Relevant to request numbers: 169-172, 174, 559]

It was reported last year (UKNDC (78) P88,pl21) that measurements of the angular dependence of the polarization of 16.1 MeV neutrons elastically scattered by Fe, Cu, I, W, Hg, and Pb showed a clear difference around 20° from calculations based on 'global' optical model parameters. Further measurements have confirmed this difference. The experimental data is now being used to search for optimum optical model parameters. An account of the first measurements (on Cu and Pb) which showed the '20° discrepancy' has been accepted for publication<sup>(1)</sup>.

(1) R.B. Galloway, and A. Waheed, Phys. Rev. C. (December 1978)

## 9.3 The polarization of 14 MeV neutrons due to elastic scattering(Amena Begum and R.B. Galloway)

In view of the significant discrepancy between the measured and calculated polarization values for the elastic scattering of 16 MeV neutrons through  $20^{\circ}$ , referred to above, measurements on 14 MeV neutrons are also being made. 14 MeV neutrons from the  ${}^{3}\text{H}(d,n)^{4}\text{He}$  reaction are unpolarized and so a double scattering measurement is being carried out, the measured asymmetry in the second scattering being equal to the square of the polarization. Measurements on  $20^{\circ}$  scattering by Cu and by Pb are consistent with the 16 MeV polarization values.

9.4 The polarization and differential cross-section for the elastic scattering of 2.9 MeV neutrons (J. Annand, Amena Begum and R.B. Galloway)

Measurements of the angular dependence of polarization due to elastic scattering and of the elastic differential cross-section for 2.9 MeV neutrons over the angular range  $20^{\circ} - 160^{\circ}$  for samples of Fe, Cu, I, Hg, and Pb have been compared with the results of combining optical model and Hauser-Feshbach calculations. The optical model calculations were performed using 'global fit' parameters as well as with parameters suggested previously for the particular nuclei. The Hauser-Feshbach calculations were performed both with and without the level width fluctuation correction <sup>(1)</sup>. It is clear that the calculations made without the level width fluctuation correction provide a better fit to the data for Fe, Cu, I and Hg and only for Pb does inclusion of the level width fluctuation provide a better fit. These optical model parameter sets are not very successful in fitting both differential cross-section and polarization data. Searches have been made for optmum parameters. This work has been prepared for publication in Nuclear Physics.

To extend this investigation measurements have also been made on W, T1, Pb, Bi and U and these are now subject to analysis.

An improved neutron polarimeter has been completed capable of making measurements at 11 scattering angles simultaneously and it should come into routine use shortly.

(1) P.A. Moldauer, Phys. Rev. 135B (1964) 642

9.5 <u>Neutron polarization in the <sup>7</sup>Li(d,n)</u> <sup>8</sup>Be reaction (R.B. Galloway, and A.M. Ghazarian) The <sup>7</sup>Li(d,n) <sup>8</sup>Be reaction is being studied with 450 keV incident deuterons as a possible useful source of polarized 14 MeV neutrons. Polarization determination is by elastic scattering by <sup>4</sup>He as used in other recent studies <sup>(1,2)</sup>. Neutron polarization values of about 0.2 to 0.3 have been found and the angular dependence is being determined.

- (1) A.M. Alsoraya and R.B. Galloway, Nucl. Phys. A280 (1977) 61
- (2) B.S. Bains and R.B. Galloway, Nucl. Instr. and Meth. <u>143</u> (1977) 295

## 9.6 The polarization of neutrons from the ${}^{2}H(d,n){}^{3}He$ reaction and neutron elastic scattering by Pb (R.B. Galloway and R.M. Lugo)

A small angle Mott-Schwinger scattering fast neutron polarimeter has been used to determine the polarization of the neutrons from the  ${}^{2}$ H(d,n) ${}^{3}$ He reaction for deuteron energies from 100 to 500 keV. The results are in excellent agreement with recent n- ${}^{4}$ He

scattering measurements. Incidental to these measurements, the total cross-section and the differential elastic cross-section for scattering by Pb through  $3.65^{\circ} \pm 1.85^{\circ}$  have been determined for neutrons from 2.2 to 3.14 MeV. An account of this work has appeared in Nuclear Instruments and Methods, <u>158</u> (1979) 153.

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