IAEA-186

TRANSACTINIUM ISOTOPE NUCLEAR DATA (TND)

VOL.I

PROCEEDINGS OF AN ADVISORY GROUP MEETING
ON TRANSACTINIUM ISOTOPE NUCLEAR DATA
ORGANIZED BY THE
IAEA NUCLEAR DATA SECTION
IN CO-OPERATION WITH THE
OECD NUCLEAR ENERGY AGENCY
HELD AT THE KERNFORSCHUNGSZENTRUM
KARLSRUHE, 3–7 NOVEMBER 1975

A TECHNICAL DOCUMENT ISSUED BY THE
INTERNATIONAL ATOMIC ENERGY AGENCY, VIENNA, 1976

The IAEA does not maintain stocks of reports in this series. However, microfiche copies of these reports can be obtained from

INIS Microfiche Clearinghouse
International Atomic Energy Agency
Kärntner Ring 11
P.O. Box 590
A-1011 Vienna, Austria

on prepayment of US $0.65 or against one IAEA microfiche service coupon.

IAEA-186

TRANSACTINIUM ISOTOPE NUCLEAR DATA (TND)

VOL.I

PROCEEDINGS OF AN ADVISORY GROUP MEETING
ON TRANSACTINIUM ISOTOPE NUCLEAR DATA
ORGANIZED BY THE
IAEA NUCLEAR DATA SECTION
IN CO-OPERATION WITH THE
OECD NUCLEAR ENERGY AGENCY
HELD AT THE KERNFORSCHUNGSZENTRUM
KARLSRUHE, 3–7 NOVEMBER 1975

A TECHNICAL DOCUMENT ISSUED BY THE
INTERNATIONAL ATOMIC ENERGY AGENCY, VIENNA, 1976

FOREWORD

The IAEA Nuclear Data Section, in cooperation with the OECD Nuclear Energy Agency, convened an Advisory Group Meeting on Transactinium Isotope Nuclear Data at Karlsruhe, FRG, from 3-7 November 1975. The meeting was attended by 45 representatives from 13 countries and 3 international organizations. It was the first international meeting on this topic.

The general conclusion of the meeting participants was that transactinium isotopes are becoming more and more important in nuclear technology, and that the present knowledge of nuclear data required to evaluate the effects of actinides in nuclear technology is not satisfactory. One of the basic recommendations, which resulted from the meeting was to initiate an internationally coordinated programme to measure, calculate, and evaluate needed transactinium isotope nuclear data which would span the next ten years. The principal aim of this effort would be to improve the status of actinide nuclear data required for nuclear technology.

Table of Contents

VOLUME I

Summary Report

Session A, Survey of TND Applications

Table of Contents

VOLUME II

VOLUME III

SUMMARY REPORT

I. INTRODUCTION

In addition to the main fertile and fissile isotopes (i.e. Th 232, U 233, U 235, U 238 and Pu 239), transactinium isotopes, comprising all isotopes heavier than actinium (i.e. $Z \geq 89$), play important roles in the nuclear fuel cycles of both thermal and fast reactors, and have found increasing areas of applications in life sciences and industry. The quantitative appraisal of the role and applications of these isotopes can only be done with an adequate knowledge of their nuclear characteristics, that is their nuclear data. Exact values of thermal and resonance neutron cross sections, fast neutron cross sections, and nuclear decay parameters of transactinium isotopes are required to calculate their effect in thermal and fast power reactors and to assess their impact on the nuclear waste management.

The Nuclear Data Section, in cooperation with the OECD Nuclear Energy Agency, convened an Advisory Group Meeting on Transactinium Isotope Nuclear Data whose principal purpose was to bring together users and producers of transactinium isotope nuclear data in order to comprehensively survey the requirements and priorities, and review the status of knowledge of transactinium isotope nuclear data with the specific aim to formulate recommendations and measures for the coordination of future work.

In order to meet these objectives, the meeting was organized around seventeen comprehensive review papers (see page 29), covering the full scope of applications in one session, and an extensive review of the status of transactinium nuclear data in a second session. These review papers, which in themselves are timely reports on important topics, provided the basis for the discussions during the first three days of the meeting and for the preparation of the conclusions and recommendations during the last two days of the meeting (see Meeting Programme page 31).

Forty five scientists, representing both the nuclear data user and producer communities, from thirteen countries and 3 international organizations, took part in the meeting (see page 33).

The transactinium isotope nuclear data which were discussed at the meeting fall into three general areas of applications

- thermal reactors and their fuel cycle,
- fast reactors and their fuel cycle, and
- nuclear waste management and isotope applications.

In order to arrive at comprehensive sets of conclusions and recommendations, and meaningful summary statements regarding the requirement and status of transactinium isotope nuclear data for each of

these three categories, the participants were assigned to three separate working groups during the last two days of the meeting. The working groups, formed according to the three data categories listed above, prepared written summary statements which were discussed during the plenary session on the last day of the meeting.

The meeting produced one set of general recommendations, and three reports on each of the three general areas of applications, each with its own conclusions and recommendations, and comparisons of trans-actinium isotope nuclear data requirements and status.

II. GENERAL RECOMMENDATIONS

Preamble

The participants of the IAEA Advisory Group Meeting on Transactinium Isotope Nuclear Data noted

a. that transactinium isotopes are becoming more and more important in nuclear technology; and

b. that the present knowledge of nuclear data required to evaluate the effects of transactinium isotopes in nuclear technology is not satisfactory.

General Recommendation 1

1.1. In view of the above, the meeting recommends that an internationally coordinated effort be implemented, and pursued during the next ten years so as to improve the status of transactinium neutron nuclear data required for nuclear technology. The results from the first phase of this effort should be made available and reviewed after the first three to five years of this effort.

1.2. This international effort should comprise the following three components:

   a. a coordinated programme of differential and integral neutron nuclear data measurements with a free exchange of information;

   b. a coordinated effort in nuclear theory computations designed to supplement the experimental information, and

   c. a coordinated programme for the evaluation of transactinium isotope neutron nuclear data.

1.3. It is understood that the IAEA Consultants' Meeting on the use of Nuclear theory in Neutron Nuclear Data Evaluation to be held at ICTP Trieste, in December 1975, as well as future meetings on this topic will deal with nuclear theory calculations of neutron nuclear data of transactinium isotopes. More detailed aspects of the suggested measurement programme are discussed under specific conclusions and recommendations, given below.

1.4. The evaluation programme should initially be concerned with two or more independent evaluations by different laboratories for mutually agreed important nuclides where differential measurements exist, supplemented by nuclear theory calculations.

1.5. The IAEA should co-ordinate the evaluation programme by:

a. convening meetings of the evaluation groups concerned in order to coordinate methods and techniques at regular intervals, and

b. sponsoring comprehensive analyses and comparisons of evaluations for the benefit of all users of transactinium isotope nuclear data.

1.6. The IAEA should reconvene another specialists' meeting on transactinium nuclear data at such a time when the evaluations, accompanying sensitivity studies, and measurements, have been completed so that the users of these nuclear data have the opportunity to assess the extent to which existing and future requirements for transactinium isotope nuclear data have been met.

General Recommendation 2

The meeting recommends that an Actinide Newsletter be published consisting of brief descriptions of measurements, compilation activities, and computer calculations carried out world-wide pertaining to the applications discussed at this meeting. It requests Dr. S. Raman of the Oak Ridge National Laboratory to coordinate all aspects concerning the preparation, publication, and distribution of such a newsletter.

General Recommendation 3

3.1. The meeting recommends that analytical results on irradiated fuel, together with the necessary reactor information (cell version) and initial fuel composition, be made available to the International Atomic Energy Agency for use by member states in order to check the reliability of calculation codes and nuclear data input.

Integral experiments have been performed in a number of reactors and different fuel types. The results of some of these experiments have not yet been distributed to enable any comparison to be carried out. Other fuel material has not yet been analysed. It would be desirable to complete existing experiments and their results be made available to the IAEA.

3.2. For this purpose, data resulting from experiments with (a) high burn-up fuel material from a Light Water Reactor (LWR) operating on the uranium cycle, (b) LWR irradiated fuel containing recycled plutonium, (c) fuel from a High Temperature Reactor, and (d) fuel from a Fast Reactor, could be used as standards. The NEACRP is willing to support this activity.

3.3. From the experience obtained so far, from the two- and three dimensional IAEA reference cases on thermal reactors proposed at the IAEA Reactor Physics Burn-up Panel in 1973, it would be useful to carry out a simpler but nevertheless representative analysis of the growth and decay of actinide nuclides for typical reference cases (LWR, HTR, LMFBR). The NEACRP is willing to support this exercise.

General Recommendation 4

The meeting recommends to develop an international cooperative research project to be coordinated by the IAEA on the measurement and evaluation of needed decay data of transactinium nuclides. The presently available accuracies in half-lives, alpha intensities and gamma-ray intensities of a number of transactinium nuclides need to be imporved to fulfil the needs for fuel analysis, safeguards applications, mass determination and the preparation of standards. New measurements, evaluations of the existing data, intercomparisons between the laboratories concerned, and exchanges of samples and techniques are recommended in order to arrive at accurate and worldwide consistent sets of these data.

General Recommendation 5

The neutron dose rates generated by (α,n) reactions in different kinds of glasses constitute a hazard during waste handling. It is therefore recommended that cross sections pertinent to elements used in nuclear waste vitrification be made available.

General Comment

1. Integral cross section measurements obtained by post-irradiation analysis of spent fuel or test irradiations can be used to check differential cross section data, provided all information (on flux, energy spectrum, reactor operation condition etc.) can be made available. Standard reference experiments are useful for this purpose.

2. Should differential cross section data not be sufficiently accurate or not available, measurement of integral cross sections can be performed easily; this information, however, is pertinent only to a specific reactor and its mode of operation.

III. CONCLUSIONS AND RECOMMENDATIONS OF WORKING GROUPS

A. Report of the Working Group on the Requirements of TND for Thermal Reactors (Chairman: R.M. Nunn)

I. Introduction

This report is concerned with the requirements, status and availability of TND in the context of thermal reactors. It is in order to make the following introductory remarks:

(i) During the discussion of the working group it became apparent that there were requirements for TND which had not been identified in the review papers, and, more importantly, that the status of some of the data required had not been included in the reviews.

(ii) Some of the reviewers had not made specific recommendations as to requirements for data or other required actions, and this increased the difficulties of the working group in reaching satisfactory conclusions and recommendations during the time available for discussion.

(iii) So far as nuclear power plants are concerned, the importance of some of the actinides has emerged only over the last few years, and the requirements for data are in many instances concerned with problems which have only recently been identified. In many cases, the development is inadequate to fully define the accuracy requirements for TND.

II. Requirements and Status of TND

The requirements for TND stem from the recognition of the importance of actinides in a wide range of reactor problems, but the actinides themselves can be grouped into two categories:

(i) Those which may be present in only very small quantities (e.g. U 232 and its daughters, or Cm 242), but which, because of their unique properties, assume considerable importance.

(ii) Those which are present in comparatively large quantities (e.g. the plutonium and americium isotopes), and because of either their decay or neutronic properties are important.

1. Core design and fuel analysis for code evaluation

a. Core design

The requirements for transactinium nuclear data for the design of thermal reactors (RP.A2) are connected with the need to derive several reactor parameters, namely: reactivity, power distribution, burn-up, kinetic response, temperature coefficients and others. In these calculations the data of prime importance are the fission and capture cross sections and it is possible to divide the transactinium nuclei into two groups.

(i) Nuclei of importance in the major (Th-U, U-Pu) fuel cycles: U 234, U 236, Pu 240, Pu 241 and Pu 242.

The requirements and data available for these nuclei have been subjected to extensive study in the past. Nevertheless, in some instances there are long outstanding and detailed requirements for data on these nuclides which have not been met. In specific cases the requests are satisfied by a combination of the available data and the results of fuel analyses.

(ii) The isotopes listed in Table 1, column 1.

Cross section requirements for these nuclei are related to reactivity calculations and are presented in Table 1. The requested accuracies quoted in Table 1 for Pa and U isotopes are based on considerations of the Th-U fuel cycle. For Am and Cm isotopes the accuracies are based on calculations for Pu recycling in thermal reactors.

The calculation of the internal neutron source strength, required for subcritical reactivity measurements during refuelling of reactors, demands a knowledge of the neutron output due to Cm 242 and to a lesser extent, Cm 244. The group is of the opinion that an accuracy of $\pm$ 50 % in the calculation of the source strength should be sufficient and, consequently, the requirements for the related data are less stringent than those derived from consideration of core physics.

b. Fuel analysis

Destructive and non-destructive analyses of irradiated fuel lead to a heavy isotopic assay which can validate the

computer codes and the data used to predict fuel burn-up. Judicious use of these techniques on selected fuel elements may differentiate deficiencies in computer codes from defects in the input data. The nuclear data requirements for these analyses are mainly the half-lives and decay schemes of the isotopes listed in Table 3. Except for the Am and Cm isotopes the data requirements are less stringent than those required for quality control (sect. II.2.a.). For the Am and Cm isotopes the half-life data appear to be adequate (see Table 3).

2. Fresh-fuel fabrication, transport and handling

The problems (and hence requirements) depend upon the type of fuel employed, and fall into five broad categories:

(i) Quality control at the fabrication plant and enrichment checks prior to loading into the reactor - i.e. destructive and non-destructive assay methods.

(ii) Health physics problems associated with fabricating, storing, handling and transportation of the fuel, particularly for fuel with energetic γ and neutron emission.

(iii) Criticality control (neutronics).

(iv) Safeguards (destructive and non-destructive assay).

(v) Fuel design.

a. Quality control and enrichment checks

The fuel fabricator will operate to tight product specification and for this purpose employs both destructive and non-destructive methods of fuel analysis. No TND cross section data are required, but the analyses require accurate data on half-lives, decay schemes and energies. The group accepted the view of Dierckx (RP.A8) on data requirements, and a comparison of the requirements and status is given in Table 3. It would appear that the half-life data for Pu 238 are inadequate. For two nuclides, Pu 239 and Pu 241, the panel noted discrepancies which suggested that the data may also be inadequate (see footnote to Table 3). In the opinion of Dierckx (RP.A8) the decay schemes are adequately known. This is confirmed by the classification of Reich (RP.B7).

b. Prediction of the isotopic content of fresh fuels

There are no requirements for fresh uranium fuel. Recycle uranium fuels may be contaminated with U 232 to an extent which

affects the design and operation of fuel fabrication plants, and of fuel handling procedures. Recycle plutonium fuels present wellknown fabrication problems, and for these, and for the design of fuel handling arrangements, TND are required to predict the likely composition of these fuels - particularly for the buildup of Pu 238 and Pu 240. Recycle Th/U 233 fuel will be heavily contaminated with U 232 and this will require the provision of remote fabrication and handling facilities.

The requirements for TND are listed in Table 1. Except for the (n,2n) cross sections, the available cross section data would seem to be adequate. The information on (n,2n) cross sections and companion (γ,n) cross sections was insufficient to judge the status of the data. From the number of measurements carried out and the evaluations available, it seems likely that the status will meet the requirements.

c. Criticality control

TND required are adequately covered by the requirements for core design (Section II.1.a. this report, also report of Fast Reactor Working group).

d. Safeguards requirements

Safeguards requirements were not discussed in detail by the group but should be covered by the requirements for fuel analysis discussed previously (also see RP.A8).

e. Fuel design

Actinides influence fuel design, from consideration of their in-reactor performance (covered by II.1. above) and through consideration of their impact on fuel fabrication problems and on subsequent in-reactor chemical performance. For the latter, only very approximate TND are required (total Pu content and total fission fraction in plutonium) and the requirements from fuel fabrication are covered by II.2.b. above.

f. Reactor shutdown heating

The contribution of actinides to total decay heat is small, and at short shutdown times the dominant actinide is Np 239. No cross-section data for this isotope is available: the accuracy requirement is for σ(n,abs) to an order of magnitude.

g. Discharge fuel handling, storage and transport

For fuel handling, the major requirement is for accurate (i.e. $\sim \pm$ 10 %) predictions of total heat production. The dominant heat source is fission product decay, and the important actinides are Cm 242 and Cm 244. The accuracy requirements are stated in Table 1 and compared in Table 2. It would appear that the presently available data are adequate. For pond storage of discharged fuel, actinides do not give rise to significant problems. For dry storage, the accuracy requirements will be similar to those required for irradiated fuel handling.

For fuel transportation, heat production and neutron emission (especially) are both of importance. The heat requirements are similar to those required for fuel handling and the prediction of neutron output is required with $\sim$ 20 % accuracy. The requirements would appear to be met by available data (see Table 2).

III. Future Requirements

This report has considered requirements presently identified by those associated with nuclear power plants. Because of the very recent recognition of the importance of some actinides, it is difficult to predict future requirements. However, now that actinide build-up codes are freely available it is necessary that data is available for calculating the build-up of all actinides to obtain estimates of inventories of the longer lived (> 20 min) isotopes to an accuracy of at least a factor of 10. As a very approximate guide it is suggested that the cross-sections involved in the predictions should be known to an accuracy of 30-50 %.

IV. Summary and Conclusions

1. As a basis for discussing the status of neutron cross-section data, the group used the review paper by Benjamin (RP.B1) supplemented by the paper of James (RP.B2).

2. In the main, available cross-section data appear to meet present requirements. There are exceptions and these are marked * in Table 2. The more important of these are:

   a. The long-standing requirement for Pu 240 resonance data, especially for low energy (< 20ev) resonances does not appear to have been met. These data are of particular importance to reactor design.

b. Very limited data exist for Np 239 and Pu 236 (reviewed in RP.B1), a 100 % accuracy is required. Data for Am 242m and Cm 243 were reviewed in RP.B1, and a 50 % evaluation of these data has recently been completed (private communication from R.W. Benjamin, 19 February 1976).

3. The half-life data for Pu 238, Pu 239 and Pu 241 do not meet the requirements for fuel quality control and may not meet the requirements for burn-up analyses.

4. Insufficient data were presented to properly assess the status of the (γ,n) and $(n,2n)$ cross-sections required to determine the buildup of important nuclides (U 232, Pu 236 and Pu 238), or for the neutron yield from (α,n) reactions.

5. Integral measurements are a powerful means of checking both data and codes and are of great importance in establishing overall prediction uncertainties for power reactors. These measurements are encouraged.

6. For data beyond Cm 244 in Table 2, no requests have been identified. However, data adequate to predict the buildup of these isotopes to an accuracy of at least a factor of 10 should be made available. Sensitivity studies are needed to identify whether this is achievable with existing data.

V. Recommendations

1. Accurate resonance parameters for Pu 240, particularly below 20 ev, should be obtained as soon as possible. The 1 ev resonance is of prime importance.

2. The status of cross section data for Np 239 and Pu 236 should be carefully assessed and the need for measurements ascertained. The status of Am 242m and Cm 243 was assessed in RP.B1.

3. For the remaining data marked with an asterisk in Table 2, evaluations should be carried out to determine whether existing data meet the requirements.

4. An assessment should be carried out to ensure that evaluations of $(n,2n)$ cross sections and of (α,n) and (γ,n) yields have led to an estimated error which satisfies the requirements shown in Table 1.

5. Evaluations should always include realistic estimates of errors.

Table 1

Cross Section Accuracy Requirements for Specific Thermal Reactor Fuel Cycle Applications

(accuracies given in percent)

Reaction and Isotope	Core Design	Fresh Fuel	Decay Heat	Discharge Fuel	Transport
Th 232 (n,2n)		20			
Pa 231 (n,γ)	10				
Pa 233 (n,γ)	10				
U 232 (n,γ)	30				
233 (n,2n)		30			
234 (n,γ)	5				
236 (n,γ)	4				
238 (n,2n)		50			
Np 237 (n,γ)	10	50			
239 (n,2n)		50			
239 (n,Abs)	100		100		
Pu 236 (n,Abs)		50			
238 (n,γ)	30				
239 (n,2n)					
*240 (n,γ)	1				
*241 (n,γ)	3				
(n,f)	1				
($\bar{\upsilon}$)	0.5				
*242 (n,γ)	5		50	30	20
Am 241**(n,γ)	10			30	20
Am 241***(n,γ)	50				
m242 (n,γ)	50				
m242 (n,f)	30				
243 (n,γ)	10				
Cm 242 (n,γ)	50				
244 (n,γ)	50				

* But note detailed requests are outstanding.
** for production of ground state Am 242.
*** for production of isomeric state.

For α emitters (α,n) yields (in UO_2) are required. For Cm 242 and Cm 244 to 100%, likewise for other isotopes.

(γ,n) production rates are required for Th 232, U 233, U 238, Np 239, Pu 239 to ~100% accuracy.

Table 2

Comparison of Requirements and Status of Cross Section Data for Thermal Reactor Applications

Isotope	$\sigma_{n\gamma}$			σ_{nf}			$I_{n\gamma}$			I_{nf}		
	Requested accuracy (1)	2200m Data accuracy	Request not satisfied (= *)	Requested accuracy (1)	2200m Data accuracy	Request not satisfied (= *)	Requested accuracy (2)	Data accuracy	Request not satisfied (= *)	Requested accuracy (2)	Data accuracy	Request not satisfied (= *)
Pa 231	10	10					20	7				
Pa 233	10	12	*				10	4				
U 232	30	2		30	12		30	6		30	13	
U 234	5	1.5					5	12	*			
U 236	4	6	*				4	6	*			
U 237	100	33					100	18	no cut off given			
Np 237	100	2					10	8				
Np 239	100	?	*									
Pu 236	100	-	*				100	-	*			
Pu 238	30	4		50	3		50	10				
Pu 240	2	0.5					1	12	*			
Pu 241	3	3		1	<1 %		10	5				
Pu 242	10	4-5					5	4				
Am 241(GS)	10	3					10	9				
Am 241(IS)	50	8					50	9				
Am 242m	50	-	*	30	4		50	-	*	30	7	
Am 243	15	5					10	3	*			
Cm 242	50	50					50	30				
Cm 243	50	-	*	50	7		50	-	*	50	21	
Cm 244	50	20					50	10				
Cm 245	50	10		50	25		100	7		100	5	
Cm 246	100	20		30	15		30	7		30	7	
Cm 247	30	15		30	15		30	15		30	7	
Cm 248	50	10					30	10				
Bk 249	30	50					30	50				
Cf 249	50	6		30	3		70	10		60	10	
Cf 250	50	15		30	-		70	7		50	-	
Cf 252	50	10		30	10		70	4		50	15	
Cf 252	30	8		30	10		50	8		50	20	
Cf 253	100	20		30	20		200	25		60	25	
Es 253(GS)	30	-					30	5				

Footnotes:

(1) The requested accuracy is for an average value over a reactor spectrum. The measured accuracy quoted is for 2200m values. These will meet the requests in the absence of serious non-$1/_v$ cross-section behaviour. All accuracies are given in %.

(2) Resonance integral (I) has been used as a means of parametrising the non-thermal requirements. In general, differential cross sections for major resonances will be required.

Comment:

Data requirements for isotopes beyond Cm 244 are associated with future requirements. See section 3. They are purely speculative, and a sensitivity analysis is required.

Table 3

Comparison of Requirements and Status of Half-life and Decay Data for Thermal Reactor Applications

Isotope	Half-life data *		Decay Scheme Status (see RP.B7, Table I)	Needed for
	Required Accuracy (%)	Achieved Accuracy (%)		
Pu 238	0.5	1	A	Quality Control
Pu 239	0.5	1	A	
Pu 240	1	1	A	
Pu 241	1	3	A(β^-),B(α)	
Am 241	4	1	A	Burnup Analysis
Cm 242	2	1	A	
Cm 244	2	1	B	

* The achieved accuracies quoted above are statistical confidence levels of 1σ and do not reflect the range of currently available data. In some instances working group 3 has assessed larger uncertainties taking this latter point into account.

B. Report of the Working Group on the Requirements of TND for Fast Reactors (Chairman: J.Y. Barré)

I. Introduction

The requests to be considered are for present-day fast reactors.

Problems considered are of three categories:

(i) in-core problems for reactivity and breeding gain

(ii) out-of-core problems for handling, reprocessing, manufacturing and shielding

(iii) fuel analysis for code tests and operation of the plants.

Only the U-Pu fuel cycle for fast reactors is considered in this document.

Any use of Th 232, including the U-Th cycle will be considered at the appropriate time.

It is not the intention here to determine requests for the higher Pu isotopes Pu 240, Pu 241, Pu 242. Requests for these isotopes have been made in many previous documents. The most challenging of these requests are for the capture cross-section of Pu 240 to about 5% and the fission cross-section of Pu 241 to about 1.5%. The capture cross-sections for Pu 241 and Pu 242 are normally requested to about 8%. The fission cross-sections of Pu 240 and Pu 242 are usually requested to about 2% and 4%, respectively.

In the long term, actinides may be recycled in a fast reactor to avoid the need to store them for thousands of years. Perhaps the most extreme version of this idea is for the fast reactor to be fuelled by the higher actinides. In this case, the accuracy requirements for these isotopes would be similar to those presently required for the plutonium isotopes. Fuel handling problems would be considerably increased due to the activity involved, and the data for decay heat and neutron emission would need to be known to high accuracy. Until more complete sensitivity studies are carried out, it is not possible to determine quantitative requests for cross-sections for this type of actinide recycle.

II. Requests

The table below lists the accuracy requirements for the isotopes which are considered to be of major importance for fast reactors.

Accuracy Requirements (in percent)

	Cross-Sections			$\bar{\upsilon}$	Half-life
	(n,γ)	fission	$(n,2n)$		
Np 237	30	50	50	50	
Np 239	20	50		50	
U 238			50		
Pu 236	50	50			
Pu 238	20	7		4	0.5
Pu 239			50		
Am 241	5	15		10	1
Am 242m	50	15		10	
Am 243	10	30		25	
Cm 242	50	25		15	3
Cm 244	50	50			3

Comments:

a. (n, gamma) cross-sections:

The most stringent requirements on accuracy for these cross-sections are fixed by the in-core reactivity estimate. The exceptions are the two curium isotopes, which limits are set by the fuel handling considerations, and Pu 236 (see comment f).

b. fission cross-sections:

Most of the required accuracies are again fixed by the reactivity effect. The curium isotopes accuracy is determined by the fuel handling.

c. Nu values:

These are fixed by the reactivity effects.

d. Alpha-decay half-lives:

Since they are considered to be sufficiently well known for most fast reactor purposes, half-lives of the isotopes for alpha-particle decay are listed only for some isotopes. An exception to this statement is for fuel analysis where the required accuracies are 0.5 % for Pu 239, and 1 % for Pu 240 and Pu 241.

e. Half-lives for spontaneous fission are required for the isotopes Cm 242 and Cm 244 to within 15 %. This accuracy is needed to estimate the rate of neutron production for fuel handling assessments.

f. Plutonium 236 and neptunium 237 are included in the table because they both lead to the production of uranium 232. The gamma-activity of the daughter of this latter isotope causes problems in the subsequent handling and refabrication of the uranium.

g. To predict nuclide concentration of actinides one has to be careful in choosing coarse energy group structure in the whole burnup calculation (see RP.A3).

h. For the capture cross-sections the energy range (0.5-100 keV) and for fission 1 keV - 2 MeV have to be taken into account (except for threshold fission).

i. After fuel unloading, the cooling and storage time play important roles with respect to the actinide concentration (especially for Am 241). This can influence the requested accuracies according to the fuel cycle chosen.

j. The information on (γ,n) cross sections is presently insufficient to be able to analyse the importance of this problem compared to $(n,2n)$ reactions: example Np 237 for Pu 236 production. Orders of magnitude for these cross-sections would be useful. The same problem exists for the (α,n) cross-section on oxygen (neutron dose).

III. Summary of Data Status for Transactinium Nuclear Data for fast Reactors

1. Theoretical

Fission probabilities from reactions such as (d,pf) and (t,pf) have been inferred for all the actinide isotopes of current fast reactor interest. These and the measured fission cross-sections, when available, can be used in theoretical calculations of σ_{nf} and $\sigma_{n,\gamma}$ in the energy range of interest to fast reactors. We estimate that the reliability of such calculations is $\pm$ 25 % for the isotopes of U, Np, Pu and Am, $\pm$ 30 % for the medium isotopes of Cm (say Cm 244, Cm 245) and deteriorating to $\pm$ 50 % for other isotopes of Cm, Bk and Cf. For Th and Pa, $\sigma_{n,\gamma}$ can be predicted by present techniques to $\pm$ 25 %, but the fission cross-sections appear to be overpredicted by perhaps a factor of 2, although modifications to the theory have been suggested which could improve this situation.

2. Experimental

Np 237: fission cross-sections above threshold agree to less than 10 % but capture data show $\simeq$ 50 % discrepancy. No plans are known for measurement.

Pu 238: estimated accuracy of σ_{nf} is $\pm$ 10 % above threshold; $\sigma_{n,\gamma}$ below 300 keV is probably good to $\pm$ 30 %, on the average over the range of interest.

Pu 240: capture cross-section data to less than 10 % below 100 keV to $\pm$ 20 % at 300 keV. Fission data accuracy is estimated at $\pm$ 10 % above threshold; recent results by Behrens at LLL should provide corroboration.

Pu 241: alpha measurements currently in progress at ORELA are expected to yield data comparable to Pu 240; Behrens' fission ratios on Pu 241 should provide highly accurate results ($\simeq$ 1-2 %).

Pu 242: capture measurements at RPI with accuracy $\leqslant$ 10 % available below 100 keV. Fission measurements above threshold also agree to $\simeq$ 10 %.

Am 241: absorption cross-section measurements at ORNL have an accuracy of 5-10 %; however the uncertainty in the fission cross-section shape and discrepancies with integral measurements suggest that the capture cross-section is probably uncertain to 30 %. A remeasurement of fission to verify the gross structure reported by Seeger et al, between 1 and 100 keV, is urgently needed.

Am 242: we estimate that uncertainties as large as ± 50 % exist in current fission data. Measurements with a better sample are planned at LLL. No radiative capture measurements exist and there are no plans for making this extremely difficult measurement.

Am 243: the fission cross-section is probably known to ± 10 % above threshold, but no radiative capture measurements have been made. These are urgently needed; the accuracy obtainable with current theoretical techniques is probably not sufficient to satisfy fast reactor needs.

Cm 242: no experimental data exist in the region of fast reactor interest.

IV. Specific Recommendations for Provisional Data

Comparison of these remarks on data status with the requirements in Table 4 reveals the following immediate data gaps:

1. Am 241. Fission cross-section needs verification in the region 1-100 keV (highly dissimilar data exist here). A measurement of the capture cross-section in this region and above is also highly desirable, particularly since there are now apparent discrepancies between experimental and theoretical differential data on the one hand, and integral data, on the other.

2. Am 242m. The capture cross-section, or alternatively very accurate measurements of both fission and absorption, are required.

3. Am 243. Capture cross-section is required.

4. Pu 240. Better capture data are required.

5. Pu 241. Perhaps new ORELA measurements now being made will fill gap here.

6. Pu 242. Capture data are required.

7. Cm 242. Capture and fission data are required.

8. Cm 244. Capture data are required.

9. Np 237. Capture data are required.

It should be stressed that while some of the above gaps can be filled by theory to within the presently needed accuracy, this does not absolve the data measuring community from measuring data on the higher transuranium isotopes, not least for the purpose of checking the theory.

V. General Remarks

The comparison of TND requests and status is summed up in Table 4.

1. For fast reactor core physics, the accuracies needed are well defined and probably not reached for all the nuclei involved.

2. For fast reactor fuel cycle,
   - it is probably necessary to have more precise knowledge of the different fuel cycles and strategies envisaged,
   - the mode of operation of fast reactors may play a role in the accuracies needed, and
   - the improvement of plutonium recovery in reprocessing plants may affect the desired accuracies.

3. For medium and long-term perspectives,
   - the accuracies actually quoted could change by some factor when the number of operating reactors will increase, and
   - if one is to envisage special fast reactors for actinide recycling, better accuracies for some of the transactinium isotopes will be needed.

General Recommendations:

1. The transactinium evaluated neutron nuclear data should be improved over the next five years in the framework of an international collaboration:

   for microscopic data:

   a. One should improve the availability of transactinium samples for laboratories already qualified to perform the measurements or for laboratories which are developing measurement techniques.

   b. Evaluations should be exchanged and compared. They must include error estimates on evaluated data.

   for integral data:

   Integral experimental results should be exchanged and compared with calculations. It seems necessary to confirm the transactinium neutron nuclear data coming from differential measurements and evaluations by integral results on irradiated fuels and critical experiments.

For data already available discrepancies should be examined and resolved.

2. Fuel cycle strategies should be better defined.

3. There should be an appropriate international diffusion of experimental, calculated and evaluated transactinium nuclear data in a form suitable for the users.

Table 4

Comparison of Requirements and Status of Nuclear Data for Fast Reactor Applications

Nuclide	Data Type	Main Energy Range	Accuracy %		Comment	Origin	Needs
			Achieved	Required			
Np 237	(n,γ)	.5 - 100 KeV	$\simeq$ 50	30	$\pm$ 25 % from theory	A2, A4, A5	In-core cycle, fuel handling, and fabrication
	(n,f)	above threshold	< 10	50	$\pm$ 25 % from theory		
	$(n,2n)$	above threshold	?	50			
	$\bar{\upsilon}$	above threshold	?	50			
Np 239	(n,γ)	.5 - 100 KeV	?	20	$\pm$ 25 % from theory	A2	In-core cycle and decay heat
	(n,f)	above threshold	?	50	$\pm$ 25 % from theory	A4,A5	
	$\bar{\upsilon}$	above threshold	?	50			
U 238	$(n,2n)$	above threshold	?	20		A4	Fuel fabrication
Pu 236	(n,γ)	.5 - 100 Kev	?	50	$\pm$ 25 % from theory	A4	Fuel fabrication
	(n,f)	1 KeV - 2 MeV	?	50	$\pm$ 25 % from theory	A5	and Pu recycling
Pu 238	(n,γ)	.5 - 100 KeV	30	20	$\pm$ 25 % from theory	A2	In-core cycle, and fuel fabrication, and fuel control
	(n,f)	1 KeV - 5 MeV	<10	7	$\pm$ 25 % from theory	A5	
	$\bar{\upsilon}$	1 KeV - 5 MeV	?	4			
	$T_{1/2}$ (α)		1	0,5		A8	
Pu 239	$(n,2n)$	above threshold	?	50		A5	Fuel fabrication
Am 241	(n,γ)	.5 - 100 KeV	30	5	$\pm$ 25 % from theory	A2, A5	In-core cycle, fuel fabrication, fuel control, and Cm 242 production.
	(n,f)	1 KeV - 5 MeV	30 ?	15	$\pm$ 25 % from theory		
	$\bar{\upsilon}$	1 KeV - 5 MeV	?	10			
	$T_{1/2}$ (α)		1	1		A8	
Am 242m	(n,γ)	.5 - 100 KeV	?	50		A2	In-core cycle
	(n,f)	1 KeV - 2 MeV	50	15			
	$\bar{\upsilon}$	1 KeV - 2 MeV	?	10			
Am 243	(n,γ)	.5 - 100 KeV	?	10	$\pm$ 25 % from theory	A2	In-core cycle and Cm 244 production
	(n,f)	above threshold	10	30	$\pm$ 25 % from theory	A5	
	$\bar{\upsilon}$	above threshold	?	25			
Cm 242	(n,γ)	.5 - 100 KeV	?	50	$\pm$ 30 % from theory	A4, A5	Fuel handling, storage, transport, and control.
	(n,f)	1 KeV - 2 MeV	?	25	$\pm$ 30 % from theory		
	$\bar{\upsilon}$	1 KeV - 2 MeV	?	15			
	$T_{1/2}$ (s.f.)		1	3		A8	
Cm 244	(n,γ)	.5 - 100 KeV	?	50	$\pm$ 50 % from theory	A4, A5	Fuel handling, storage, transport, and control
	(n,f)	above threshold	30 ?	50	$\pm$ 50 % from theory		
	$T_{1/2}$ (s.f.)		1	3		A8	

C. Report of the Working Group on the Requirements of TND in Waste Management and isotope applications (Chairman: L. Hjaerne)

This working group was the 'catch-all' for everything other than the explicit applications of actinide cross sections in thermal and fast reactor development, design and operation. Hence the subject matter for discussion in the working group was concerned with various energy and non-energy applications of actinide nuclear data, with some emphasis on the back end of the nuclear fuel cycles, which is a matter of much current interest.

The working group first brought together a list of requests for actinide cross section data. From that list were then excluded all those requests which have obviously been met by measurements. The working group then attempted to assign priorities to the individual requests according to the urgency and timeliness of the needs in the various laboratory and national programmes, as expressed in reports and papers available at the time of the meeting. The criteria for priorities are of the usual kind described in WRENDA-75 (INDC(SEC)-46, June 1975). Data requests which have been assigned priority 1 have, in other words, been selected for maximum practicable attention. Priority 2 has been assigned to requests for data which will be required during the next few years in the energy programs, particularly in the area of waste management. It was convenient to list all $\bar{\nu}$-requests in a separate table (Table 7). Decay data in Table 6 are more difficult to categorize according to priorities, as this would often mean a subjective comparison of incomparable entities and the working group therefore abstained from making such an excercise. Table 5 lists the actinide cross section requirements for energy and related applications.

Table 5

Comparison of Requirements and Status of Cross Section Data for Energy and Isotope Applications

Nuclide (half-life)	Data type	Energy range	Achieved Accuracy	Required Accuracy	Needs	Reference Review Paper	Priority
Th 232 (1.407×10^{10} y)	(n,3n)	fast	no	50 %	Fuel handling	A6	3
	(n,2n)	fast	no	50 %	Fuel handling	A6	3
	delayed neutron yield				Spectrum measurement	A9	3
U 233	(n,2n)	fast	10 % (fission spectrum)	10 %	Fuel handling	A1	2
Np 237 (2.1×10^{6} y)	(n,2n) (γ,n)	—	12 %	10 %	Radioisotope Power Sources	A1	2
Pu 236 (2.8y)	(n,γ) (n,f)	resonance	no	20 %	Fuel handling	A1	3
Pu 238 (8.8y)	(n,3n)	resonance and fast	no	50 %	Fuel handling	A1	3
	(n,f)	resonance and fast	40 %	20 %	Actinide recycling	A6	2
Pu 239 (24000 y)	(n,2n)	fast	no	10 %	Fuel handling	A1	2
Pu 240 (6540 y)	(n,γ) resonance parameters	1 ev resonance		5 %	Discrepancies	B1	2
Pu 242 (3.9×10^{5} y)	(n,γ)	fast	no	30 %	Actinide recycle	A6	1
Am 241 (433 y)	(n,γ) branching ratios	thermal, fast (cut-off energies unknown)	10 %	10 %	Radioisotope power sources, fuel handling and others	A1 B1	1
Am 242g (16 h)	(n,γ) (n,f) total	thermal, fast	no	50 %	Actinide recycle	A1 A6	3
Am 242m (152 y)	(n,γ) (n,f) total	thermal, fast		20 %	Actinide recycle	B1 B6 B1	1
Am 243 (7.37×10^{3} y)	(n,γ) (n,2n)	fast		50 %	Actinide recycle Radioisotope power sources	A1, A6 A1	1 3
Cm 242 (163 d)	(n,γ)	thermal, resonance	(50 %)	20 %		B1	2
		fast	no	30 %		A6	2
	(n,f)	fast	no	20 %		A6	2
	total	thermal, resonance	no	20 %		B1	2
Cm 243 (30 y)	(n,γ)	all	no	30 %		A1, A6	2
	Γ_t	resonance	20 %	15 %		B1	2
	(n,f)	fast	?	20 %		A6	2
Cm 244 (17.9 y)	(n,γ)	fast	no	30 %		A6	2
	(n,f)	fast	?	20 %		A6	3
Cm 245 (8.5×10^{3} y)	(n,γ)	fast	no	50 %		A6	2
	α	fast	no	20 %			2
Cm 246 (4.76×10^{3} y)	(n,γ)	thermal, resonance	20 %	10 %		B1	2
		fast	no	factor 2		A6	3
Cm 247 (1.54×10^{7} y)	(n,f)	thermal, resonance 30 eV	no	5–10 %		WRENDA 75	3
		fast	20 %	10 %		B4	3
Bk 249 (311 d)	$I_{n\gamma}$	(cut-off energies unknown)	10 %	10 %		B1	2
Cf 250 (13.1 y)	(n,γ)	thermal	15 %	10 %		B1	2
	(n,f)	resonance	no	10 %		B1	3
Cf 251 (900 y)	(n,f)	thermal, resonance	no	10 %		B1	2

Table 6

Comparison of Requirements and Status of Half-life and Decay Data for Energy and Isotope Applications

Nuclide	Data Type	Achieved Accuracy *	Required Accuracy	Needs	Reference Review Paper
Th 232	yield of 55-s and 22-s delayed-neutron groups	unknown	5 %	neutron spectrum determination	A8
U 233	yield of 55-s and 22-s delayed neutron groups	unknown	5 %	neutron spectrum determination	A8
U 234	α-intensity	3 %	1 %	mass determination	A9
	γ-intensity	factor of 5	5 %	nondestructive and destructive fuel assay	A8
U 235	$T_{1/2}$ (α)	2 %	1 %	mass determination fuel assay	A8, A9
	α-intensities	10 %	1 %	mass determination	A9
	γ-intensities	unknown	1 %	fuel assay, mass determination	A8, A9
	yield of 55-s and 22-s delayed neutron groups	unknown	5 %	neutron spectrum determination	A8
U 236	$T_{1/2}$ (α)	2 %	1 %	mass determination	A8, A9
U 238	α-intensities	5 %	1 %	mass determination	A9
	yield of 55-s and 22-s delayed neutron groups	unknown	5 %	neutron spectrum determination	A8
Np 237	α-intensities	20 %	1 %	mass determination	A9
	yield of 55-s and 22-s delayed neutron groups	unknown	5 %	neutron spectrum determination	A8
Pu 238	$T_{1/2}$ (α)	1.5 %	0.5 %	destructive fuel assay	A8
	"	"	"	Calorimetry	A8
	"		0.02 %	high-precision mass standard	(Aten)
	γ-intensities	25 %	1 %	nondestructive fuel assay	A8
	L-x ray intensity	?	30 %	health-physics applications	A9
	α-intensities	1 %	0.1 %	mass determination	A9
Pu 239	$T_{1/2}$ (α)	1 %	0.2 %	destructive fuel assay, calorimetry, mass determination for safeguards	A8
	γ- intensities	10 %	1 %	nondestructive fuel assay for safeguards	A8
	specific power	1 %	0.2 %	calorimetry	A8
	L-x rays	100 %	30 %	chemical research	A9
	α-intensities	2 %	1 %	mass determination	A8
	yield of 55-s and 22-s delayed neutron groups	unknown	5 %	neutron spectrum determination	A8
Pu 240	$T_{1/2}$ (α)	5 %	0.2 %	destructive fuel assay, calorimetry, mass determination	A8
	γ-intensities	factor of 4	1 %	nondestructive fuel assay	A8
	α-intensities	1 %	0.2 %	mass determination	A8
	L-x ray intensity	unknown	30 %	chemical research	A9
Pu 241	$T_{1/2}$ (α)	5 %	1 %	destructive fuel assay	A8
		"	2 %	fast reactor	A8
	γ-intensities	5 %	1 %	nondestructive fuel	A8
Pu 242	$T_{1/2}$ (α)	5 %	1 %	mass determination	A9
	α-intensities	10 %	4 %	mass determination	A9
Am 241	γ-intensities	2 %	1 %	intensity standards	A9
	L-x ray intensity	5 %	1 %	intensity standards	A8
	yield of 55-s and 22-s delayed neutron groups	unknown	5 %	neutron spectrum determination	A8
Cm 242	$T_{1/2}$ (α)	1 %	0.1 %	decay correction in destructive fuel assay	A8
Cf 252	$T_{1/2}$ (α)	1 %	0.2 %	decay correction	A9

* In the column "Achieved Accuracy" the listed values generally represent the range of the measured data.

Table 7

Comparison of Requirements and Status of Neutron Induced $\bar{\upsilon}$ Data for Energy and Isotope Applications

Nuclide	$\bar{\upsilon}$	Energy	Achieved Accuracy	Required Accuracy	Priority	Reference Review Paper
Pa 231	$\bar{\upsilon}_{pr}$	fast	-	10 %	3	A6
U 232	"	fast	-	10 %	3	A6
Np 237	"	fast	-	10 %	2	A6
Pu 238	"	fast	-	10 %	2	A6
Pu 242	"	fast	10 %	3 %	2	B1
Am 241	"	fast	-	10 %	2	A6
Am 242	"	fast	-	10 %	2	A6
Am 243	"	fast	-	10 %	2	A6
Cm 242	"	fast	-	10 %	2	A6
Cm 243	"	fast	-	10 %	2	A6
Cm 244	"	fast	-	10 %	2	A6
Cm 245	"	fast	-	20 %	3	A6
U 236	$\bar{\upsilon}$ del	fast	-	10 %	2	B1
Pu 240	"	fast	-	20 %	2	B1
Pu 241	"	all	-	10 %	2	B1
Pu 242	"	fast	-	20 %	3	B1

ADVISORY GROUP MEETING ON TRANSACTINIUM ISOTOPE NUCLEAR DATA

Karlsruhe, FRG, 3-7 November 1975

LIST OF REVIEW PAPERS

A. Survey of TND Applications

1. General Survey of Applications of Actinide Nuclear Data in the Nuclear Fuel Cycle and the Nuclear Industry. (S. Raman)

2. Importance of TND in the Physics Design of Fast and Thermal Reactor Cores. (J.Y. Barré, J. Bouchard)

3. Sensitivity Studies of Build-up of Actinides during Normal Reactor Operations. (H. Kuesters)

4. Importance of TND for Engineering Design and Operations of Reactors. (R.G. Nunn)

5. Importance of TND for Fuel Handling (excluding waste management). (R.F. Burstall)

6. European Programmes in Waste Management (Incineration) of Actinides. (L. Koch)

7. US Programmes in the Waste Management (Incineration) of Actinides. (S. Raman)

8. Importance of TND for Fuel Analysis. (R. Dierckx)

9. Importance of TND in Non-reactor Applications. (A.H.W. Aten, Jr.)

B. Review of TND Status

1. Status of Measured Neutron Cross Sections of Transactinium Isotopes for Thermal Reactors. (R.W. Benjamin)

2. Status of Neutron Cross Sections of Transactinium Isotopes in the Resonance Energy Region - Linear Accelerator Measurements. (G.D. James)

3. Status of Neutron Cross Section of Transactinium Isotopes in the Resonance and Fast Energy Regions - Underground Nuclear Explosion Measurements. (M.S. Moore)

4. Status of Measured Neutron Cross Section of Transactinium Isotopes in the Fast Region. (S. Igarasi)

5a. Status of Transactinium Isotope Evaluated Neutron Data in the Energy Range 10^{-3} eV to 15 MeV. (S. Yiftah)

5b. Evaluation and Theoretical Calculation of TND. (J.E. Lynn)

6. Status of Alpha Decay Data of the Transactinium Isotopes. (A.G. Zelenkov/S.A. Baranov)

7. Status of Beta and Gamma Decay and Spontaneous Fission Data from Transactinium Isotopes. (C.W. Reich)

ADVISORY GROUP MEETING ON

TRANSACTINIUM ISOTOPE NUCLEAR DATA

Karlsruhe, FRG, 3-7 November 1975

MEETING PROGRAMME

Monday, 3 November

- Opening addresses by J.J. Schmidt and Prof. Böhm
- Outline of meeting procedure and objectives
- Organization of Working Groups
- Appointment of chairmen
- Presentation and discussion of Review Papers A1 to A5.

Tuesday, 4 November

- Presentation and discussion of Review Papers A6 to A9, and B1 to B4
- Short meeting of Working Groups.

Wednesday, 5 November

- Presentation and discussion of Review Papers B5 to B7
- Visit to the Karlsruhe Nuclear Research Centre.

Thursday, 6 November

- Independent work sessions of working groups to draft conclusions and recommendations.

Friday, 7 November

- Final plenary session for the presentation, discussion and adoption of recommendations and working group reports.

ADVISORY GROUP MEETING ON

TRANSACTINIUM ISOTOPE NUCLEAR DATA

Karlsruhe, FRG, 3-7 November 1975

LIST OF PARTICIPANTS

(Reviewers indicated by an R after their name)

Aleta, C.		Philippine Atomic Energy Agency Don Mariano Marcos Ave. Diliman, Quezon City, Philippines
Aten, A.H.W.	R	IKO Institute for Nuclear Research 18 Oosterringdijk P.O. Box 4395 Amsterdam-1006 The Netherlands
Bambynek, M.		CBNM Steenweg op Retie 2440 Geel Belgium
Barré, J.Y.	R	DEDR/DRE/SPNR CEN-Cadarache B.P. No. 1 13115 Saint Paul lez Durance France
Benjamin, R.	R	Savannah River Laboratory E.I. du Pont de Nemours & Co. Aiken, South Carolina 29801 USA
Bouchard, J.	R	DEDR/DRE/SPNR CEN-Cadarache B.P. No. 1 13115 Saint Paul lez Durance France
Burstall, R.F.	R	Central Technical Services Risley, Warrington, Lancashire, WA3 6AT United Kingdom

Cierjacks, S.		Kernforschungszentrum Karlsruhe Postfach 3640 7500 Karlsruhe Fed. Rep. of Germany
Cricchio, Dr.		C.E.C. Direction Generale Affaires Industrielles at Technologiques 200 rue de la Loi 1049 Bruxelles Belgium
Derrien, H.		NEA Neutron Data Compilation Centre B.P. No. 9 91190 Gif-sur-Yvette France
Deruytter, A.J.		University of Gent Instituut voor Kernwetenschappen 48 Proeftuinstraat 9000 Gent Belgium
Dierckx, R.	R	Centre Commun de Recherche C.E.C, Physics Division 21020 Ispra (Varese) Italy
Fudge, A.J.		U.K.A.E.A. Bldg. 220 Atomic Energy Research Establishment Harwell, Didcot, Oxfordshire OX 11 ORA United Kingdom
Glover, Mrs. K.M.		U.K.A.E.A. Bldg. 220 Atomic Energy Research Establishment Harwell, Didcot, Oxfordshire, OX 11 ORA United Kingdom
Grenier, G.		CEN-Bruyeres-le-Chatel B.P. No. 61 92120 Montrouge France
Hage, M.W.		Centre Commun de Recherche C.E.C, Physics Division 21020 Ispra (Varese) Italy

Haug, H.		Kernforschungszentrum Karlsruhe Postfach 3640 7500 Karlsruhe Fed. Rep. of Germany
Hjaerne, L.		ASEA-ATOM Box 53 72104 Vaesteras 1 Sweden
Igarasi, S.	R	Japan Atomic Energy Research Inst. Tokai-Mura, Naga-gun Ibaraki-ken 319-11 Japan
James, G.D.	R	U.K.A.E.A. Atomic Energy Research Establishment Harwell, Didcot, Oxfordshire, OX11 ORA United Kingdom
Kalebin, S.M.		Institute of Theoretical and Experimental Physics Moscow, USSR
Koch, L.	R	European Institute for Transuranium Isotopes P.O. Box 2266 7500 Karlsruhe Fed. Rep. of Germany
Kulakov, V.M.	R	I.V. Kurchatov Atomic Energy Inst. Moscow USSR
Kuesters, H. (local Secretary)	R	Kernforschungszentrum Karlsruhe Postfach 3640 7500 Karlsruhe Fed. Rep. of Germany
Lalovic, M.		Kernforschungszentrum Karlsruhe Postfach 3640 7500 Karlsruhe Red. Rep. of Germany

Legrand, J.		CEN-Saclay B.P. No. 2 91190 Gif-sur-Yvette France
Liljenzin, J.		Chalmers Tekniska Hoegskola Department of Nuclear Chemistry Fack 402 20 Goeteborg 5 Sweden
Lorenz, A. (Scientific Secretary)		Nuclear Data Section International Atomic Energy Agency P.O. Box 590 1011 Vienna Austria
Lutz, D.		Institut f. Kernenergietechnik der Universitaet Stuttgart Pfaffenwaldring 31 7 Stuttgart 80 Fed. Rep. of Germany
Lynn, J.E.	R	U.K.A.E.A. Atomic Energy Research Establishment Harwell, Didcot, Oxfordshire, OX 11 ORA United Kingdom
Matsuura, Sh.		Japan Atomic Energy Research Inst. Tokai-Mura, Naga-gun Ibaraki-ken 319-11 Japan
Menapace, E.		Centro di Calcolo del C.N.E.N. Via Mazzini 2 40138 Bologna Italy
Moore, M.S.	R	Los Alamos Scientific Laboratory P.O. Box 1663 Los Alamos, New Mexico 87544 USA
Nunn, R.M.	R	Berkeley Nuclear Laboratory C.E.G.B. Berkeley Gloucestershire, GL 13 9PB United Kingdom

Parus, J.L.		Institute of Nuclear Research Warsaw Poland
Philis, C.		CEN-Bruyeres-le-Chatel B.P. No. 61 92120 Montrouge France
Raman, S.	R	Oak Ridge National Laboratory Physics Division P.O. Box X Oak Ridge, Tennessee 37830 USA
Reich, C.W.	R	Idaho National Engineering Laboratory Aerojet Nuclear Co. Idaho Falls, Idaho 83401 USA
Rohr, G.		CBNM Steenweg op Retie 2440 Geel Belgium
Salvy, J.		CEN-Bruyeres-le-Chatel B.P. No. 61 92120 Montrouge France
Sola, A.		Centre Commun de Recherche C.E.C. Physics Division 21020 Ispra (Varese) Italy
Schatz, M.		Kraftwerk Union Erlangen Fed. Rep. of Germany
Schmidt, J.J. (Soientific Secretary)		Nuclear Data Section International Atomic Energy Agency P.O. Box 590 1011 Vienna Austria
Schmitt, A.P.		DEDR/DRE/SENR CEN-Cadarache B.P. no. 1 13115 Saint Paul lez Durance France

Scholtyssek, W.		Kernforschungszentrum Karlsruhe Postfach 3640 7500 Karlsruhe Fed. Rep. of Germany
Weston, L.W.		Neutron Physics Division Oak Ridge National Laboratory P.O. Box X Oak Ridge, Tennessee 37830 USA
Yiftah, S.	R	Department of Nuclear Engineering Technion Israel Institute of Technology Haifa Israel

Review Paper A1

General Survey of Applications Which Require Actinide Nuclear Data*

S. Raman

Oak Ridge National Laboratory, Oak Ridge, Tennessee 37830

> 'You don't seem to give much thought to the matter in hand,' I said at last, interrupting Holmes' musical disquisition.
>
> 'No data yet,' he answered. 'It is a capital mistake to theorize before you have all the evidence. It biases the judgment.'
>
> – *A Study in Scarlet,* Sir Arthur Conan Doyle

This review paper discusses the actinide waste problem, the buildup of toxic isotopes in the fuel, the neutron activity associated with irradiated fuel, the ^{252}Cf buildup problem, and the production of radioisotope power sources as broad areas that require actinide cross-section data. Decay data enter into the area of radiological safety and health physics. This paper also discusses a few cross-section measurements in progress at the Oak Ridge Electron Linear Accelerator. The availability of actinide samples through the Transuranium Program at Oak Ridge is discussed in considerable detail. The present data status with respect to the various applications is reviewed along with recommendations for improving the data base.

I. INTRODUCTION

Since the discovery of fission in 1939, nuclear physicists and chemists have been called upon to produce neutron cross sections and other nuclear data for a large number of actinides (Ac and higher Z elements). And produce they did in impressive quantity and quality with the end result that electric power generation through fission reactors has become a practical reality. However, it appears that any large-scale development of nuclear energy, while solving many pressing problems, also creates new ones. A major problem is the potential hazard associated with the fuel, fertile material, and wastes. These products must be processed, transported, stored, and safeguarded with

*Research sponsored by the U. S. Energy Research and Development Administration under contract with Union Carbide Corporation.

extreme circumspection. Reactor fuel elements need to be processed in order to recover reusable fuel and fertile material. The unrecovered portions are usually called "wastes" which can be broadly divided into fission products and actinides. Most fission products decay to harmless nonradioactive elements in a relatively short span of <1000 years. It is generally believed that adequate storage means, either underground or above, can be found to handle most fission product wastes. The fuel, fertile material, and actinide wastes pose a different type of problem because several isotopes among them possess very long half-lives. For example, the main new fuel produced in a light-water ^{235}U–^{238}U reactor is ^{239}Pu which has a half-life of 24,400 yr. The half-life of ^{243}Am, a waste material, is 7370 yr. The hazards associated with the fissionable isotopes of the fuel are, however, of secondary concern because these isotopes can be depleted through their use in reactors. Similarly, the hazards due to the fertile materials can be minimized through recovery and reuse. This leaves us to contend with a third category of nuclides, specifically Np, Am, and Cm isotopes and the unrecovered portions of U and Pu isotopes, that would be, using present technology, routinely discharged along with the fission products in the high-level waste stream and temporarily stored above ground pending their final disposition. One should perhaps add to this category the nonfissionable isotopes of U and Pu fuels which are produced in substantial quantities (see Table I) through reactor operations. There are many who feel that the very future of nuclear power may be governed by how satisfactorily we can determine, evaluate, and handle the risks from the fission products and the actinides.

Consider the magnitude of the actinide waste problem. With present-day fuel processing methods, it is estimated that in the United States about 110 metric tons of actinides will be discharged in waste annually by the year 2000, and that the accumulated inventory would be in excess of 1300 metric tons [2]. These quantities are based on a projection of 1200 reactors of 1000 MW(e) each by the year 2000. By comparison, the fuel loading of a 1000-MW(e) light-water reactor is about 80 metric tons.

The actinide wastes must be interred nearly forever somewhere — but where? Some geologists have argued that geological stability and complete containment cannot be easily guaranteed for periods of the order of 250,000 years required for the actinides to decay to innocuous levels. Therefore, methods other than simple burial need to be studied. One suggested method for alleviating the actinide waste problem would involve fissioning the actinides in a suitable reactor. Among Pu, Am, and Cm isotopes, odd-neutron nuclides (^{239}Pu, ^{241}Pu; ^{242}Am, ^{244}Am; ^{243}Cm, ^{245}Cm) possess appreciable

TABLE I

Approximate amounts of transactinium isotopes in a 1000-MW(e) light-water reactor at the end of three-year power generation and three-year cooling. The initial fuel composition was ^{234}U (20 kg), ^{235}U (2630 kg), and ^{238}U (77,350 kg). The calculations were made with the ORIGEN code [1]. The flux was 2.9×10^{13} n/cm^2 · sec.

Isotope	Quantity	Isotope	Quantity	Isotope	Quantity
^{238}U	75,400 kg	^{241}Am	13 kg	^{247}Cm	230 mg
^{235}U	640 kg	^{234}U	10 kg	^{230}Th	154 mg
^{239}Pu	420 kg	^{243}Am	8 kg	^{232}Th	50 mg
^{236}U	360 kg	^{244}Cm	2 kg	^{232}U	49 mg
^{240}Pu	170 kg	^{245}Cm	150 g	^{231}Pa	43 mg
^{241}Pu	70 kg	^{242m}Am	74 g	^{248}Cm	15 mg
^{237}Np	39 kg	^{246}Cm	18 g	^{236}Pu	12 mg
^{242}Pu	30 kg	^{242}Cm	8 g	^{250}Cf	180 μg
^{238}Pu	14 kg	^{243}Cm	6 g	^{252}Cf	3 μg
		^{233}U	424 mg		

fission cross sections, and the remaining isotopes can be transmuted to these by neutron capture. Therefore, in principle, the actinides can be partly converted into more manageable fission products.

A detailed analysis is needed to establish the technical and economic feasibility of this actinide disposal method. To carry out such an analysis, basic research is needed in the following three disciplines: (i) chemical techniques to effect maximal separation of the actinides and the more abundant fission products, (ii) neutron cross-section measurements where gaps in needed data exist, and (iii) reactor sensitivity and transmutation calculations.

The transmutation of actinides is a function of the product of the flux, the absorption cross section, and the irradiation time. There are conspicuous lacunae in our knowledge of actinide cross sections. The "differential" cross sections, i.e., the particular reaction probabilities as a function of neutron energy, have so far been measured only for those isotopes for which enriched isotopic samples were readily available. New actinide cross-section measurements are needed, at the very least, for detailed testing of the actinide recycle concept in existing reactors and for implementing the concept. At most, these new measurements would dictate recycling strategy and perhaps result in entirely new types of reactors.

A second major application with numerous nuclear data requirements is fuel management. The efficient operation of reactors requires a detailed knowledge of transuranium isotope buildup and burnout. The planning of operating modes, core loadings, and fuel requirements depends on the quantities of various isotopes in the fuel. Computer programs, validated with dissolved fuel isotopic analysis, are now employed to calculate fuel burnout. These codes are reasonably successful in predicting most Pu concentrations, but the determination of transplutonium isotopes is less certain. Experimentally measured Am and Cm isotopic compositions and improved neutron cross-section values are required to reduce the uncertainties in the calculations of the actinide isotope concentrations. Of special interest are improved neutron capture and absorption cross sections for ^{242}Pu, ^{241}Am, ^{242g}Am, ^{242m}Am, and ^{242}Cm. Generally, such cross sections are needed with 3-10% accuracy for the thermal through the resonance neutron energy region. With the availability of improved cross-section data, more realistic estimates of Pu, Am, and Cm isotopes in discharged fuel can be obtained. In particular, ^{241}Am, ^{242}Pu, and ^{243}Am are starting points for producing Cm isotopes and higher Z elements, and ^{238}Pu, ^{242}Cm, and ^{244}Cm are neutron emitters. Another major neutron emitter, ^{252}Cf, would be produced in increasing quantities if the actinides were recycled. Inventories of the above isotopes are especially needed for determining the impact of their heat and neutron generation on irradiated fuel handling, storage, and transportation.

The two nuclides which appear most attractive for use in compact energy sources are ^{238}Pu and ^{244}Cm. These isotopes combine reasonable power densities with reasonable half-lives [3]. The ^{238}Pu is currently used as an energy source for heart pacemakers and will most likely be used in the development of an artificial heart. Problem areas common to both ^{238}Pu and ^{244}Cm are technological ones such as demonstration of compatibility of these isotopes with encapsulants, evaluation of safety problems both in normal and accident modes, control of decay-produced helium, etc. The nuclear characteristics and production methods for both isotopes are fairly well established, and additional investigations in this area would be data refinement rather than determination of new data. In production methods, the two materials are different; ^{238}Pu is made by neutron irradiation of specially prepared targets while ^{244}Cm is a by-product of the operation of nuclear power reactors. Thus, ^{244}Cm will be available in tremendous amounts (see Table I) in the near future. However, there are at present no plans for the isolation of ^{244}Cm from reactor wastes, so its use in power sources is only a potential, not a reality. One problem which is very difficult to evaluate, and which probably applies more to ^{238}Pu than to ^{244}Cm, is that of public acceptance. The general public

has a proclivity to translate "plutonium" as "bomb", with no discrimination made amongst the different isotopes of plutonium.

Cross-section measurement programs to study the uncharted actinide regions have been proposed or are currently underway in several laboratories in the United States [4]. Proposed measurements with an underground nuclear explosion will be discussed by Moore [5]. Preliminary cross-section results for the ^{241}Am(n, γ), the ^{245}Cm(n,fission), and the ^{249}Bk(n,total) reactions, carried out at Oak Ridge, are described in a later section. The minimum quantities of separated isotope needed are about 30 μg, 10 mg, and 100 mg for fission, total, and capture cross-section measurements, respectively, although it might be feasible to work with even smaller amounts. In many cases, the required sample sizes and isotopic purities restrict presently possible measurements. For the (n, f) measurements on Cm isotopes, it would be important, for instance, that the concentration of the even isotopes ^{244,246,248}Cm be very low because of their spontaneous fission rates. Since sample acquisition, purification, and preparation require major efforts, a detailed discussion of the problems encountered on an isotope-by-isotope basis has been included in a later section.

In the areas of radiological safety and health physics, actinide nuclear data are important for establishing permissible organ burdens through the estimation of internal radiation dose from ingested or inhaled radionuclides. These organ burdens are combined with biological transport rates (concentration in food chains, deposition on tobacco, etc.) to arrive at maximum permissible concentrations (MPC) in air, water, and food. The MPC values govern allowable routine releases from reactors. They also determine the design of containment for radiological protection in fuel handling facilities and in research laboratories.

The internal dose is dependent on the concentration of the radionuclide in the organs and on the decay scheme of the nuclide and its progeny. The chemical properties and biological processes will influence the deposition of elements in the organs. The biological aspects are of special concern because studying them is both time consuming and expensive. Obviously, a detailed knowledge of emitted radiations and the decay scheme is an essential prerequisite for computing the dose rate from a particular nuclide.

In the preceding pages, several areas have been specified which require actinide nuclear data. In the following pages, these areas are examined in greater detail.

II. CROSS-SECTION NEEDS FOR RECYCLING ACTINIDES

Figure 1, adapted from the GE Chart of the Nuclides [6], shows the major pathways for the buildup of the heavier actinides. The quantitative prediction of the various

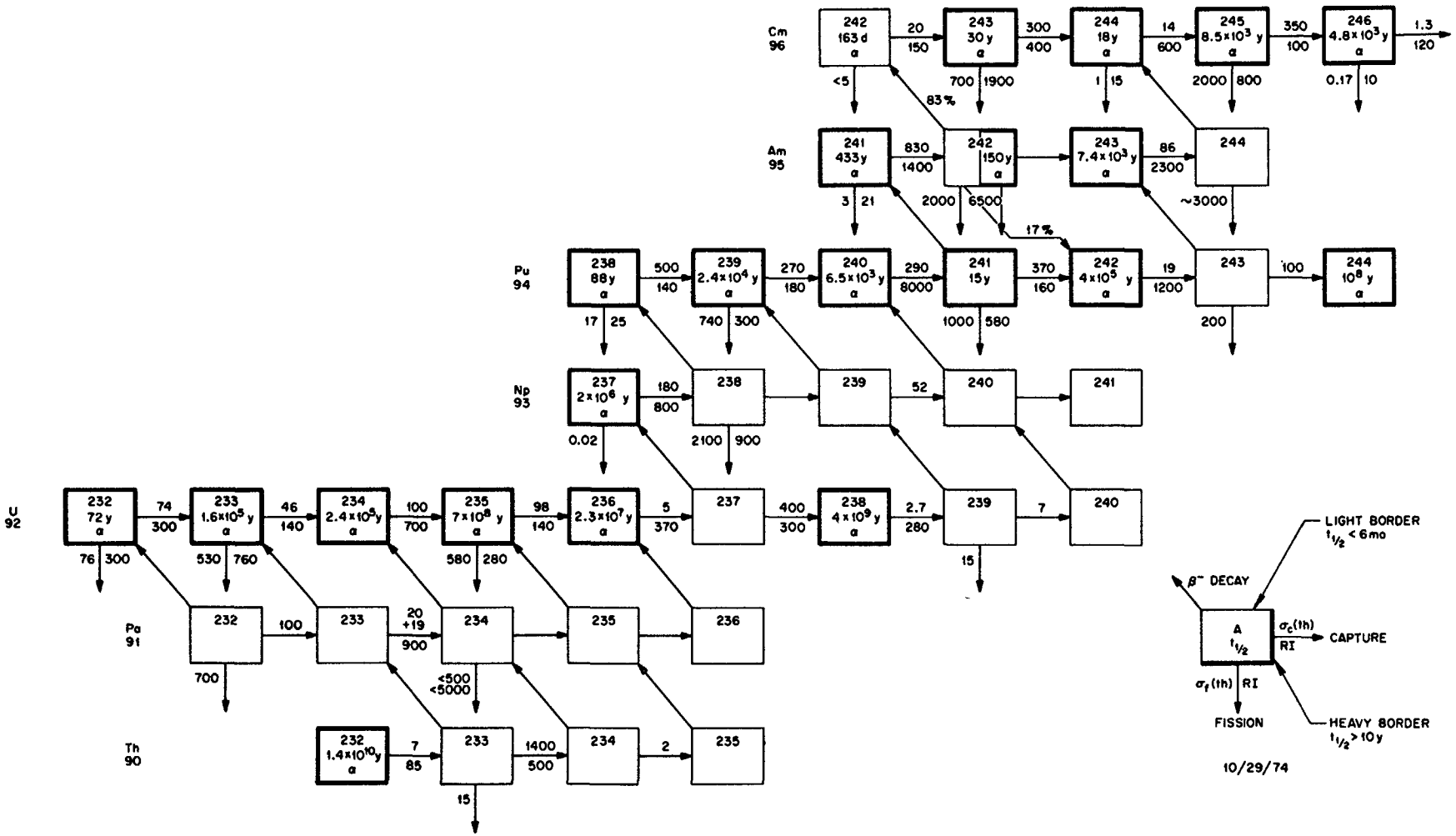


Fig. 1. Some nuclides and cross sections of interest to actinide waste management.

nuclides produced, transmuted, and fissioned in reactors is necessary for systematic actinide management. These quantitative predictions are made with special computer programs, an example of which is the code ORIGEN [1]. This code requires as input values all relevant cross sections of all the actinides in the region of interest. At present, this code utilizes effective values at thermal, resonance, and fast neutron energies obtained from suitable weighted averages over the reactor spectrum under consideration.

Benjamin [7] has surveyed neutron cross sections for the actinides with emphasis on those desired for thermal and near-thermal reactors and has presented a recommended set of thermal neutron cross sections and infinite dilution resonance integrals for the major actinides. A two-group self-consistent set of cross sections for the heavier actinides has been developed at the Oak Ridge National Laboratory-High Flux Isotope Reactor (ORNL-HFIR) by King, Bigelow, and Collins [8]. This set is shown in Table II. Benjamin, McCrosson, Vandervelde, and Gorrell [9] have also developed a self-consistent set of cross sections (divided into 84 energy bins) and resonance parameters for isotopes from ^{242}Pu to ^{253}Es using evaluated nuclear data, reactor production experiments, multigroup production codes, and theory. Benjamin [7] has made approximately a dozen recommendations for new measurements in the thermal and resonance neutron energy region that require special efforts in preparing samples and in developing measurement techniques. Better fission data will be required later because irradiation of the expected large quantities of the heavier actinides will alter reactor charge design characteristics and reactivity transient effects. Reactor sensitivity calculations underway in different laboratories should identify the cross sections that should be known with greater accuracy. In general, it appears that current nuclear data are reasonably adequate for evaluating heavy-actinide production and removal in *thermal* reactors.

The reactor types under active consideration for actinide recycling are not limited to thermal reactors as discussed in another review paper [4]. The neutron spectrum in a typical Liquid Metal Fast Breeder Reactor (LMFBR) is shown in the top part of Fig. 2. In the bottom part of the same figure is shown the neutron flux spectrum in the innermost water blanket zone of a conceptual D-T fusion reactor [10]. A fission reactor operating entirely on the higher actinide "wastes" would have a harder neutron spectrum than that shown for the LMFBR. A D-D plasma fusion reactor would have its maximum neutron flux at a neutron energy of ~2.6 MeV. All these reactor types have been suggested as potential actinide waste-burning systems. Therefore, the range over which the capture and fission cross sections of the actinides need to be known extends from below thermal to ~18-MeV neutron energy unless some of these systems are excluded from consideration on the basis of other criteria.

The present status of the measured fission, capture, and total cross-section data is shown schematically in Fig. 3. This figure is based partly on the review papers by Benjamin [7] and by Igarasi [11] (both presented at this meeting) and partly on the cross-section curves provided by the Brookhaven National Neutron Cross Section Center [12]. These curves for the actinides (except for ^{232}Th, ^{233}U, ^{235}U, ^{238}U, and ^{239}Pu) have been included in this paper as Appendix A. In addition to indicating the energy regions

TABLE II

Neutron cross sections used to compute transmutations in HFIR target irradiations

Nuclide	Half-Life	Capture			Fission		
		2200-m/s Cross Section (barns)	Resonance Self-Shielding Constant	Resonance Integral (barns)	2200-m/s Cross Section (barns)	Resonance Self-Shielding Constant	Resonance Integral (barns)
^{238}Pu	87.404 y	560	0	150	16.5	0	25
^{239}Pu	2.4413×10^4 y	265.7	0	195	742.4	0	324
^{240}Pu	6580 y	290	0	8453	0.05	0	0
^{241}Pu	14.98 y	360	0	166	1011	0	541
^{242}Pu	3.869×10^5 y	19.5	6.20	1280	0	0	0
^{243}Pu	4.955 h	80	0	0	210	0	0
^{244}Pu	8.28×10^7 y	1.6	0	0	0	0	0
^{245}Pu	10.6 h	277	0	0	0	0	0
^{246}Pu	10.85 d	0	0	0	0	0	0
^{243}Am	7370 y	105	0	1500	0	0	0
^{244}Am	10.1 h	0	0	0	2300	0	0
^{244m}Am	26 m	0	0	0	0	0	0
^{244c}Am[a]	49 m	0	0	0	1128	0	0
^{245}Am	2.07 h	0	0	0	0	0	0
^{246}Am	25.0 m	0	0	0	0	0	0
^{244}Cm	18.099 y	10.0	4.0	650	1.2	4.0	12.5
^{245}Cm	8265 y	343	2.4	120	1727	2.4	1140
^{246}Cm	4655 y	1.25	0	121	0	0	0
^{247}Cm	1.56×10^7 y	60	0	500	120	0	1060
^{248}Cm	3.397×10^5 y	3.56	2.0	170	0	0	0
^{249}Cm	64 m	2.8	0	0	50	0	0
^{250}Cm	1.74×10^4 y	2	0	0	0	0	0
^{249}Bk	314 d	1451	2.4	1240	0	0	0
^{250}Bk	3.222 h	350	0	0	3000	0	0
^{251}Bk	57 m	0	0	0	0	0	0
^{249}Cf	352 y	450	1.46	750	1690	5.8	2920
^{250}Cf	13.08 y	1900	20	11600	0	0	0
^{251}Cf	900 y	2850	14	1600	3750	14	5400
^{252}Cf	2.646 y	19.8	0	44	32	0	110
^{253}Cf	17.812 d	12.6	0	0	1300	0	0
^{254}Cf	60.5 d	50	0	1650	0	0	0
^{255}Cf	1.5 h	0	0	0	0	0	0
^{253}Es	20.467 d	345	0	0	0	0	0
^{254}Es	276 d	20	0	0	3060	0	0
^{254m}Es	39.3 h	1.26	0	0	1840	0	0
^{255}Es	39.8 d	60	0	0	0	0	0
^{256}Es	25 m	0	0	0	0	0	0
^{254}Fm	3.24 h	76	0	0	0	0	0
^{255}Fm	20.07 h	26	0	0	100	0	0
^{256}Fm	2.62 h	45	0	0	0	0	0
^{257}Fm	94 d	10	0	0	5500	0	0
^{258}Fm	380 μs	0	0	0	0	0	0

[a] To simplify calculations we use a fictitious isotope, ^{244c}Am, which combines the properties of ^{244m}Am and ^{244}Am according to their relative rates of production from ^{243}Am.

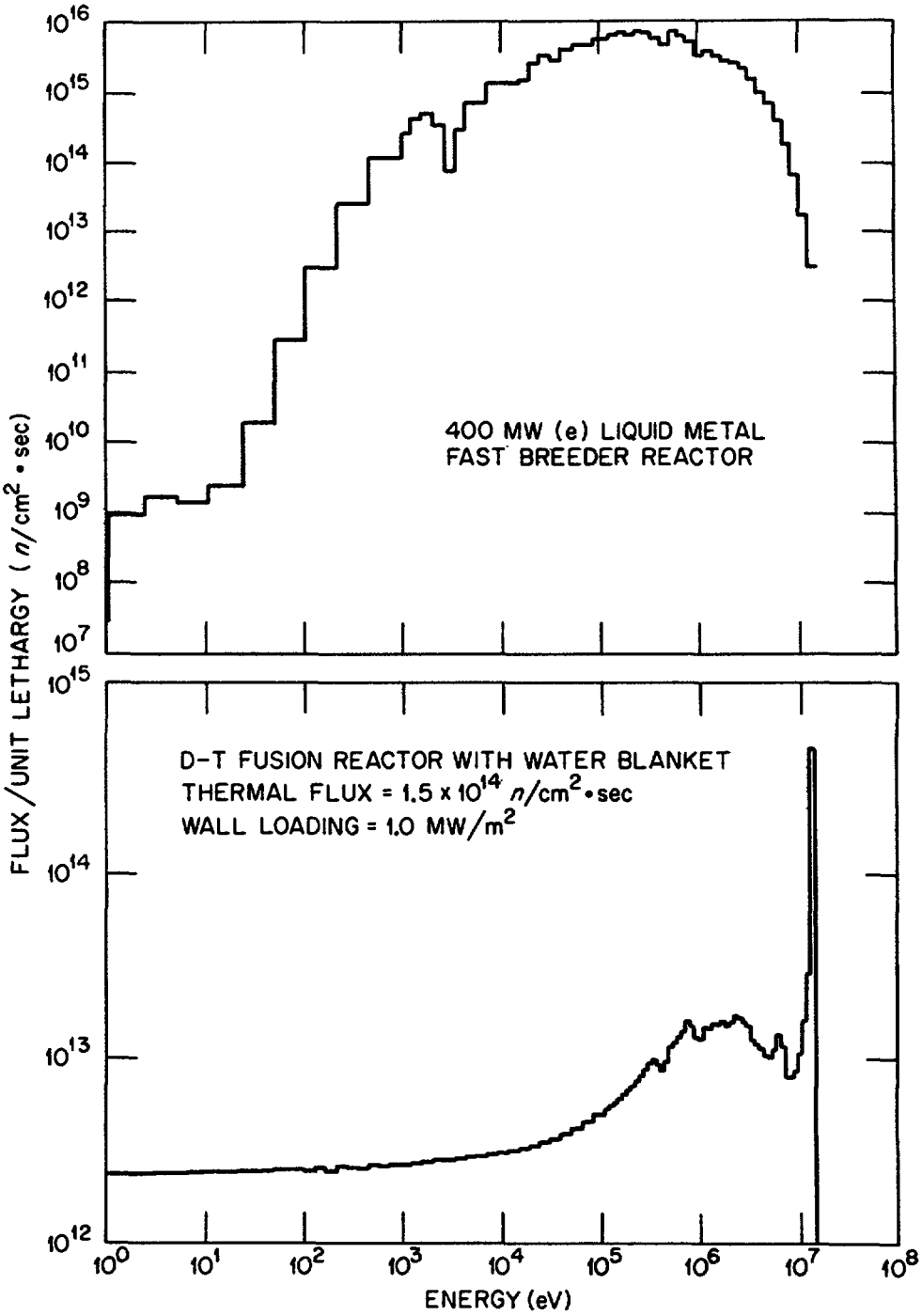


Fig. 2. Neutron spectrum from an LMFBR and from a D-T fusion reactor.

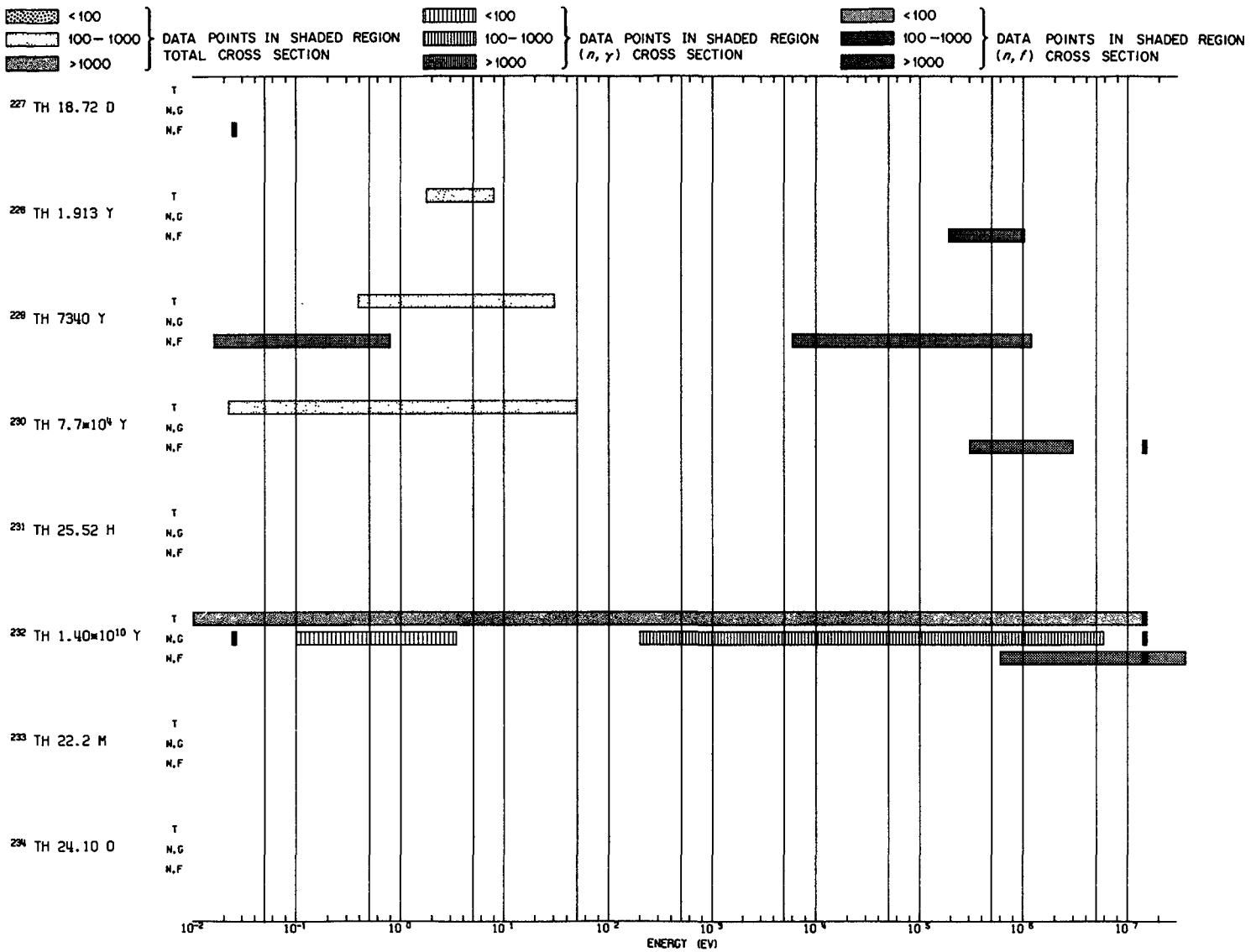


Fig. 3. Present status of actinide cross sections.

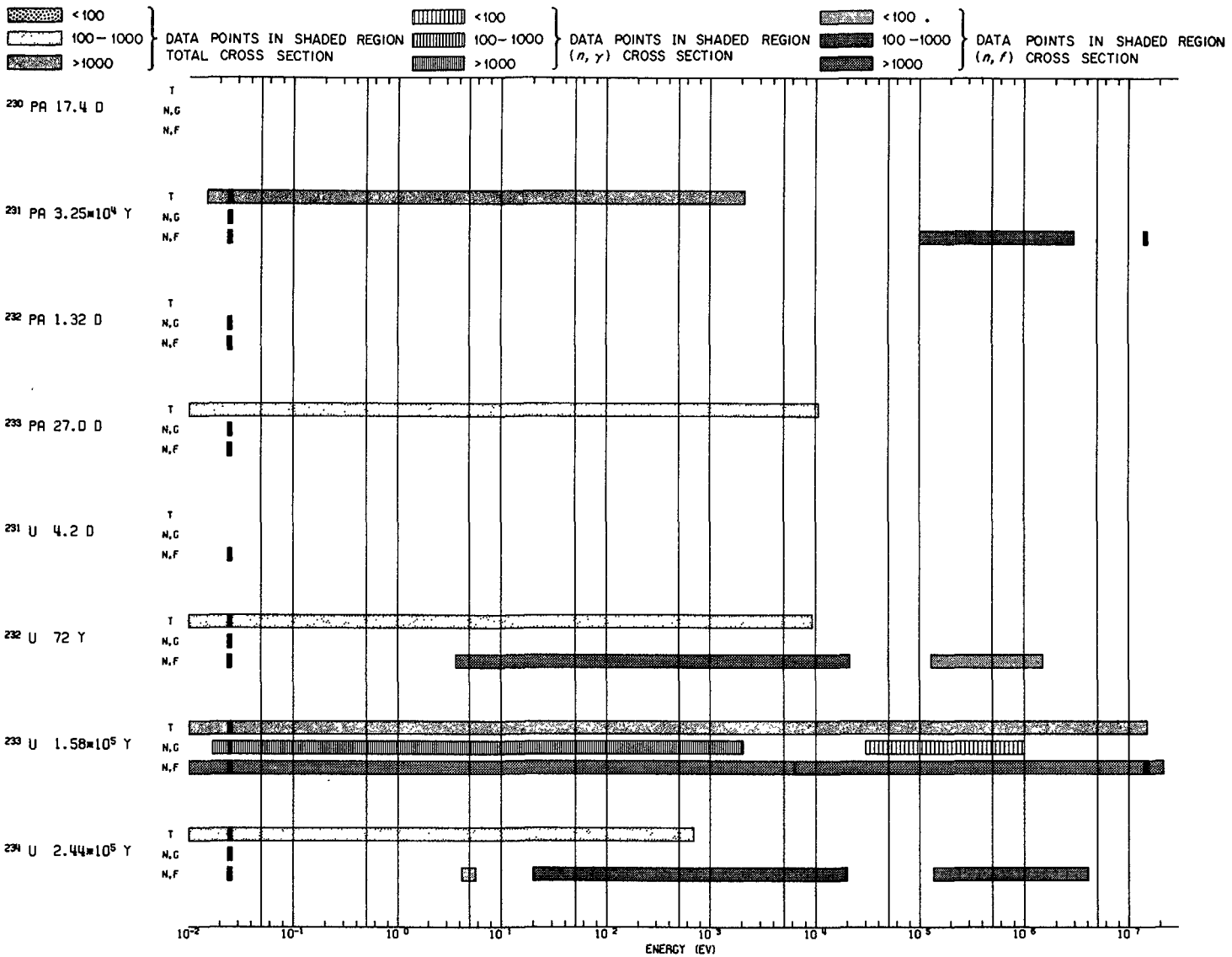


Fig. 3. (continued) Present status of actinide cross sections.

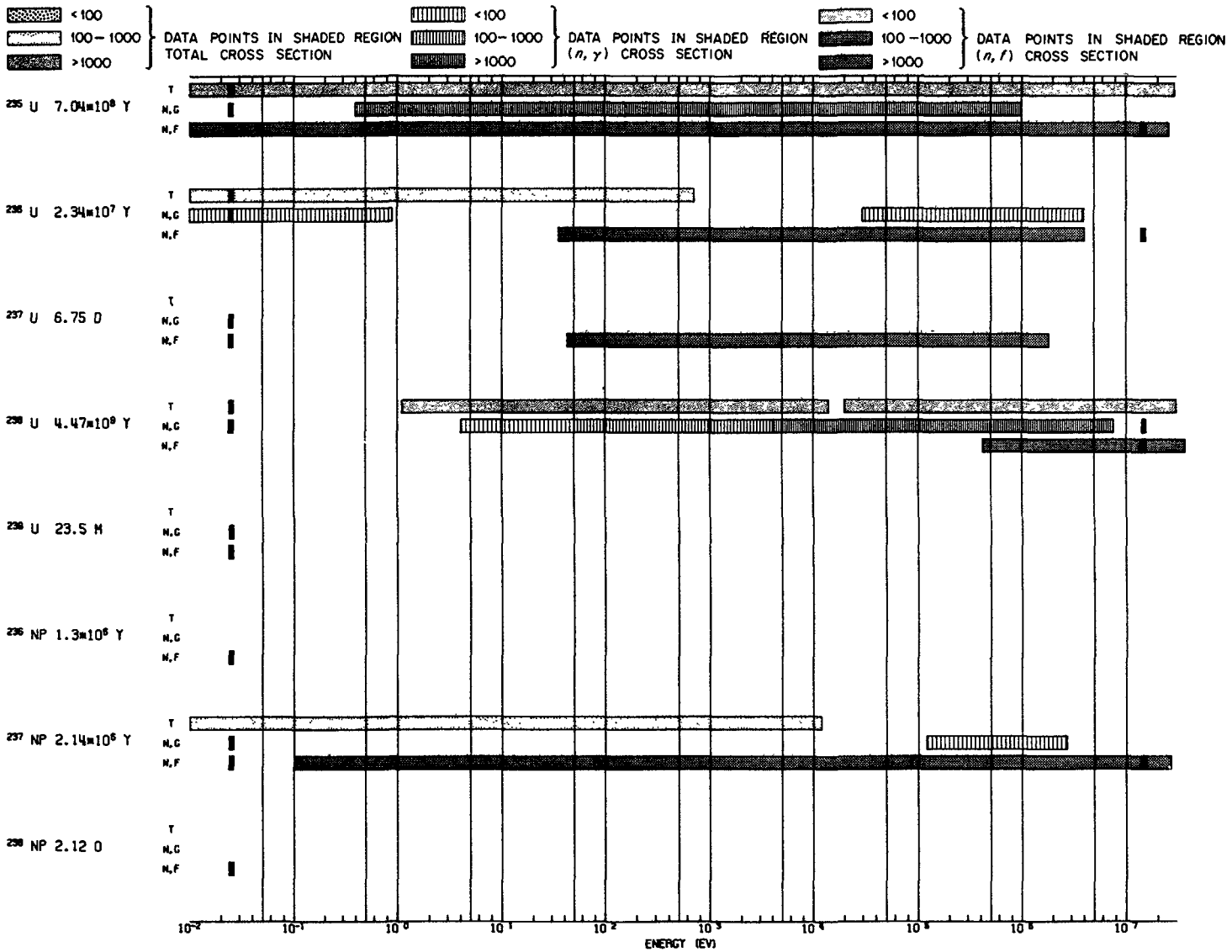


Fig. 3. (continued) Present status of actinide cross sections.

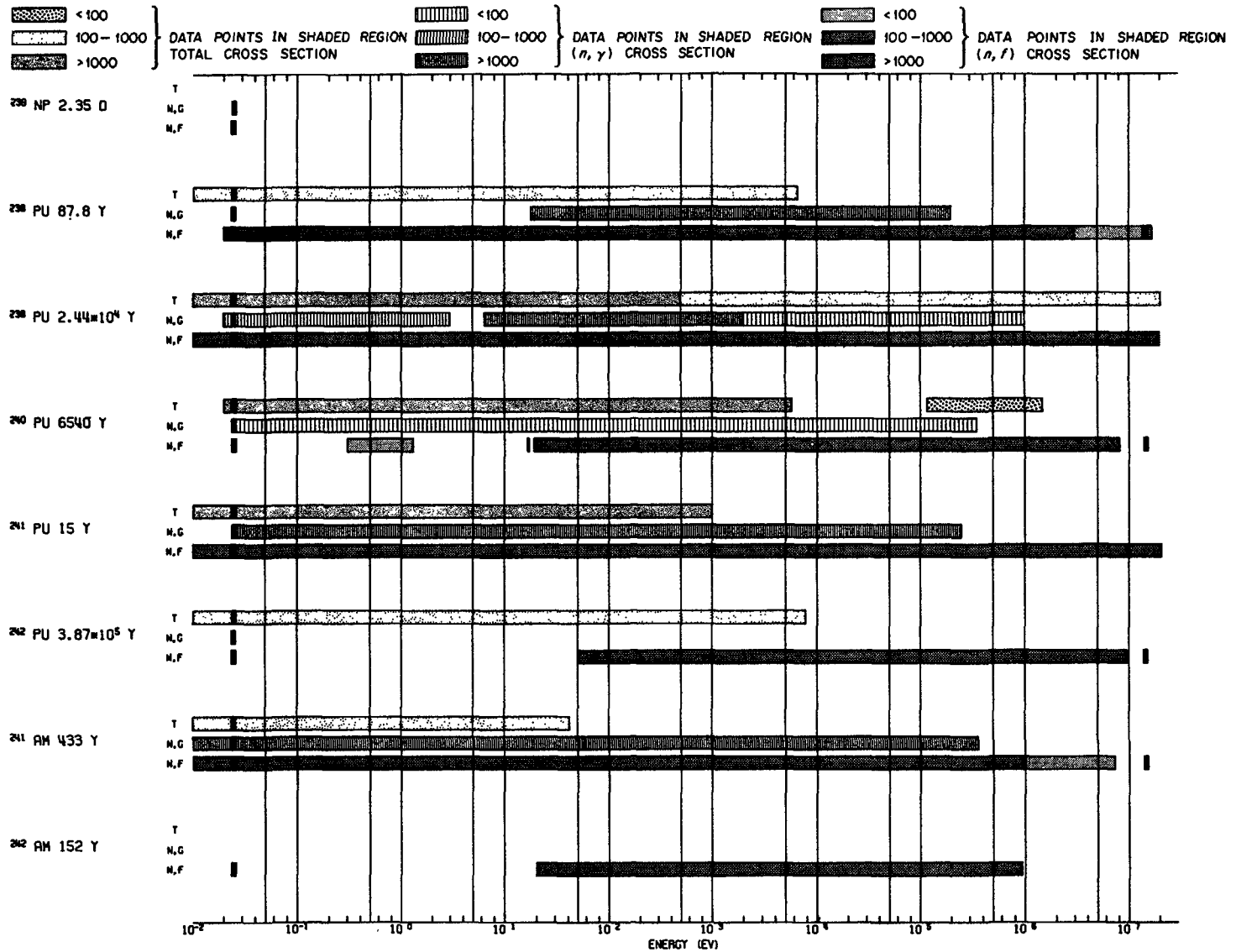


Fig. 3. (continued) Present status of actinide cross sections.

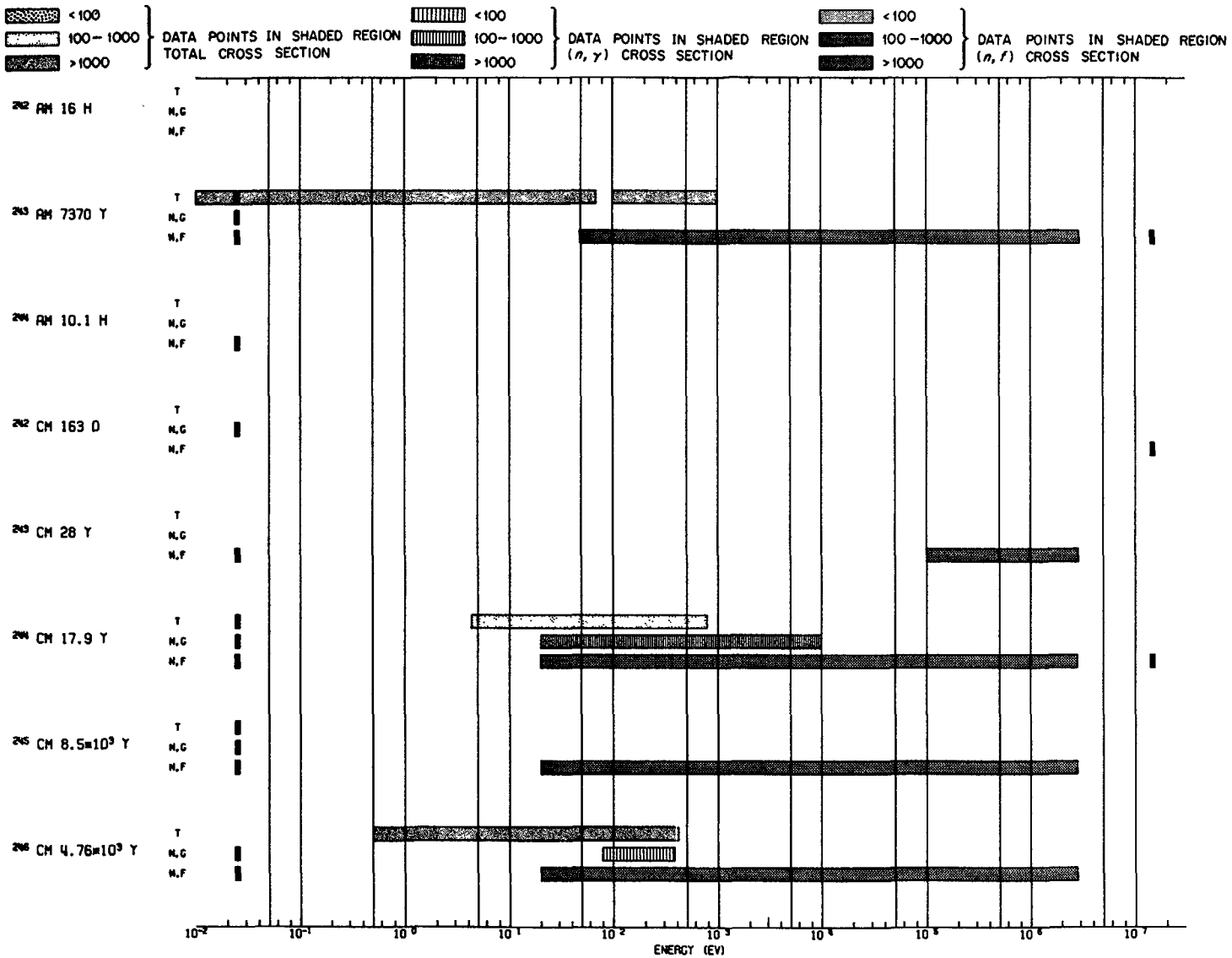


Fig. 3. (continued) Present status of actinide cross sections.

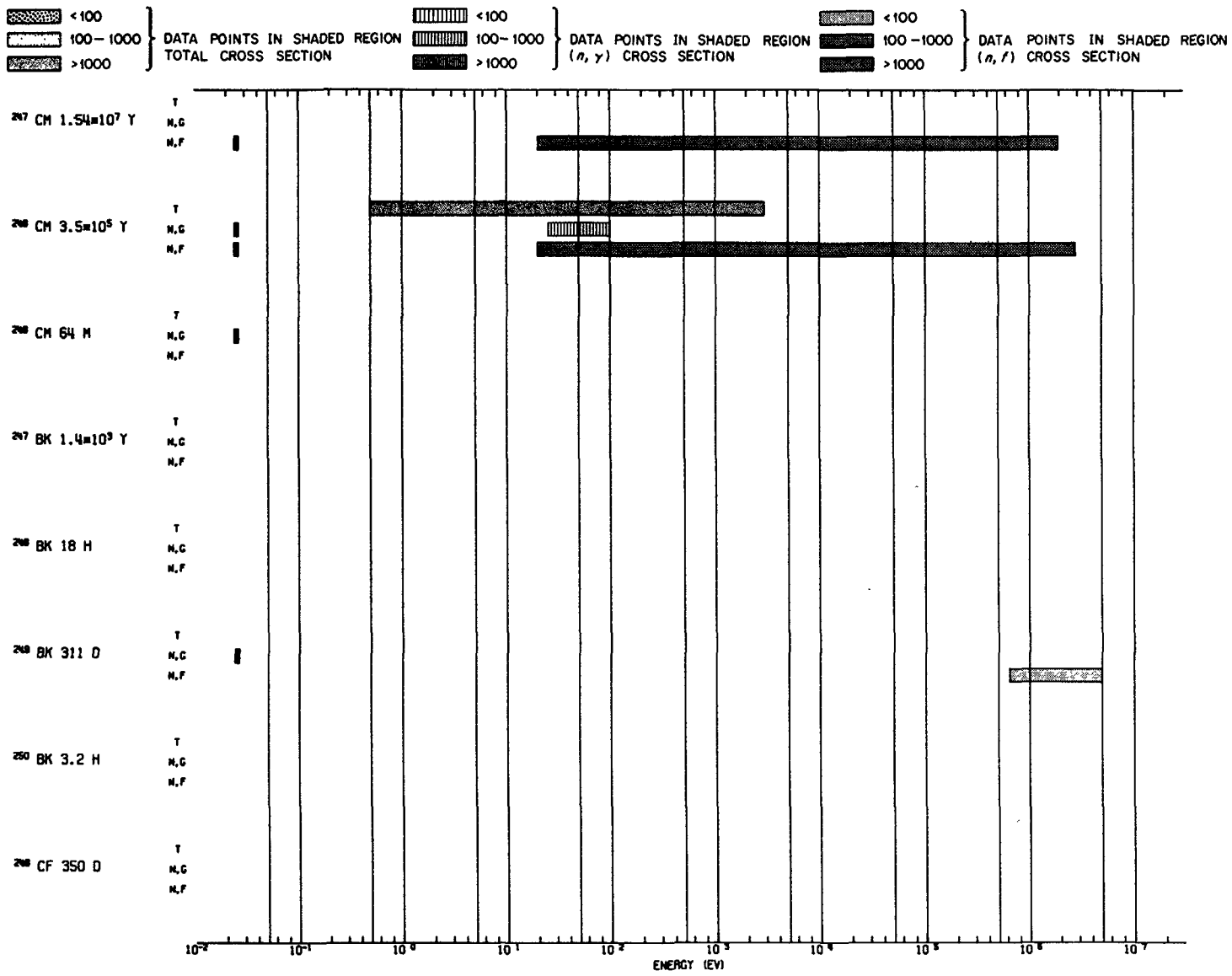


Fig. 3. (continued) Present status of actinide cross sections.

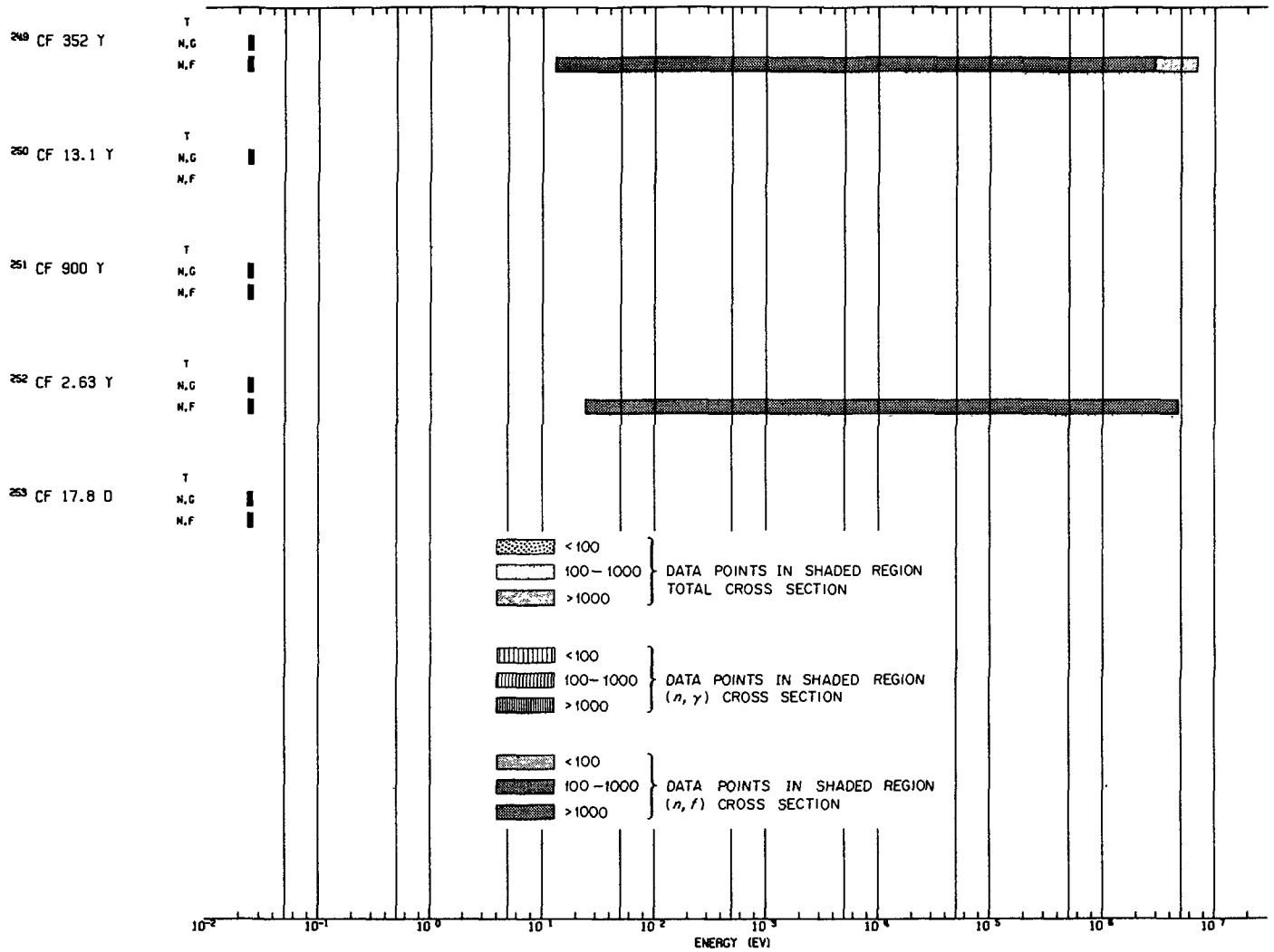


Fig. 3 (continued) Present status of actinide cross sections.

in which cross sections have been obtained as a function of neutron energy, Fig. 3 explicitly shows whether or not cross sections have been measured at thermal and at 14.6-MeV neutron energies.

In Table I, the nuclides are arranged in the order of decreasing occurrence in a thermal reactor. With the aid of this table and Fig. 3, it is relatively straightforward to identify those nuclides for which new cross sections are urgently needed. For example, the existence of data appears to be adequate for the first six nuclides in Table I, until ^{237}Np and ^{242}Pu are reached. For these nuclides, Fig. 3 shows that (n,γ) cross sections are not available. (However, at low energies, σ_γ can be estimated from σ_T.) The next hiatus is again (n, γ) cross section, this time for ^{243}Am. Further down, one finds that ^{242m}Am and the Cm isotopes have not been extensively studied. The deficiencies in capture cross sections are numerous. In addition, the fission cross sections are also lacking below 15 eV for several fissile isotopes of Cm and Cf. Since actinide waste recycling in thermal reactors is a concept under active scrutiny at this juncture and since the elimination of problem actinides depends on our ability to induce them to fission, a knowledge of fission cross sections in the thermal ~20-eV range for ^{242}Am and Cm isotopes has great importance. A list of needed cross sections for actinide recycling studies has been provided at the end of Section III.

III. DATA NEEDS RELATED TO FUEL COMPOSITION

The neutron activity of a metric ton of fresh Pu-enriched LWR fuel is $\sim 6 \times 10^7$ n/sec. This activity in the same fuel after a three-year irradiation increases to $\sim 6 \times 10^9$ n/sec [13]. One of the principal results of recycling actinides would be a further increase in the neutron activity associated with the fuel. The main neutron-active isotope in actinides recycled in thermal reactors is ^{252}Cf. In the case of actinides recycled in an LMFBR, the principal neutron-active isotopes are ^{244}Cm, ^{250}Cf, and ^{252}Cf. The neutron source strength associated with the irradiated fuel is an important factor in the design of shielding and in the determination of the reactivity status of a shut-down reactor. Johnson [14] has recently evaluated the neutron yields from spontaneous fission of transuranium isotopes. The spontaneous fission yield, Y (in neutrons/gram · sec), is related to the nuclear data parameters by the following expression,

$$Y(S.F.) = \bar{\nu}_{S.F.} \lambda_{S.F.} N,$$

where $\bar{\nu}_{S.F.}$ is the average number of neutrons emitted per spontaneous fission, $\lambda_{S.F.}$ is the decay constant for spontaneous fission, and N is the number of atoms per gram of the isotope. The unmeasured $\lambda_{S.F.}$ values were estimated with a correlation developed by Swiatecki [15]. The unmeasured $\bar{\nu}_{S.F.}$ values were estimated through an approxi-

mate correlation between $\bar{\nu}_{S.F.}$ values and a parameter representing the kinetic energy of the fission fragments as discussed by Viola [16]. The neutron yields from spontaneous fission, estimated by Johnson [14], are shown in Table III.

Because it is used as a standard in the measurements of $\bar{\nu}$ for neutron-induced fission, the value of $\bar{\nu}$ for the spontaneous fission of ^{252}Cf must be known with high accuracy, preferably ±0.25%. No reported measurement has reached this accuracy level. The three measurements with the smallest reported uncertainties (±0.4%) yield the following values: 3.713±0.0015 by Colvin and Sowerby [17] with a graphite pile + BF_3 detector, 3.725±0.0015 by de Volpi and Porges [18] with a Mn bath, and 3.738±0.0015 by Boldeman [19] with a Gd-poisoned liquid scintillator. The first and third values differ by 0.7%. The spread in all reported values is about 2.5%. A recent study [20] of the liquid-scintillator tank method of measuring $\bar{\nu}_p$ for ^{252}Cf indicates that there is a neutron energy dependence of the capture gamma-ray detection efficiency for these devices, which in previous measurements was assumed constant. It appears that the inclusion of a correction for this effect would reduce the overall spread in the previously reported values [21]. However, the above study [20] consisted of Monte Carlo calculations of the neutron absorption efficiency and the neutron capture gamma-ray interaction probability while assuming that the third factor in the overall tank neutron efficiency, viz., the light collection efficiency, is independent of the position where the light was created. It is not clear whether this assumption is valid for the several tank configurations (shape of the tank, number of photomultipliers, the size of the through tube, etc.) employed in the previous measurements. It would be desirable to have an experimental determination of the overall detection efficiency of the tank for fission spectrum neutrons. Spencer, Gwin, Ingle, and Weaver [22] at Oak Ridge are planning to carry out such a measurement and a remeasurement of the $\bar{\nu}_p$ for ^{252}Cf utilizing a Gd-poisoned tank. They plan to use a proton recoil detector for the efficiency measurements along with a very small effective through hole in the tank and a well-collimated, 1.25-cm diameter, neutron beam. These measurements are being carried out in support of a broader program of relative $\bar{\nu}$ *vs.* neutron energy determinations on the fissile isotopes with the same tank and the Oak Ridge Electron Linear Accelerator (ORELA) intense neutron source.

Proceeding as before, if we consider both the amounts produced in a reactor (Table I) and the neutron yield (Table III), the isotopes we must contend with are ^{244}Cm, ^{242}Cm, ^{246}Cm, ^{240}Pu, ^{242}Pu, and ^{238}Pu. (It is not a coincidence that these isotopes are also the ones for which neutron yields have been experimentally measured as given in Table III.) With actinide recycling, the list should also include ^{250}Cf and ^{252}Cf. The cross-section data needed to predict the buildup of these nuclides are of special signifi-

TABLE III

Neutron yields from spontaneous fission

Isotope	$T_{1/2}$ (S.F.) years measured	$T_{1/2}$ (S.F.) years estimated	$\bar{\nu}$ (S.F.) measured	$\bar{\nu}$ (S.F.) estimated	Neutron Yield (n/gm.sec) measured	estimated
^{235}U	$(3.5\pm0.9)\times10^{17}$			1.695		2.73×10^{-4}
^{236}U	2.0×10^{16}		1.65±0.12	1.784	4.64×10^{-3}	5.00×10^{-3}
^{237}U		6.3×10^{19}		1.872		1.66×10^{-6}
^{238}U	$(1.01\pm0.03)\times10^{16}$		2.00±0.08	1.960	$(1.10\pm0.06)\times10^{-2}$	1.08×10^{-2}
^{239}U	8.9×10^{18}			2.048		1.27×10^{-5}
^{236g}Np		6.3×10^{17}		1.783		1.59×10^{-4}
^{236m}Np		6.3×10^{17}		1.790		1.59×10^{-4}
^{237}Np	$>10^{18}$			1.873		$<1.05\times10^{-4}$
^{238}Np		2×10^{18}		1.963		5.45×10^{-5}
^{239}Np		6.3×10^{16}		2.053		1.80×10^{-3}
^{236}Pu	3.5×10^{9}		2.22±0.20	1.793	3.55×10^{4}	2.87×10^{4}
^{237}Pu		2×10^{15}		1.886		5.26×10^{-2}
^{238}Pu	$(5.0\pm0.6)\times10^{10}$		2.28±0.08	1.977	$(2.54\pm0.32)\times10^{3}$	2.20×10^{3}
^{239}Pu	5.5×10^{15}		2.24±0.11	2.069	2.26×10^{-2}	2.08×10^{-2}
^{240}Pu	$(1.33\pm0.12)\times10^{11}$		2.16±0.02	2.160	$(9.0\pm0.9)\times10^{2}$	8.95×10^{2}
^{241}Pu		2.5×10^{15}		2.250		4.94×10^{-2}
^{242}Pu	$(7.4\pm0.2)\times10^{10}$		2.15±0.02	2.340	$(1.58\pm0.04)\times10^{3}$	1.72×10^{3}
^{243}Pu		2×10^{15}		2.430		6.61×10^{-2}
^{244}Pu	$(2.5\pm0.8)\times10^{10}$		2.30±0.19	2.518	$(5.0\pm1.7)\times10^{3}$	5.46×10^{3}
^{240}Am		1×10^{15}		2.290		1.26×10^{-1}
^{241}Am	$(2.3\pm0.8)\times10^{14}$			2.383		5.69×10^{-1}
^{242g}Am		1×10^{10}		2.475		1.35×10^{2}
^{242m}Am	$(9.5\pm3.5)\times10^{11}$		2.59±0.10	2.481	$(1.5\pm0.6)\times10^{2}$	1.43×10^{2}
^{243}Am	$(3.3\pm0.3)\times10^{13}$		2.52±0.11	2.566	4.2±0.5	4.23
^{244g}Am		7.9×10^{13}		2.657		1.82
^{244m}Am		7.9×10^{13}		2.665		1.83
^{240}Cm	1.9×10^{6}			2.406		6.98×10^{7}
^{241}Cm		1.6×10^{12}		2.500		8.58×10^{1}
^{242}Cm	7.2×10^{6}		2.59±0.09	2.594	1.97×10^{7}	1.97×10^{7}
^{243}Cm		1.2×10^{11}		2.687		1.22×10^{3}
^{244}Cm	$(1.30\pm0.07)\times10^{7}$		2.76±0.07	2.780	$(1.15\pm0.07)\times10^{7}$	1.16×10^{7}
^{245}Cm		4.0×10^{12}		2.872		3.88×10^{1}
^{246}Cm	$(1.80\pm0.01)\times10^{7}$		3.0±0.2	2.964	$(9.0\pm0.7)\times10^{6}$	8.85×10^{6}

cance. In addition to the usual (n,f), (n,γ), and total cross-section data, (n,2n) and (n, 3n) cross sections might be important in this instance. In a few cases where metastable states are involved, one might also need (n, n') cross sections, e.g., $^{242m}Am(n,n')^{242g}Am \xrightarrow{\beta} {}^{242}Cm$, which is an exoenergetic reaction.

Several cross sections are needed to estimate the production of ^{232}U and ^{236}Pu in irradiated fuel elements. These isotopes are objectionable because they produce radioactive daughters, some of which emit penetrating gamma radiation requiring heavy shielding. Another precursor of toxic isotopes is ^{231}Pa produced via the $^{232}Th(n,2n)^{231}Th \xrightarrow{\beta^-} {}^{231}Pa$ channel. A knowledge of the $^{232}Th(n,3n)$ cross section leading to ^{230}Th is also important since ^{230}Th and its daughters are extremely toxic.

TABLE IV

Needed cross sections in the actinide region

Target	σ (type)	Why needed and other comments
^{230}Th	(n, γ)	To calculate production rate of ^{232}U
^{232}Th	(n, 3n)	To calculate production rate of ^{230}Th
^{231}Pa	(n, γ)	To calculate production rate of ^{232}U
^{233}Pa	(n, γ); total	Actinide recycle (Priority II) ^{233}U–^{232}Th thermal breeder
^{232}U	(n, γ)	To calculate production rate of ^{232}U
^{233}U	(n, 2n)	To calculate production rate of ^{232}U
^{237}Np	(n, γ); (n,2n); (γ, n)	Actinide recycle (Priority I) To calculate production rate of ^{236}Pu
^{236}Pu	(n, γ); (n, f); total	To calculate production rate of ^{236}Pu
^{238}Pu	(n, 3n)	To calculate production rate of ^{236}Pu
^{239}Pu	(n, 2n)	To calculate production rate of ^{238}Pu
^{242}Pu	(n, γ)	Actinide recycle (Priority I)
^{241}Am	(n, γ)	Capture branching ratios to the ground and isomeric states of ^{242}Am needed to estimate production of ^{242m}Am and ^{242}Cm
^{242g}Am	(n, f); (n, γ); total	Actinide recycle (Priority II)
^{242m}Am	(n, f); (n, γ); total	Actinide recycle (Priority II) Thermal σ(n, γ) value, 1400 ± 860 b, has 60% uncertainty
^{243}Am	(n, γ); (n,2n)	Actinide recycle (Priority I) To calculate production rate of ^{238}Pu

(continued on next page)

Table IV lists some of the additional cross sections needed for actinide recycling studies and for estimating fuel composition. It is recognized that direct measurements of some of these cross sections might be impractical for various reasons. In such cases, it might be necessary to employ neutron cross sections extracted from indirect charged-particle measurements [4] or obtained from nuclear model calculations [23] and systematics.

TABLE IV (continued)

Target	σ (type)	Why needed and other comments
^{242}Cm		Actinide recycle (Priority II)
^{243}Cm		Actinide recycle (Priority II)
^{244}Cm		Actinide recycle (Priority II)
^{245}Cm	(n, f); (n, γ); total	Actinide recycle (Priority I)
^{246}Cm		
^{247}Cm		
^{248}Cm		
^{249}Cm		
^{249}Bk		
^{250}Bk		
^{249}Cf		
^{250}Cf		
^{251}Cf		
^{252}Cf		
^{253}Cf		Actinide recycle (Priority II)
^{254}Cf		Actinide recycle (Priority II)

IV. THE ^{252}Cf BUILDUP AND RELATED PROBLEMS

One gram of ^{252}Cf emits 2.34×10^{12} n/sec. A rapid increase in the ^{252}Cf buildup is a corollary to all actinide recycling schemes involving fission reactors. This buildup places severe demands on the design of shielding for facilities involved in the handling, storing, transporting, and processing of irradiated fuel. The production path for ^{252}Cf is well-illustrated in Fig. 4 prepared by Crandall [24]. Approximately 1800 neutrons and 1000 atoms of ^{238}U are required to produce one atom of ^{252}Cf in a typical thermal reactor. In order to estimate the amount of ^{252}Cf produced in the United States between now and the year 2000, we follow Crandall's arguments. The present predictions are that reactors would generate $\sim 10^{10}$ megawatt-days of heat in the above time interval. The total amount of Pu generated (without breeders) would be ~5000 metric tons on the basis of 0.5 grams of Pu for each megawatt-day. If all of the Pu is recycled, ~7% will transmute to Am and Cm, resulting in ~360 metric tons of these elements. The burnup of Am and Cm will ultimately result in the production of ~5 metric tons of ^{252}Cf over a period of 25 years! Fortunately, the half-life of ^{252}Cf is only 2.6 yr.

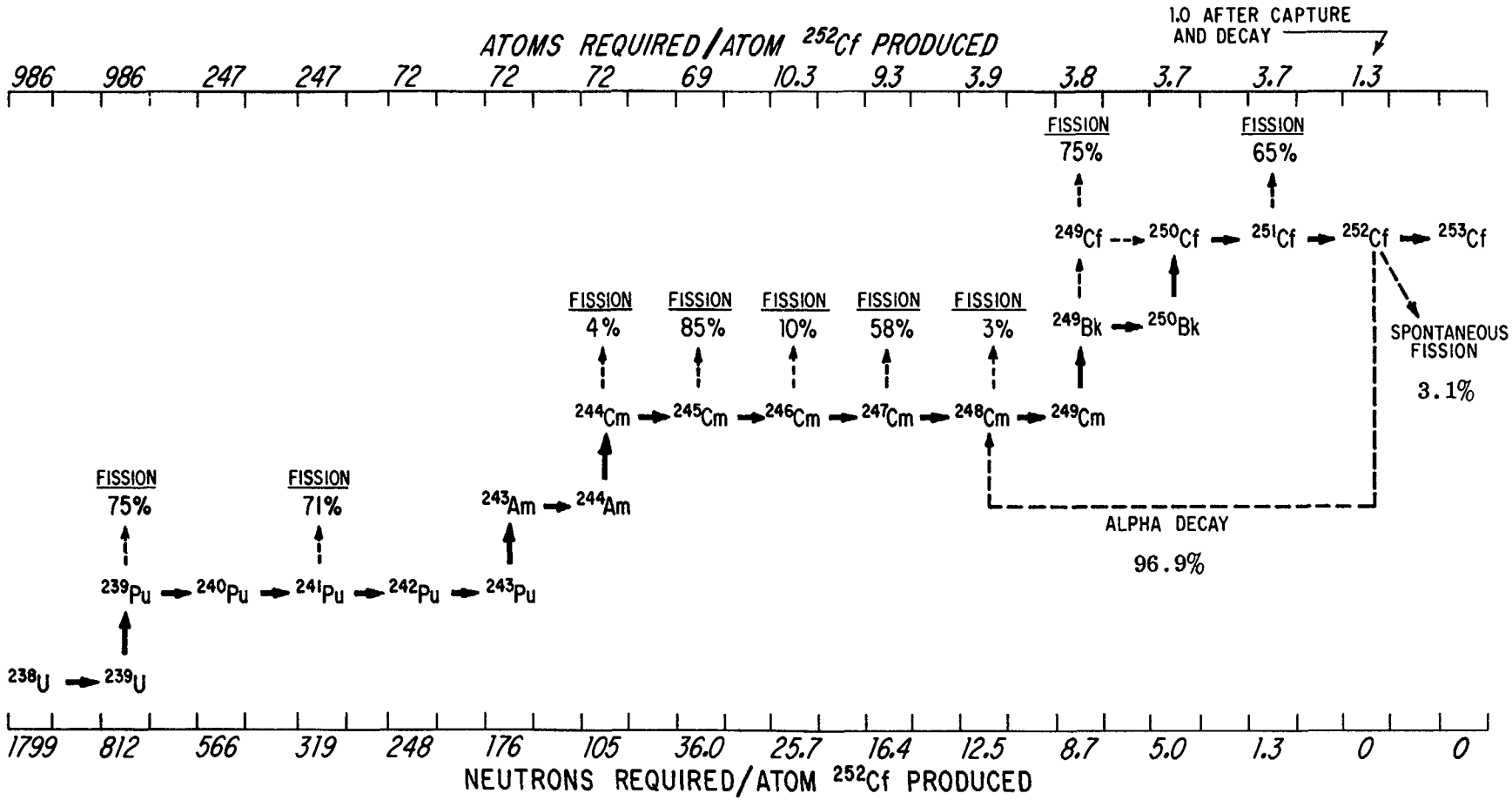


Fig. 4. Production path for ^{252}Cf.

In this context, the experience gained at the Savannah River reactors and more recently at the ORNL-HFIR in producing gram quantities of ^{252}Cf for its multifarious applications should be extremely valuable. The data set shown in Table II has yielded fairly satisfactory results in predicting the contents of irradiated HFIR target rods, usually within 10% for most nuclides up to ^{253}Es. However, there still remain some interesting problems and puzzles as outlined below.*

One region which was originally ill-defined but now better understood is the ^{252}Cf–^{253}Cf–^{254}Cf region. The first isotope, ^{252}Cf, is the major transcurium product of the transmutation chain. Its relatively small capture cross section causes it to have relatively high abundance in the irradiated targets. Its short half-life (2.6 yr) results in very high specific activities. The high yield of spontaneous fission (3%) makes ^{252}Cf the major radiation hazard. Another hazard is ^{254}Cf which is produced in much smaller yield but has 500 times the specific neutron emission rate of ^{252}Cf. Extrapolations of the ^{254}Cf abundance in the HFIR product, based on early cross-section data, suggested that this nuclide might produce neutron activity equal to that of ^{252}Cf, thus doubling the hazard. However, actual experience has shown the maximum yield of ^{254}Cf in HFIR production is such that it produces only 30% as much neutron radiation as the ^{252}Cf in the same sample. In working out these apparent discrepancies, Bigelow [25] found not only that ^{252}Cf is slightly fissionable with thermal neutrons, but also that ^{253}Cf has a large thermal fission cross section. Thus, in the HFIR, over 90% of the ^{253}Cf atoms fission before they can decay or capture a neutron to produce ^{254}Cf. This fortunate circumstance might not occur in a high-flux, high neutron energy spectrum reactor like an LMFBR. If such a device were used as an actinide burner, the yield of ^{254}Cf might be higher, and thus the neutron dose from a unit of Cf at discharge might be several times what it is in HFIR californium.

Another problem that had to be solved in the HFIR calculations was that of resonance self-shielding. The density of several of the nuclides in the target rods can be fairly high (on the order of 2×10^{21} nuclei/cm^3), and, considering the strong absorption resonances, there can be appreciable self-shielding. While a single-level Breit-Wigner formalism, with a semi-empirical constant, gave fairly good results for ^{242}Pu and ^{243}Am burnout curves, this procedure has not been successful for ^{244}Cm probably due to resonance flux depression caused by ^{244}Cm in adjoining target rods. This is an effect which could also be important in actinide burner calculations.

A final example that might be of interest is the case of the missing ^{255}Es. This neutron-rich β-emitting isotope would be a good target for producing new super-heavy

*Notes for the remainder of Section IV were provided by Bigelow [25].

elements provided it could be produced in multimicrogram amounts. Unfortunately, the yield of this isotope is minuscule; the highest yield in the HFIR is only about 0.07% of the total of all einsteinium isotopes at discharge.

There are three routes for producing ^{255}Es: neutron capture in 39-h ^{254m}Es, neutron capture in 276-d ^{254}Es, and neutron capture in 60-d ^{254}Cf followed by β-decay of the short-lived ^{255}Cf. Absorption cross sections for these three nuclides can be estimated fairly well from observed burnout. If a reasonable value is assumed for the capture-to-fission ratio for any of these nuclides, the amount of ^{255}Es produced ought to be 10-100 times what is actually observed. Instead, the apparent capture cross sections are small: σ_c = 1.2 b *vs.* σ_a = 1800 b for ^{254m}Es, σ_c = 20 b *vs.* σ_a = 3000 b for ^{254}Es, and σ_c = 1 b *vs.* σ_a = 150 b for ^{254}Cf. More investigations will be required to elucidate these unusual relationships. Another possible explanation is that normal cross sections apply, but that they lead to an isomeric state of ^{255}Es which decays in a relatively short time (hours or less) by β-emission or spontaneous fission. Whatever the explanation, it may also apply in the case of ^{250}Cm which too is produced in unusually small amounts.

V. RADIOISOTOPE POWER SOURCES

To date, the use of actinides for power sources has been restricted almost exclusively to ^{238}Pu-fueled units, although the original "atomic battery" used in space was fueled with ^{210}Po. A considerable amount of development work has been done with ^{244}Cm and some with ^{242}Cm, but no operational sources have been put into service with these isotopes. The largest use for ^{238}Pu-fueled sources, in terms of amount of ^{238}Pu, is in thermoelectric generators to provide electrical power in space applications. Units of this type have been used in satellites, on the moon, and in space probes. Another application, requiring less ^{238}Pu but involving more devices, is in heart pacer power units.

Possible uses of ^{238}Pu and ^{244}Cm include additional outer space applications, terrestrial applications requiring reliable power in remote areas, oceanographic applications of various types, and others. Most applications which have been seriously considered involve the generation of electrical power from the decay heat of the isotope, but there are many possible applications in which the thermal energy would be used directly. For the general types of uses mentioned, ^{238}Pu and ^{244}Cm can be considered as competitors; each has some advantages and some disadvantages, compared to the other. One use which is probably restricted to ^{238}Pu is as a power source in an artificial heart. The potential requirements for radioisotope power sources, costs, and

availability have been the subject of numerous studies. The results of these studies vary considerably, but most of them indicate annual requirements in the 10–100-KW(th) range.

There are two different ways of producing ^{238}Pu. The direct way is by irradiating separated ^{237}Np with neutrons to produce ^{238}Np which β-decays to ^{238}Pu (see Fig. 1). Since ^{237}Np target material will become available as a by-product of U fuel reprocessing, there will be no dearth of feed material. The major problem with this approach is that the (n, 2n) and (γ, n) reactions on ^{237}Np lead to ^{236}Np and thence to ^{236}Pu, which is an undesirable material because of the decay chain it initiates. Both of these reactions have a 6.6-MeV threshold and can be initiated only by the high-energy fission neutron tail and by the capture γ-rays from structural elements (Al and Fe), respectively. The desirability of obtaining better cross sections for the (n, 2n) and (γ, n) reactions will be discussed in another review paper [7].

An alternate way of producing ^{238}Pu is through the irradiation of ^{241}Am with neutrons to produce ^{242}Am followed by β-decay to ^{242}Cm and α-decay to ^{238}Pu (see Fig. 1). This approach produces ^{238}Pu which is essentially free from ^{236}Pu contamination. The ^{241}Am source material is a byproduct of Pu fuel reprocessing and will be available in less quantity than ^{237}Np, but adequate to meet expected requirements. The source material, however, will probably also contain ^{243}Am so that any subsequent irradiation will result in a product consisting of both ^{238}Pu and ^{240}Pu (resulting from α-decay of ^{244}Cm) which might be inconsequential in some applications but not in others. Fortunately, the ^{241}Am has a much higher thermal cross section.

Nuclear data applicable to both of the above methods for producing ^{238}Pu are reasonably well-established. Minor uncertainties exist in the choice of the branching ratio for neutron capture in ^{241}Am to the ground state and isomeric state of ^{242}Am, but these are not crucial problems.

The production of ^{244}Cm is also well-understood in terms of available cross-section data. The feed material in this instance might be ^{243}Am or intermediate Pu isotopes. There should be no shortage of ^{243}Am in the future; indeed, the problem may be how to dispose of the unwanted surplus amounts.

VI. RECENT ACTINIDE CROSS-SECTION MEASUREMENTS AT THE OAK RIDGE ELECTRON LINEAR ACCELERATOR (ORELA)

While the underground nuclear explosion technique has provided fission cross sections for several nuclides, the capture cross sections are not nearly as well-known. At ORELA, Weston and Todd [26] have recently carried out (n, γ) measurements on ^{240}Pu, ^{241}Pu, and ^{241}Am. Figure 5 shows the average absorption cross section of

^{241}Am. The cross section is predominantly capture since fission is small at these neutron energies. The ENDF/B-IV, MAT 1056 evaluation is also shown in Fig. 5. The ENDF evaluation at keV neutron energies is based almost entirely on resonance parameters from total cross sections in the resolved resonance region. The data are appreciably higher than the ENDF evaluation above 20 keV and lower than the ENDF evaluation below 100 eV. Actinide buildup and burnout calculations carried out until now have employed the ENDF evaluation and have, therefore, overestimated the ^{242}Am and ^{242}Cm production in a thermal reactor and underestimated the production in a fast reactor.

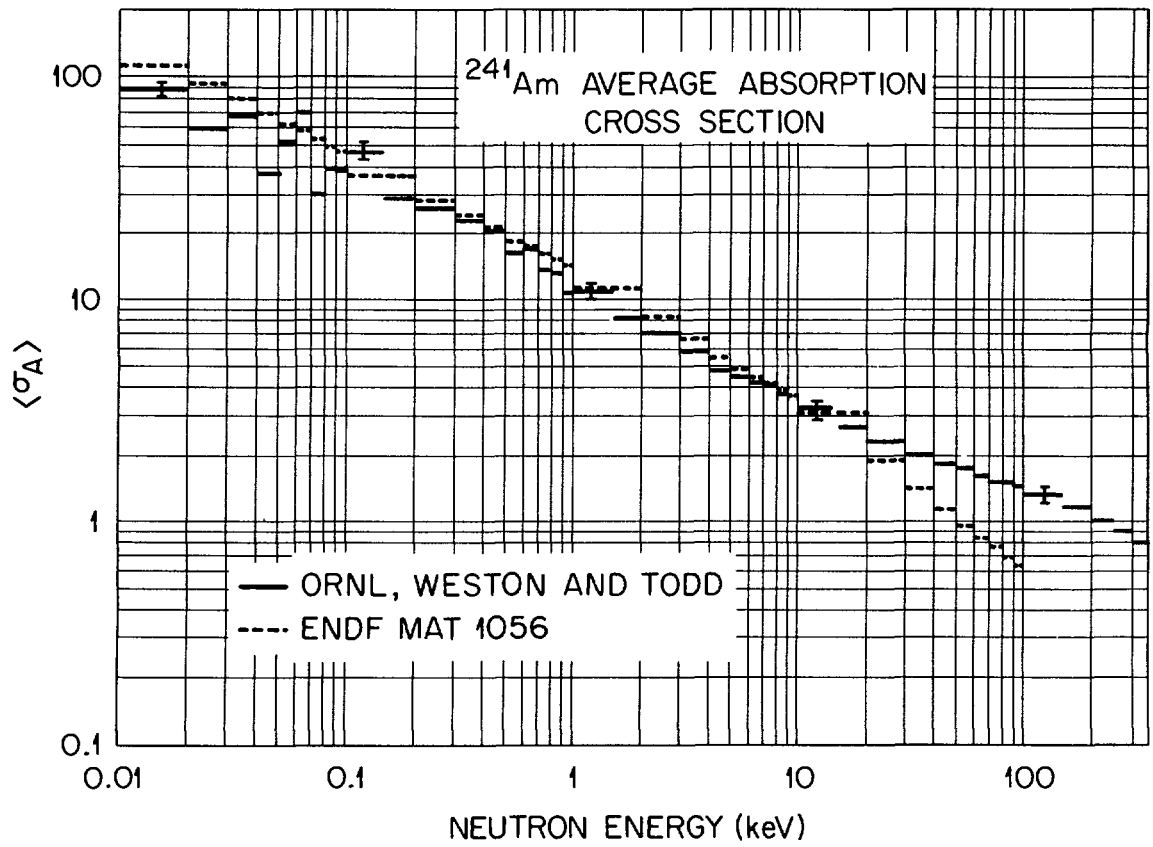


Fig. 5. Absorption cross section of ^{241}Am measured at ORELA

Since ^{242}Cm is a major spontaneous fission neutron source in used fuel elements, the ability to correctly predict the ^{242}Cm content would be important in fuel handling. The above case exemplifies the need for reliable cross-section measurements.

Fission cross-section measurements can be severely hampered by the presence of intense alpha activity or even by moderate amounts of spontaneous fission activity. In the latter case, underground nuclear explosions can provide a solution, but with the well-known limitation that no neutrons below 20 eV are available. With typical samples of short-lived alpha emitters (for example, 60 μg of 30-yr ^{243}Cm), one must deal with

pulse-pileup problems resulting from ~10^8 alpha pulses per second. In order to carry out fission cross-section measurements on such samples, a new spherical plate chamber has been recently developed at ORNL [27]. Measurements have been completed with a 12-μg sample of 99.96% enriched ^{245}Cm ($T_{1/2}$ = 8500 yr) on a 9.5-meter flight path at ORELA. The accelerator burst width was ~40 nsec, and 800 pulses per second (pps) were used. The spherical plate chamber operates on 1.3 atm of CH_4 gas. The chamber plates have radii of 8 and 10 mm. The sharp curvature of the outer plate restricts the maximum α-particle track length and leads to a 15:1 discrimination in pulse height between minimum fission and maximum alpha events. The pulse collection time is 25 nsec. Figure 6 shows a sample of the raw time-of-flight data below 130 eV. Ten

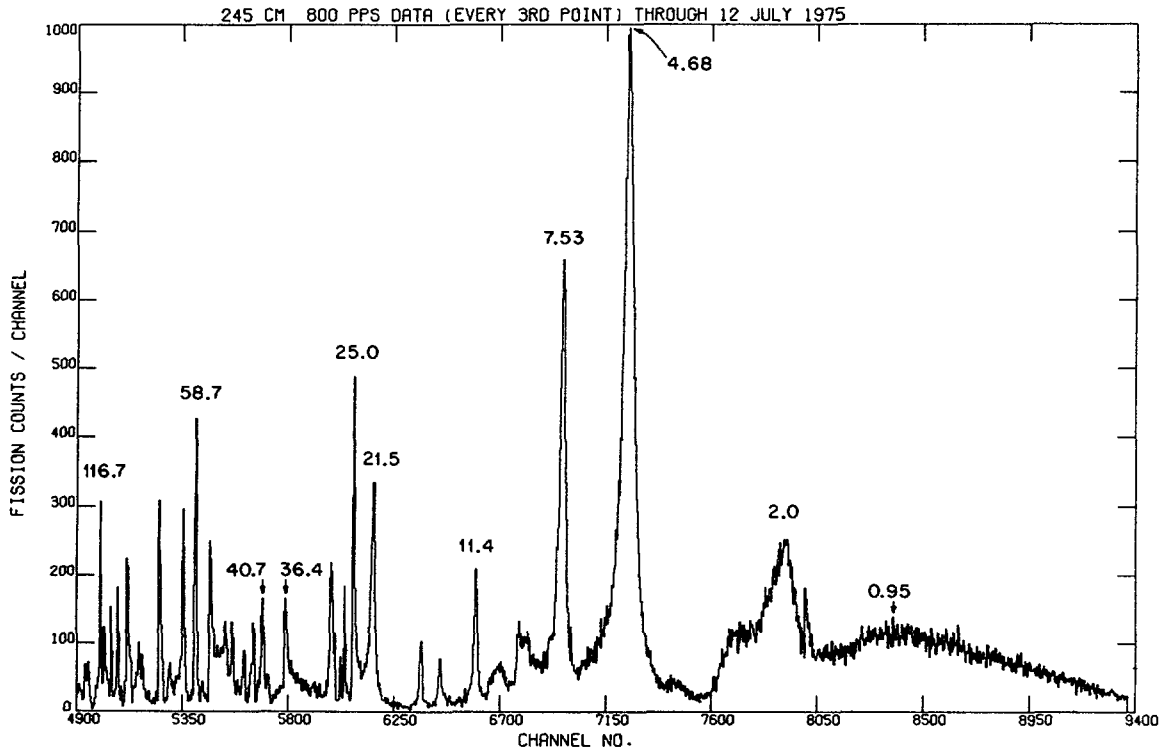


Fig. 6. Raw data from fission measurements on ^{245}Cm at ORELA. With the spectrum above channel number 8001 as the basis, the spectrum to the left should be multiplied by two at channel numbers 8000, 7000, 6000, and 5000. The resonance energies are in eV.

resonances below 20 eV (channel 6200) were observed in fission for the first time. The low-energy end is depressed by a Cd cutoff filter; other data were obtained down to thermal neutron energies, primarily for normalization purposes.

Neutron transmission measurements have been made recently upon two samples of ^{249}Bk at ORELA [28]. The 5.3-mg "thick" sample and the ≈0.8-mg "thin" sample were cooled with liquid nitrogen to reduce the Doppler broadening. Measurements were made through the samples using an 11.0-cm diameter, 1.3-cm thick, ^{6}Li glass scintillator neutron detector located at a 17.87-meter flight path. Typical results are shown in Fig. 7. A total of 47 resonances below ~130 eV were observed. A large resonance observed at 0.197 eV (not shown in Fig. 7) is responsible for most of the thermal absorption cross section which departs from a 1/v energy dependence. The average level spacing

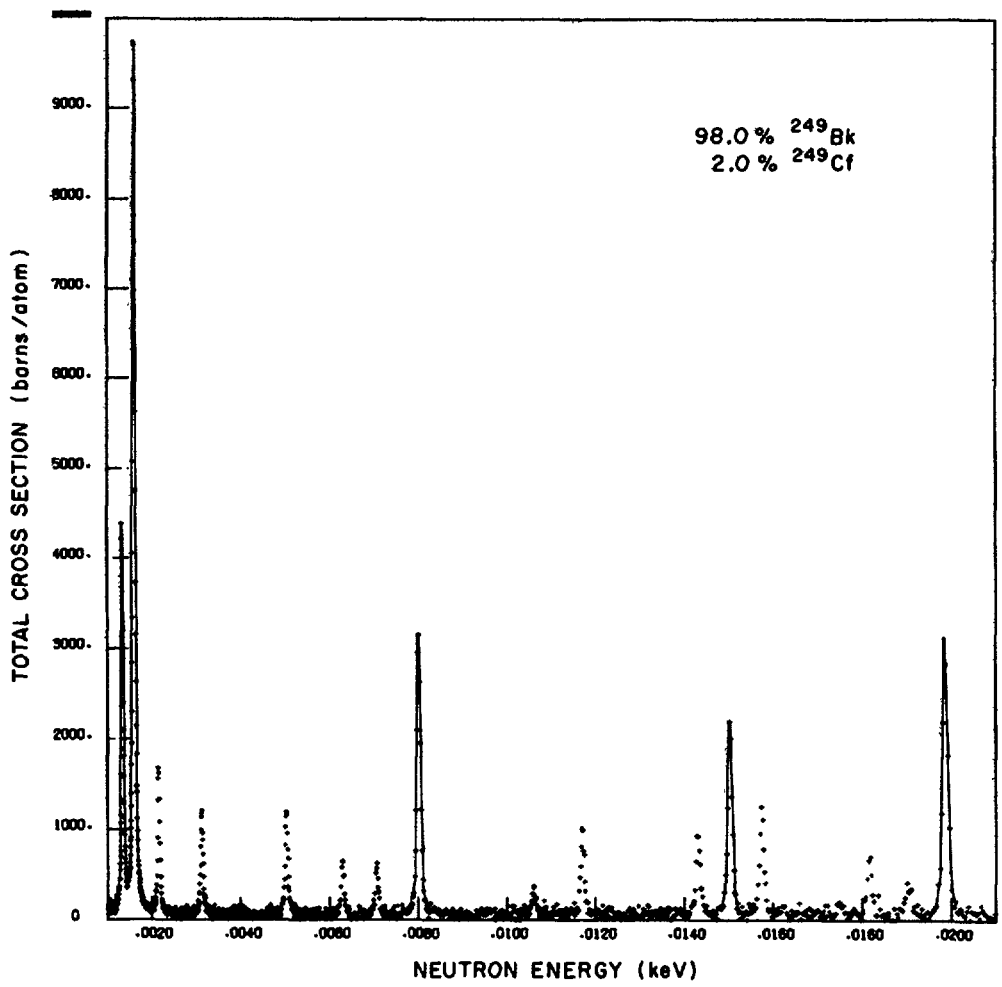


Fig. 7. Preliminary results in the 1–21-eV range of total cross-section measurements of ^{249}Bk at ORELA.

based on the resonances up to 20 eV is 1.1 eV. The large resonance in ^{249}Cf at 0.70 eV was also observed even though there was only ~2% ^{249}Cf in the sample at the time of the measurement. Additional measurements will be made after about half of the 311-d ^{249}Bk has decayed into ^{249}Cf in order to obtain parameters for the resonances in ^{249}Cf

as well as the thermal absorption cross section. These measurements are part of a cooperative program between Savannah River Laboratory and Oak Ridge National Laboratory to determine the heavy-actinide cross sections for more accurate calculation of ^{252}Cf production.

VII. PROBLEMS IN PREPARING TRANSACTINIUM SAMPLES FOR CROSS-SECTION MEASUREMENTS

The availability of high-purity samples in sufficient quantities is the controlling factor in any extensive actinide cross-section measurement program. The basic capability for preparing many actinide samples already exists at the Oak Ridge National Laboratory (ORNL) as part of the Transuranium Element Program (TRU) and the High Flux Isotope Reactor (HFIR), supplemented by the Transuranium Element Research Program (TRL), the Oak Ridge Research Reactor (ORR), and the Y-12 calutron facility. Nevertheless, in many cases, there are great difficulties to be overcome, as discussed below.*

^{227}Ac — Available in gram amounts from Union Minière de Belgique. Produced by irradiation of ^{226}Ra. Hazardous to handle because of radon emanation.

^{228}Th — Same as above (except in multi-milligram amounts). Also, several milligrams could be milked from the 750-mg stock of ^{232}U being held at ORNL. Hazard increased due to rapid growth of ^{208}Tl. Experimental difficulties great.

^{229}Th — ORNL is presently saving some ^{229}Th recovered from aged ^{233}U. Total might be as much as 1-2 g. However, the concentration is only a few percent in the presence of much ^{232}Th, and isotope separation would be required for suitable cross-section work, thus reducing the availability by an order of magnitude. The presence of contaminant ^{228}Th might rule out the possibility of using the calutrons for this purpose.

^{230}Th* — This isotope is a member of the ^{238}U decay chain and can be recovered from uranium ores. ORNL is currently processing some ^{230}Th through the calutrons. More material is expected to become available (see footnote below).

^{232}Th — Available.

*Notes for the remainder of Section VII were provided by Bigelow [25].

^{231}Pa* — This isotope is also a naturally occurring radioactive element. Recovery is extremely expensive. ORNL is holding a stock of 2.5 g. More material is expected to become available (see footnote below).

^{233}Pa — Can be produced by irradiation of ^{232}Th. A very important isotope to study, but experimental difficulties would be extreme.

^{232}U — ORNL is holding a stock of approximately 750 mg. Hard to handle. Further production entails irradiation of ^{231}Pa.

^{233}U — Available; 850 mg at 99.99% isotopic purity.

^{234}U — Available; 5 g at 99.1% isotopic purity.

^{235}U — Available; 2.5 g at 99.91% isotopic purity.

^{236}U — Available; 1 g at 89.2% isotopic purity.

^{237}U — Not available because of extreme experimental difficulties.

^{238}U — Available; > 10 g at 99.999% isotopic purity.

^{237}Np — Available; > 1 g at > 97% radiochemical purity.

^{238}Np — Not available because of extreme experimental difficulties.

^{239}Np — Could probably be separated in multi-milligram amounts from irradiated ^{238}U. Rapid growth of ^{239}Pu in sample would complicate measurements enormously.

^{238}Pu — Available; > 1 g at 97.4% isotopic purity.

^{239}Pu — Available; > 1 g at > 99.9% isotopic purity.

^{240}Pu — Available; > 1 g at 98.3% isotopic purity.

^{241}Pu — Available; 1.5 g at 99.7% isotopic purity.

^{242}Pu — Available; > 1 g at 99.8% isotopic purity.

*For the *aficionados*, the story on protactinium-231 and thorium-230 is as follows: Beginning in 1942, the Mallinckrodt Chemical Company processed Congo and native ores by extracting the uranium from a nitric acid feed solution with diethyl ether. After removal of the radium as a sulfate and the excess sulfuric acid as barium sulfate, the filtered raffinate was limed, and the limed cake became the St. Louis Airport Residues. The Cotter Corporation of Golden, Colorado, acquired these residues and began processing them in 1968 for uranium and other valuable metals. When the process was altered in 1971 to recover protactinium-231 and thorium-230, only 17,000 tons remained of the original 41,000 tons of residues. In all, approximately 80 grams of protactinium-231 and 14.5 kilograms of thorium-230 were collected in about 200 tons of material. A pilot plant scale recovery and purification system is in operation at Mound Laboratory which, when operating at capacity, will produce approximately 180 grams of thorium-230 and one gram of protactinium-231 per year. Both isotopes will be distributed from the Oak Ridge National Laboratory. [The author is indebted to R. M. Watrous of the Mound Laboratory, Monsanto Research Corporation, Miamisburg, Ohio, for providing these historical notes.]

^{243}Pu — Not available; $T_{1/2}$ = 5 hr.

^{244}Pu — Produced as by-product of Cf-I program at Savannah River and calutron enriched to 98-99%. About 3 g have been produced to date. Several more grams may become available in next few years.

^{241}Am — Available; > 10 g at > 97% radiochemical purity.

^{242m}Am — Multi-milligram amounts have been produced at 20% enrichment in the calutrons. Better material would require difficult irradiation of ^{241}Am in the ORR using cadmium-shielded targets, followed by chemical processing and calutron separations.

^{243}Am — Readily available, but special processing required to remove neutron-emitting contaminants.

^{242}Cm — Multi-gram quantities can be produced by irradiation of ^{241}Am in the ORR. Experimental difficulties with high specific activity product.

^{243}Cm — An existing stock of 60 mg (55% ^{243}Cm, 45% ^{244}Cm) was prepared by irradiating ^{241}Am, followed by long decay time to remove ^{242}Cm. Multi-gram amounts could be produced by this route, but long decay time plus high gamma radiation make the prospects dim.

^{244}Cm — Available at 95% isotopic purity. A 40-mg sample at > 99% purity exists.

^{245}Cm — Like ^{242m}Am, a difficult irradiation in a Cd filter would be required to produce a ^{245}Cm concentrate feed for isotope separation. Perhaps milligram amounts could be produced in laboratory isotope separators, but gram amounts cannot be produced until a doubly contained calutron facility is constructed. Later on, power reactor curium would be a good feed stock for separation of ^{245}Cm.

^{246}Cm — Presently available in gram amounts at 60% isotopic purity from HFIR irradiations. See comments above regarding isotope separation of ^{245}Cm.

^{247}Cm — Like ^{245}Cm, only slightly more difficult irradiation.

^{248}Cm — Available in multi-milligram amounts at 97% isotopic purity from decay of ^{252}Cf. Supply in 1976 may reach 100 mg. In 1980, may be 1 g.

^{249}Cm — Not available; $T_{1/2}$ = 1 hr.

^{250}Cm — Available only in picogram to nanogram quantities.

^{249}Bk — Available in multi-milligram amounts, isotopically pure, as a by-product of ^{252}Cf production. Because of short half-life, maximum amount that could be accumulated in foreseeable future is 100 mg.

^{249}Cf — From decay of ^{249}Bk. Probably could reach 500 mg in a decade. A valuable isotope to study chemistry of californium.

^{250}Cf — HFIR has produced 1 mg by irradiation of ^{249}Bk, at isotopic purity of 89%. Possible to produce 10 mg in this manner, but a batch of 100 mg is nearly inconceivable.

^{251}Cf — Maximum isotopic concentration that can be produced in HFIR is 15%. This could possibly be doubled by irradiation of ^{250}Cf in Cd-shielded position in ORR. Isotope separation capabilities much restricted by neutron radiation hazard from ^{250}Cf and ^{252}Cf.

^{252}Cf — Possible to produce a 1 g lot of ^{252}Cf at 98% purity. Cross-section measurements would be very difficult on such a spontaneous fission neutron source (2×10^{12} n/sec).

^{253}Cf — Can only be produced by irradiation of ^{252}Cf. Maximum isotopic concentration produced in HFIR irradiations is 1.3%.

^{254}Cf — Same as ^{253}Cf, except maximum concentration is about 0.06%.

VIII. NUCLEAR DECAY DATA NEEDS IN RADIOLOGICAL SAFETY AND HEALTH PHYSICS

In the areas of radiological safety and health physics, the needs for nuclear decay data on the actinide elements can be divided into two categories: data for measurement of body burden and for estimation of internal dose. For the first category, the needs are limited to the abundances of penetrating photons, but the accuracy should be relatively high, $\leq 5\%$. For estimation of internal dose, the details of the entire decay schemes are needed, but to a much lower accuracy, perhaps $\pm 15\%$. The details should include quantities which are not usually measured, e.g., average β-energies, Auger electron yields, X-ray yields, etc. If a particular nuclide is the progenitor of a number of daughters, the entire decay chain must be known and taken into consideration in the calculation of dose.

In vivo measurements of body burden depend on the detection of X-rays and γ-rays from the parent + daughters. In practice, since X-rays are far more common than γ-rays in the case of the actinides, a knowledge of the K and L X-ray intensities is extremely important for relating the measurements to the amounts of actinides retained in the body. If the energies and intensities of the different members of the L X-ray multiplet are known [29] and if the intensities are measured with a high-resolution detector after passage through the tissues, it should be possible to estimate not only the amount

but also the depth of the deposit through recourse to the photoelectric absorption coefficients (which are highly dependent on energy).

Since many actinides emit photons only in the low-energy (strongly absorbed) region, the estimation of body burden in deep tissues often must be based on excretion analysis. Chemical analyses can be tedious, particularly those involving neighboring actinides with similar chemistry. In such cases, the measurement of α-spectra could lead to the identification of the actinides.

Internal dose calculations require calculations of average β-energies, Auger electron yields, and X-ray yields, all of which depend on the details of the decay scheme. These data, and also the (measured) α-, β-, and γ-energies and their abundances, permit calculation of a parameter termed the "equilibrium dose constant". This parameter represents the equilibrium dose rate for each type of radiation per unit concentration of the radionuclide, assuming uniform distribution through an infinite absorbing medium. It serves as a starting point for estimating organ doses. For many isotopes of interest in nuclear medicine, the equilibrium dose constants have been given by Martin and Blichert-Toft [30] and by Dillman and von der Lage [31]. The results are also stored for computer retrieval in the ORNL Nuclear Data Project (see Appendix B) and in the ORNL Health Physics Information System.

Several of the transactinium nuclides decay by spontaneous fission as well as by other modes, and if decay occurs by fission as much as 1% of the time, the resulting dose will be comparable to that from all other modes of decay. Since most of the energy of fission is released as kinetic energy of the fission fragments, over 80% of the dose from fission will be imparted to the organ in which the radionuclide is deposited, except for the gastrointestinal (G.I.) tract. In the G.I. tract, the fission fragments do not penetrate the mucosa overlying the radiosensitive cells to an appreciable extent so that a considerable portion of the significant dose is imparted by neutrons, β-particles, and γ-rays. Under these conditions, cross-irradiation of neighboring organs must be taken into account in estimating radiation doses.

A method has been developed by Dillman and Jones [32] for estimating the dose to various organs from spontaneous fission. For the kinetic energy of fission fragments, they used an empirical expression, accurate to $\pm 1\%$, developed in a compilation by Unik and Gindler [33]. The beta and gamma components were estimated for the various nuclides from a model based on the fission of $^{236}U^*$. The neutron component was based on ^{252}Cf. The calculated results were compared with experimental results for Th, U, and Pu isotopes. The agreement was adequate, that is, within the precision of the measurements ($\pm 20\%$). These treatments of the dosimetry for fission indicate that values

can be derived which are of acceptable accuracy. However, additional measurements of fission energy carried away by prompt and delayed γ-rays and by β-particles would refine the models and indicate the validity of the assumptions involved in their development.

In health physics, the end use of nuclear data and dose calculations is the estimation of permissible organ burdens. The organ burdens are used along with biological transport rates to establish limits on maximum permissible concentrations (MPC) in air, food, and water. The MPC values are promulgated by the International Commission on Radiation Protection and by the various national groups. In general, for nuclides expected to be important in the near future from a radiological safety viewpoint, the existing nuclear data appear to be adequate. Any uncertainties in the nuclear data are overshadowed by the uncertainties in the tracing of biological transport processes which depend on chemical properties. Problems regarding better access to nuclear data in convenient and desirable form will be discussed by Reich [34]. Additional decay data and parameters required pertain to M, N, and higher shell processes, data on which are seriously lacking. These low-energy high-intensity radiations can be of great significance in the calculation of internal dose.

IX.A. NUCLEAR DATA FOR SAFEGUARDS

B. COMPILATION ACTIVITIES

Nuclear decay data needs for safeguards have been discussed by Higinbotham and Weinstock [35] and by Weitkamp and Ottmar [36]. Higinbotham and Weinstock [35] have concluded that, in general, an adequate data base exists for active safeguards assay systems or for many of the passive techniques. Areas in which better data would be useful include γ-ray intensities for Pu-Am isotopic measurements, half-lives of Pu isotopes, and fission rates and neutrons per fission for the even isotopes of plutonium.

Because of differences in reported values of the half-lives of many of the actinide isotopes and their effect on nuclear materials accountability and safeguards, the Half-life Evaluation Committee* has initiated a program to resolve these discrepancies. The goal of this program is to measure the half-lives of the Pu and other accountable isotopes in such a manner that an authoritative recommendation can be made for adoption of half-lives. Measurements have been completed on one sample of ^{239}Pu and are planned on samples of ^{238}Pu, ^{239}Pu, and ^{240}Pu [37].

*This committee, consisting of representatives from Argonne National Laboratory, Lawrence Livermore Laboratory, Los Alamos Scientific Laboratory, Mound Laboratory, and Rocky Flats Plant, is coordinated by W. W. Strohm (Mound Laboratory).

It has been recognized that the ENDF/B-IV data files for Am, Cm, Bk, and Cf isotopes are inadequate or nonexistent for many applications. New evaluations, primarily concerned with the capture and fission cross sections, are underway [38] at the Savannah River Laboratory by McCrosson, at the Hanford Engineering Development Laboratory by Schenter, and at the Lawrence Livermore Laboratory by Howerton. Their cross-section evaluations will appear in ENDF/B-V along with decay data evaluations by Reich [34].

The Nuclear Data Project at ORNL is building a computer-based file [39] of nuclear structure data, which is intended for both basic and applied users. Their Evaluated Nuclear Structure Data File (ENSDF) now contains over 1500 data sets (decay data, reaction data, adopted level properties, etc.) spanning the $3 \leq A \leq 262$ mass range. From the ENSDF data sets, a computer program (MEDLIST) prepares tabulations of the energies and intensities of all nuclear and atomic radiations involved in a specific decay. The MEDLIST program also calculates the equilibrium dose constant, Δ, (referred to in Section VIII of this paper) for each type of radiation. As an example, Appendix B contains a list of radiations (and Δ values) associated with ^{231}Pa + daughters [40].

X. EPILOGUE

The actinide waste problem, lurking in the background for some time, is suddenly looming large as a result of increased environmental concerns. Fissioning the actinides in an appropriate reactor has been suggested as an alternative to interment. Preliminary results, promising enough to warrant more detailed studies, have been obtained through computer calculations of actinide buildup and burnout rates in both fission and fusion reactors. More refined calculations require neutron cross sections which are more complete and of higher accuracy than are now available.

In the existing literature on the actinide waste problem, the standard procedure is to postulate > 99.5% recovery of U and Pu (and occasionally ^{237}Np as well) from irradiated fuel and to treat the remainder as "wastes" to be normally discarded in a waste stream. The early segregation of Pu, however, tends to obscure two things: (i) the Pu thus removed contains (see Table I) both fissionable ^{239}Pu and ^{241}Pu and "nonfissionable" ^{240}Pu and ^{242}Pu, and (ii) if the Pu is not utilized promptly, there is an appreciable buildup of ^{241}Am because of the relatively short half-life of ^{241}Pu ($T_{1/2}$ = 15 yr). It could be argued that the nonfissionable Pu isotopes also constitute "wastes", albeit of a different sort since they cannot be selectively discarded in a waste stream. The ^{241}Am buildup produces many complications. The ingestion hazard due to ^{241}Am is much higher than that due to ^{241}Pu. The presence of ^{241}Am in the fuel implies that ^{242}Am

and ^{242}Cm will be produced during reactor operation. The ^{242}Cm is an important source of neutrons in plutonium-oxide fast-reactor fuel because of both spontaneous fission and the ^{17,18}O(α,n) reactions. [In carbide fuels, it is the ^{13}C(α,n) reaction.]

For obtaining better predictions of the nuclidic composition of irradiated fuel elements, the emphasis is different, but the cross sections are substantially the same as those desired for better testing of the actinide recycle concept. The production paths leading to the even Cm and Cf isotopes are especially important because the buildup of these neutron-active isotopes, including ^{252}Cf, could be a very ticklish prospect, if not the bane, of the actinide recycle concept in particular reactor types. To keep a lid on the neutron activity, it has been suggested that it might be preferable not to recycle the Cm isotopes. Another argument in favor of leaving out the Cm isotopes is based on the observation that the heavier Cm isotopes are much longer lived than the lighter ones. Therefore, transmutation of the lighter ones to the heavier ones, implicit in recycling, would be counterproductive in terms of reducing the long-term waste hazard.

If, during the past, many neutron cross-section measurements were carried out explicitly in support of reactor programs, they were also carried out to gain better insight into basic physics phenomena. Therefore, these measurements have extended to many nuclides of little or no importance to reactors. If cross-section data for a particular nuclide are lacking today, the reasons can probably be traced to experimental difficulties including lack of suitable samples, interference from α-emission and spontaneous fission, short half-lives, etc. Nuclear physicists and chemists have now been called upon again to carry out extremely relevant measurements by overcoming these difficulties. They will undoubtedly rise to the challenge.

XI. ACKNOWLEDGMENTS

Several people sent contributions (see table below) for use in this review paper. Their assistance is gratefully acknowledged. Even with these contributions at hand, the author might be forgiven for his generous use of additional examples culled from ORNL activities. This is inevitable, given the task.

Two valuable contributions received from J. E. Bigelow (all personnel mentioned in this paragraph, like Bigelow, belong to ORNL unless otherwise stated) were used in Sections IV and VII. Valuable contributions were received also from R. R. Spencer (Section III), from C. L. Ottinger (Section V), and from W. W. Parkinson (Section VIII). The computer-drawn figures (Fig. 3) were provided by C. W. Nestor, Jr., on short notice. The author has benefited through discussions with several colleagues, especially J. W. T. Dabbs, L. W. Weston, and C. E. Bemis. He is also grateful to J. A. Harvey and A. Zucker and to G. L. Rogosa (U. S. Energy Research and Development Adminis-

tration) for their interest and encouragement. The lion's share of the credit for emphasizing the role physicists should play in the attempts to solve the actinide waste problem and for actively involving them in the needed measurements goes to P. H. Stelson.

Contributor	Institution
R. W. Benjamin (Section V)	Savannah River Laboratory Aiken, South Carolina
A. G. Croff (Table IV)	Oak Ridge National Laboratory Oak Ridge, Tennessee
R. Dierckx (Section III)	Euratom Ispra, Italy
D. J. Horen (Appendix B)	Oak Ridge National Laboratory Oak Ridge, Tennessee
D. L. Johnson (Section III)	Hanford Engineering Development Laboratory Richland, Washington
B. R. Leonard, Jr. (Table IV)	Battelle Pacific Northwest Laboratories Richland, Washington
F. J. McCrosson (Section III)	Savannah River Laboratory Aiken, South Carolina
S. Pearlstein (Appendix A)	Brookhaven National Laboratory Upton, L. I., New York
H. Toffer (Section III)	United Nuclear Industries, Inc. Richland, Washington

XII. REFERENCES

[1] BELL, M. J., ORIGEN — The ORNL Isotope Generation and Depletion Code, ORNL Report 4628 (1973). Program modified by KEE, C. W., and by HERMANN, O. W.

[2] BLOMEKE, J. O., KEE, C. W., NICHOLS, J. P., Projections of Radioactive Wastes to be Generated by the U. S. Nuclear Power Industry, ORNL Report TM-3965 (1974)

[3] HENNELLY, E. J., Large-Scale Production of Radioisotopes, in Radioisotope Engineering (G. G. Eichholz, ed.) M. Decker, Inc., New York (1972).

[4] RAMAN, S., Review Paper A7, this meeting.

[5] MOORE, M. S., Review Paper B3, this meeting.

[6] HOLDEN, N. E., WALKER, F. W., Chart of the Nuclides, Knolls Atomic Power Laboratory, General Electric Company, Schenectady, New York (1972).

[7] BENJAMIN, R. W., Review Paper B1, this meeting.

[8] KING, L. J., BIGELOW, J. E., COLLINS, E. D., Transuranium Processing Plant Semiannual Report, ORNL Report 5034 (1975).

[9] BENJAMIN, R. W., McCROSSON, F. J., VANDERVELDE, V. D., GORRELL, T. C., A Consistent Set of Heavy Actinide Multigroup Cross Sections, USERDA Report DP-1394 (1975).

[10] DRAPER, Jr., E. L., PARISH, T. A., University of Texas at Austin, private communication.

[11] IGARASI, S., Review Paper B4, this meeting.

[12] GARBER, D. I., KINSEY, R. R., Neutron Cross Sections, Volume 2, Curves, BNL Report 325 (1975) to be published.

[13] CROFF, A. G., Oak Ridge National Laboratory, private communication.

[14] JOHNSON, D. L., Hanford Engineering Development Laboratory, Richland, Washington, private communication.

[15] SWIATECKI, W. J., Phys. Rev. 100 (1955) 937.

[16] VIOLA, Jr., V. E., Nuclear Data A1 (1966) 391.

[17] COLVIN, D. W., SOWERBY, M. G., Proceedings of IAEA Symposium on Physics and Chemistry of Fission, Salzburg II (1965) 25.

[18] DE VOLPI, A., PORGES, K. G., Phys. Rev. C1 (1970) 683.

[19] BOLDEMAN, J. W., Nucl. Sci. Eng. 55 (1974) 188.

[20] POITOU, J., SIGNARBIEUX, C., Nucl. Instr. Methods 114 (1974) 113.

[21] LEMMEL, H. D., Bull. Am. Phys. Soc. 20 (1975) 147.

[22] SPENCER, R. R., GWIN, R., INGLE, R., WEAVER, H., Oak Ridge National Laboratory, private communication.

[23] LYNN, J. E., Review Paper B5(b), this meeting.

[24] CRANDALL, J. L., Paper presented at the Symposium Commemorating the 25th Anniversary of the Discovery of Elements 97 and 98, Berkeley (January 1975).

[25] BIGELOW, J. E., Oak Ridge National Laboratory, private communication.

[26] WESTON, L. W., TODD, J. H., Measurement of the Neutron Cross Sections of the Actinides, to be published in the Proceedings of the Conference on Nuclear Cross Sections and Technology, Washington, D. C. (March 1975).

[27] DABBS, J. W. T., HILL, N. W., BEMIS, Jr., C. E., RAMAN, S., Fission Cross-Section Measurements on Short-Lived Alpha Emitters, to be published in the Proceedings of the Conference on Nuclear Cross Sections and Technology, Washington, D. C. (March 1975).

[28] HARVEY, J. A., BENJAMIN, R. W., RAMAN, S., HILL, N. W., Oak Ridge National Laboratory, private communication.

[29] BEMIS, C. E., at Oak Ridge National Laboratory, has recently measured the absolute L-series X-ray yields for all major actinides, private communication.

[30] MARTIN, M. J., BLICHERT-TOFT, P. H., Radioactive Atoms, Auger-Electron, α-, β-, γ-, and X-Ray Data, Nuclear Data Tables 8 (1970) 1.

[31] DILLMAN, L. T., VON DER LAGE, F. C., Results published as pamphlets which are numbered supplements to the Journal of Nuclear Medicine, 211 E. 43 Street, New York.

[32] DILLMAN, L. T., JONES, T. D., Internal Dosimetry of Spontaneously Fissioning Nuclides, Health Physics 29 (1975) 111.

[33] UNIK, J. P., GINDLER, J. E., A Critical Review of the Energy Released in Nuclear Fission, ANL Report 7748 (1971).

[34] REICH, C. W., Review Paper B7, this meeting.

[35] HIGINBOTHAM, W. A., WEINSTOCK, E. V., Nuclear Data Needs for Safeguards, Trans. Am. Nucl. Soc. 21 (1975) 120.

[36] WEITKAMP, C., Nuclear Data for Safeguards, Can Better Data Improve Present Techniques?; OTTMAR, H., WEITKAMP, C., Comments on Isotopic Analysis of Plutonium by Gamma Spectrometry, Papers presented at the Symposium on Practical Applications of Research and Development in the Field of Safeguards, Rome (March 1974).

[37] STROHM, W. W., Mound Laboratory, Miamisburg, Ohio, private communication.

[38] SCHENTER, R., Hanford Engineering Development Laboratory, Richland, Washington, private communication.

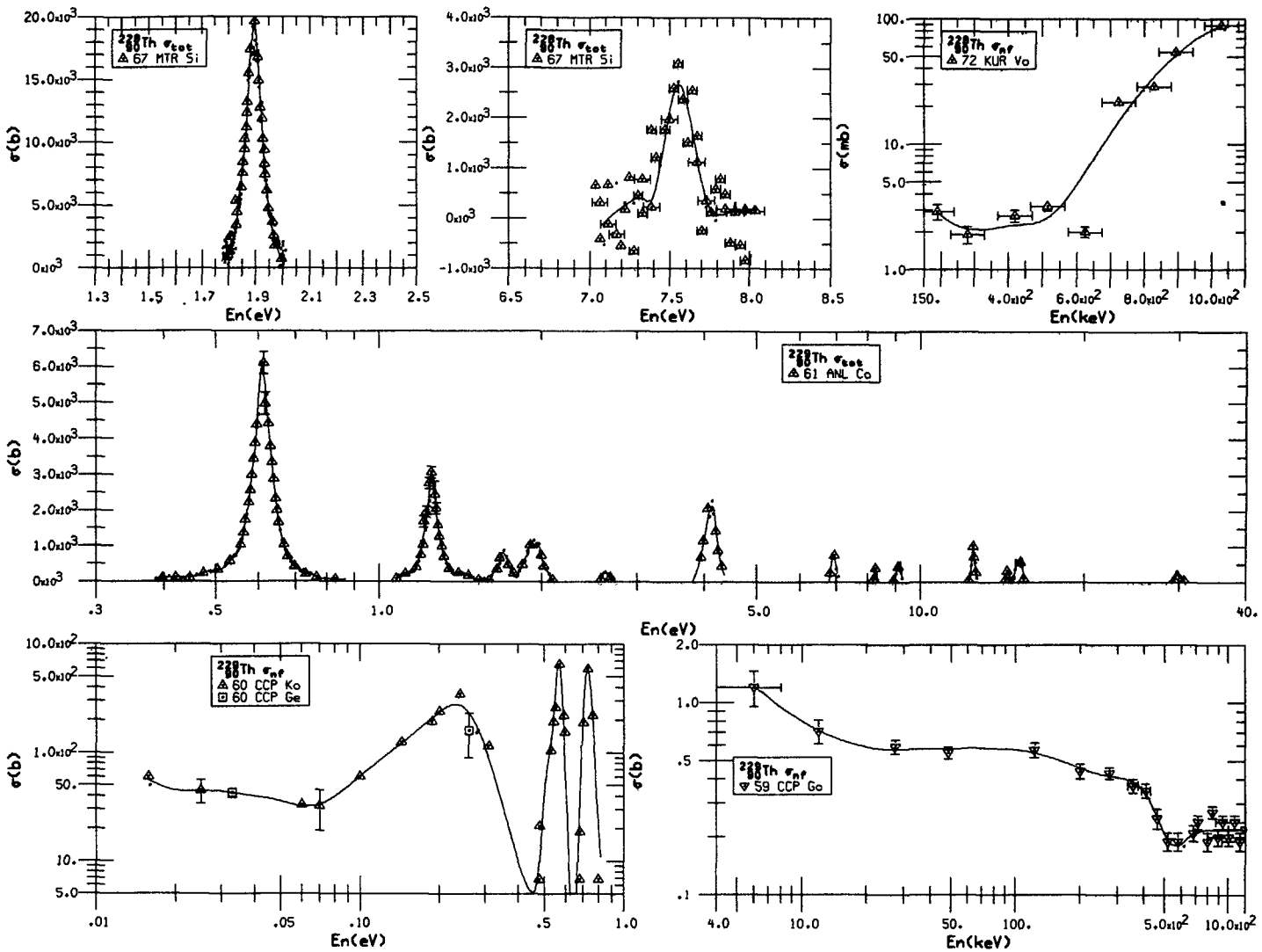
[39] EWBANK, W. B., SCHMORAK, M. R., BERTRAND, F. E., FELICIANO, M., HOREN, D. J., Nuclear Structure Data File, A Manual for Preparation of Data Sets, ORNL Report 5054 (1975).

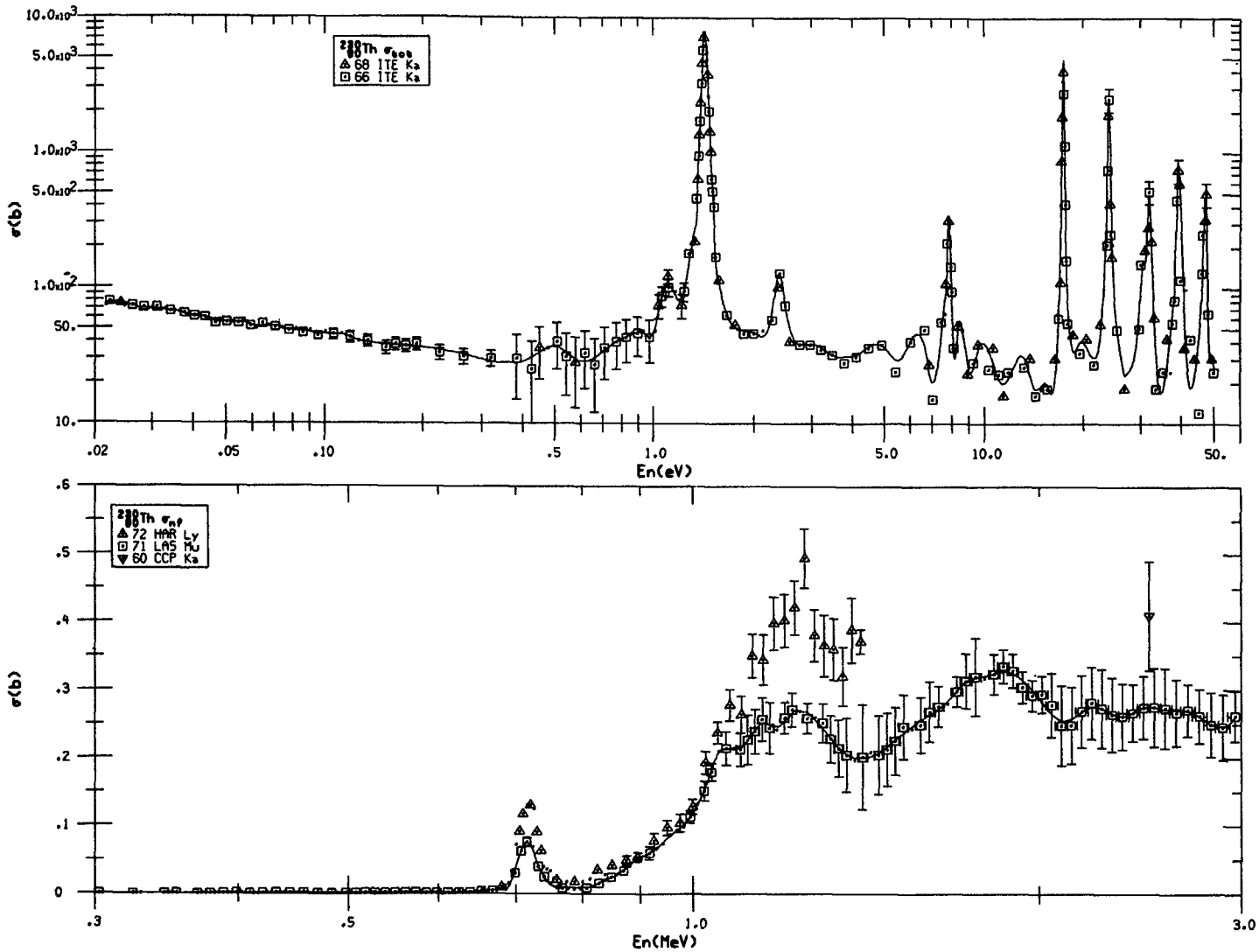
[40] MARTIN, M. J., SCHMORAK, M. R., HOREN, D. J., EWBANK, W. B., Oak Ridge National Laboratory, private communication.

APPENDIX A

Actinide Neutron Cross Sections

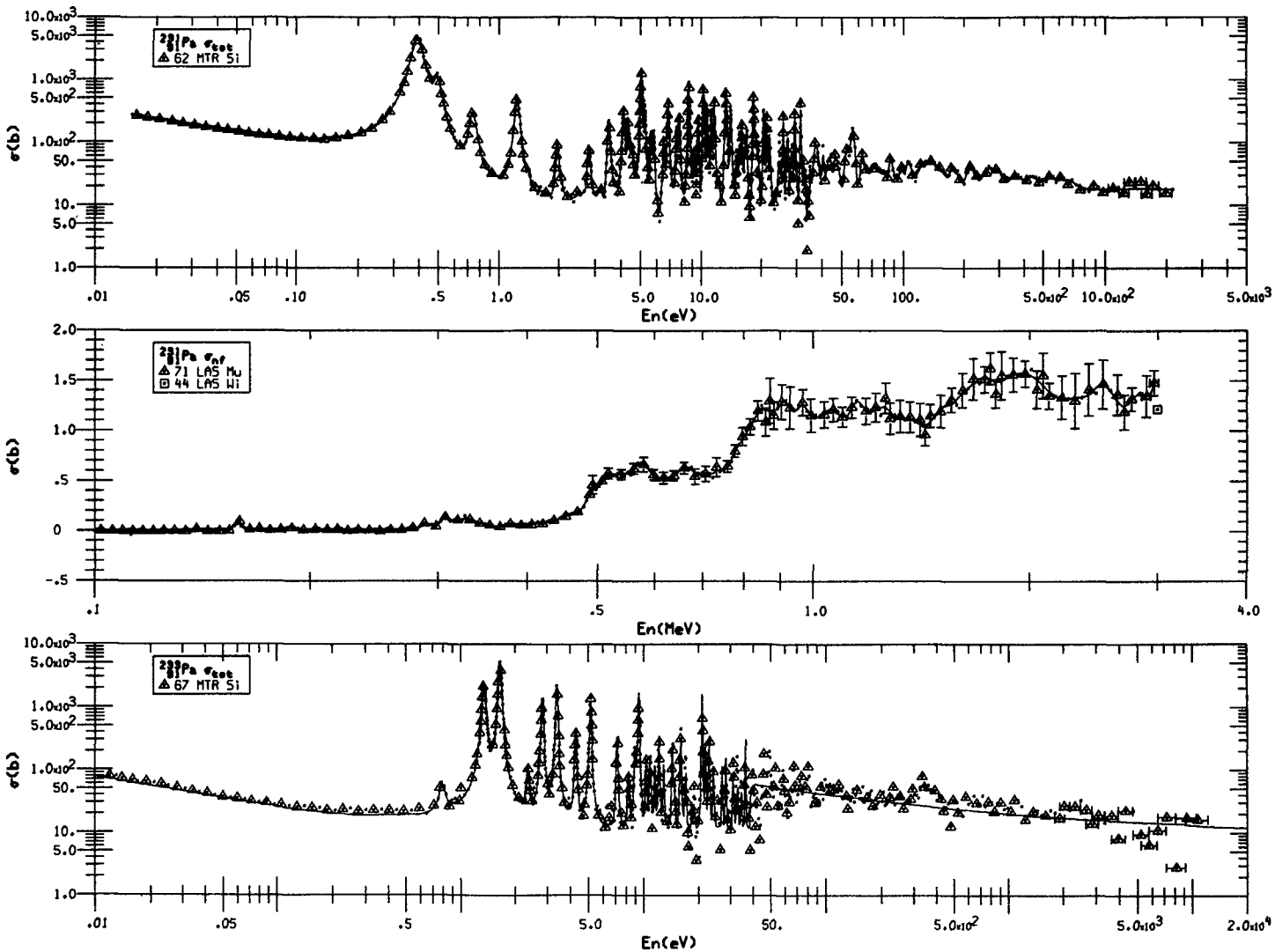
The next 34 pages contain measured neutron cross-section data (with references) for the actinides (except for ^{232}Th, ^{233}U, ^{235}U, ^{238}U, and ^{239}Pu). This material, part of a forthcoming BNL 325 Report [12], was made available by the Brookhaven National Neutron Cross Section Center to assist the participants of this meeting in their discussions. The reference style is the same as that of CINDA.


$^{228}_{90}$Th σ_{tot}
67 MTR Si
$^{228}_{90}$Th σ_{tot}
67 MTR Si
$^{228}_{90}$Th σ_{nf}
72 KUR Vo
$^{228}_{90}$Th σ_{tot}
61 ANL Co
$^{228}_{90}$Th σ_{nf}
60 CCP Ko
60 CCP Ge
$^{228}_{90}$Th σ_{nf}
59 CCP Go
σ(b)
σ(mb)
En(eV)
En(keV)

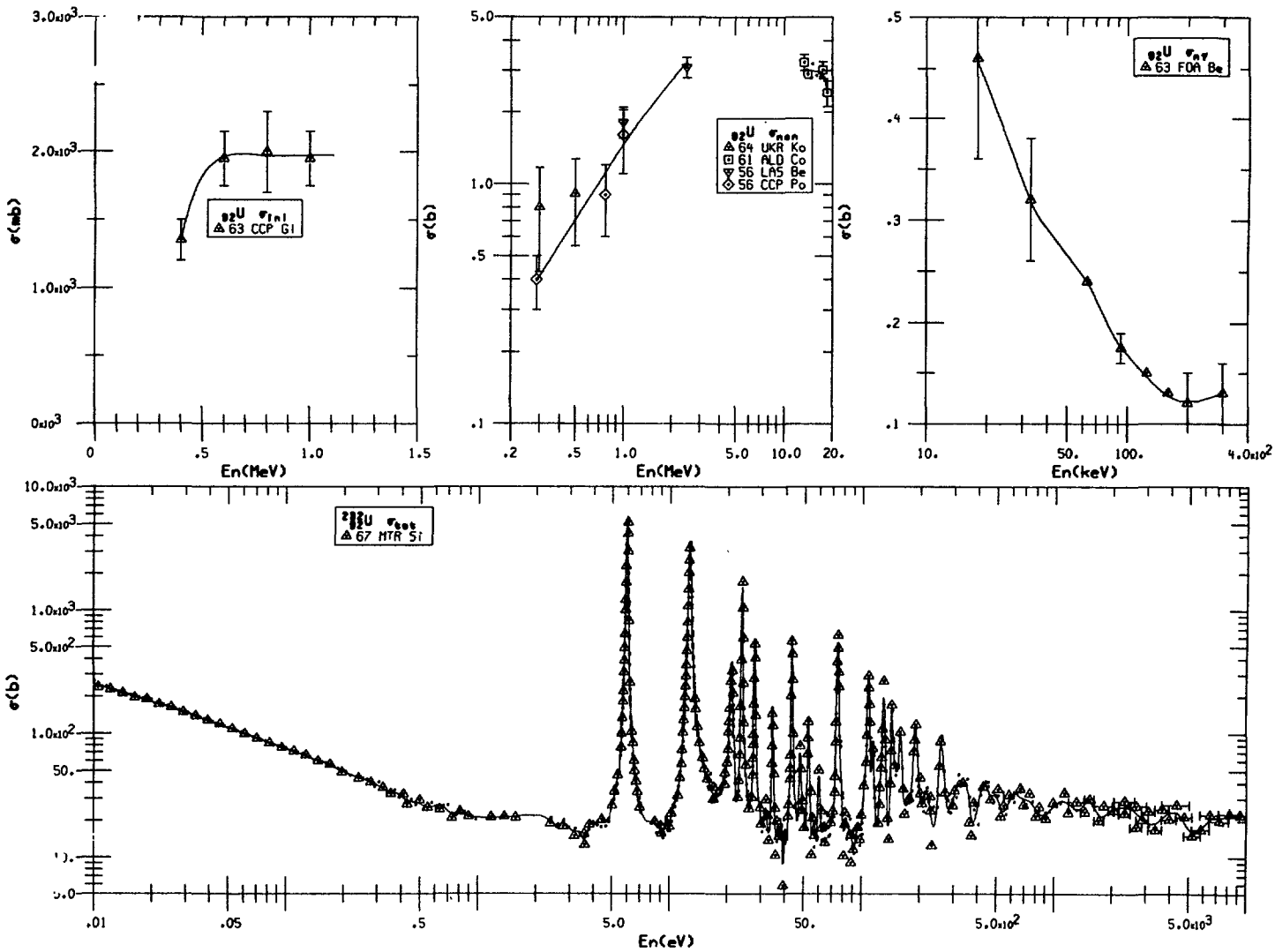

$^{232}_{90}$Th σ_{tot}
68 ITE Ka
66 ITE Ka
σ(b)
En(eV)
$^{232}_{90}$Th σ_{nf}
72 HAR Ly
71 LAS MJ
60 CCP Ka
σ(b)
En(MeV)

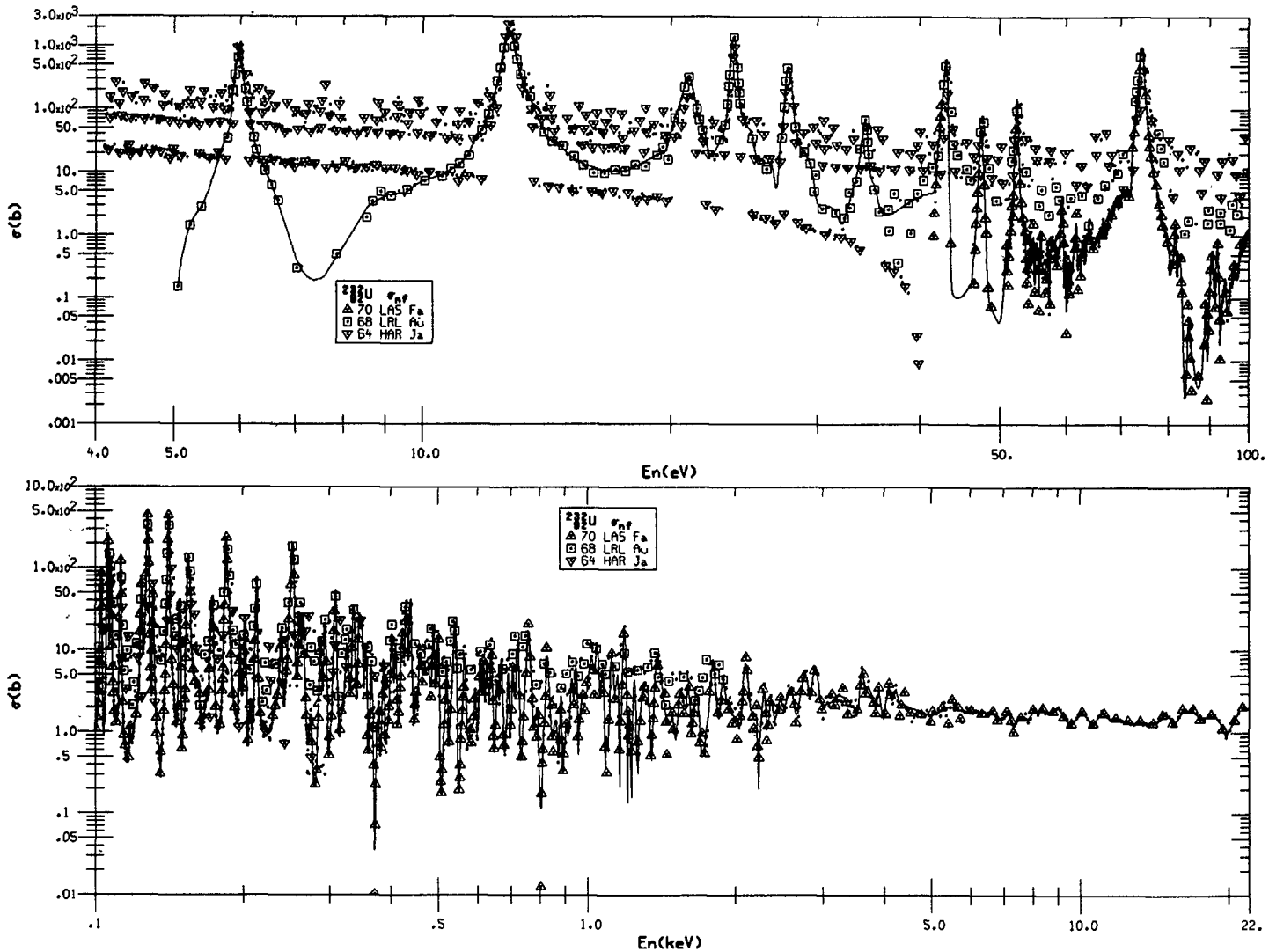
Yr	Lab	Author	Reference	Points	Range	Standard
			$^{232}_{90}$Th σ_{nf}			
73	MOL	Del Marmol,et.al	JIN,35,4323,73	1	.203 kb at thermal	^{197}Au $\sigma_{n\gamma}$
			$^{232}_{90}$Th σ_{tot}			
67	MTR	Simpson,et.al.	NSE,29,423,67	213	1.779 eV to 8.038 eV	
			$^{232}_{90}$Th σ_{nf}			
72	KUR	Vorotnikov+	YFI-14,6,72	9	.190 MeV to 1.030 MeV	
			$^{232}_{90}$Th σ_{tot}			
61	ANL	Cote,et.al.	BAP,6,417 C1 ,61	231	.387 eV to 30.7 eV	
			$^{232}_{90}$Th σ_{nf}			
60	CCP	Konakhovich+	AE,8,47,60	26	.0158 eV to .800 eV	^{229}Th σ_{nf}
60	CCP	Gerasimov	AE,8,47,60-	3	.0160 eV to .260 eV	^{229}Th σ_{nf}
59	CCP	Gokhberg,et.al.	DOK,128,911,59	23	6.000 keV to 1.200 MeV	
			$^{232}_{90}$Th σ_{tot}			
68	ITE	Kalebin,et.al.	68DUBNA,(ACC-68/18)	221	.0217 eV to 50.1 eV	abs
66	ITE	Kalebin,et.al.	66PARIS,1,71,66	221	.0217 eV to 50.1 eV	abs
			$^{232}_{90}$Th σ_{na}			
59	ALD	Coleman,et.al.	PPS,73,215,59	1	4.600 mb at 14.5 MeV	^{27}Al σ_{na}
			$^{232}_{90}$Th σ_{nf}			
72	HAR	Lynn,et.al.	NP/A,189,225,72	50	.625 MeV to 1.400 MeV	^{230}Th σ_{nf}
71	LAS	Muir,et.al.	71KNOX,1,292,71	181	.300 MeV to 2.965 MeV	^{239}Pu σ_{nf}
67	CCP	Vorotnikov	YF,5,295,67	12	.650 MeV to 1.230 MeV	
65	ORL	Lamphere	65SALZBURG,1,63,65	42	.675 MeV to 2.930 MeV	
60	CCP	Kazarinova+	AE,8,139,60	2	2.500 MeV to 14.6 MeV	
			$^{232}_{90}$Th σ_{tot}			
71	BNW	Foster Jr,et.al.	PR/C,3,576,71	242	2.263 MeV to 14.9 MeV	
71	BET	Green,et.al.	71KNOX,1,325,71	644	.501 MeV to 9.599 MeV	
70	SAC	Ribon	RIBON,70	4117	.212 keV to 10.7 keV	abs
68	JNE	Manero	AR5,64,373,68	60	3.340 MeV to 5.100 MeV	
68	FEI	Filipov,et.al.	68DUBNA,(ACC-68/17)	8	20.0 keV to .350 MeV	
66	HAR	Uttley,et.al.	66PARIS,1,165,66	28	6.500 keV to .950 MeV	abs
66	PAD	Zago	ZAGO,66	139	1.470 MeV to 8.635 MeV	
65	ORL	Rayburn,et.al.	NP,61,381,65	1	13.3 b at 1.440 eV	
65	ALD	Batchelor,et.al.	NP,65,236,65	4	2.000 MeV to 7.000 MeV	abs
64	COL	Garg,et.al.	PR/8,134,985,64	5877	82.3 eV to 3.961 keV	
64	DKE	Tabony,et.al.	SETH,64	102	30.0 keV to .650 MeV	
64	HAR	Pattenden	PATTENDEN,64	832	15.3 eV to .289 keV	
63	SAC	Leroy,et.al.	JPR,24,826,63	56	1.980 MeV to 14.5 MeV	
61	LVN	Deconninck,et.al	JPR,22,652,61	1	5.490 b at 28.4 MeV	
61	HAR	Uttley,et.al.	61SACLAY,,109,61	33	.350 keV to 60.0 keV	
61	CCP	Averchenkov+	SOV.PR.NP.,,191,61	17	1.900 MeV to 8.000 MeV	abs
60	JAE	Tsukada,et.al.	JPJ,15,1994,60	89	3.370 MeV to 5.070 MeV	
58	BNL	Seth,et.al.	PR,110,692,58	21	.526 eV to 53.7 eV	
55	BNL	Pilcher	PILCHER,55	139	21.3 eV to .580 keV	

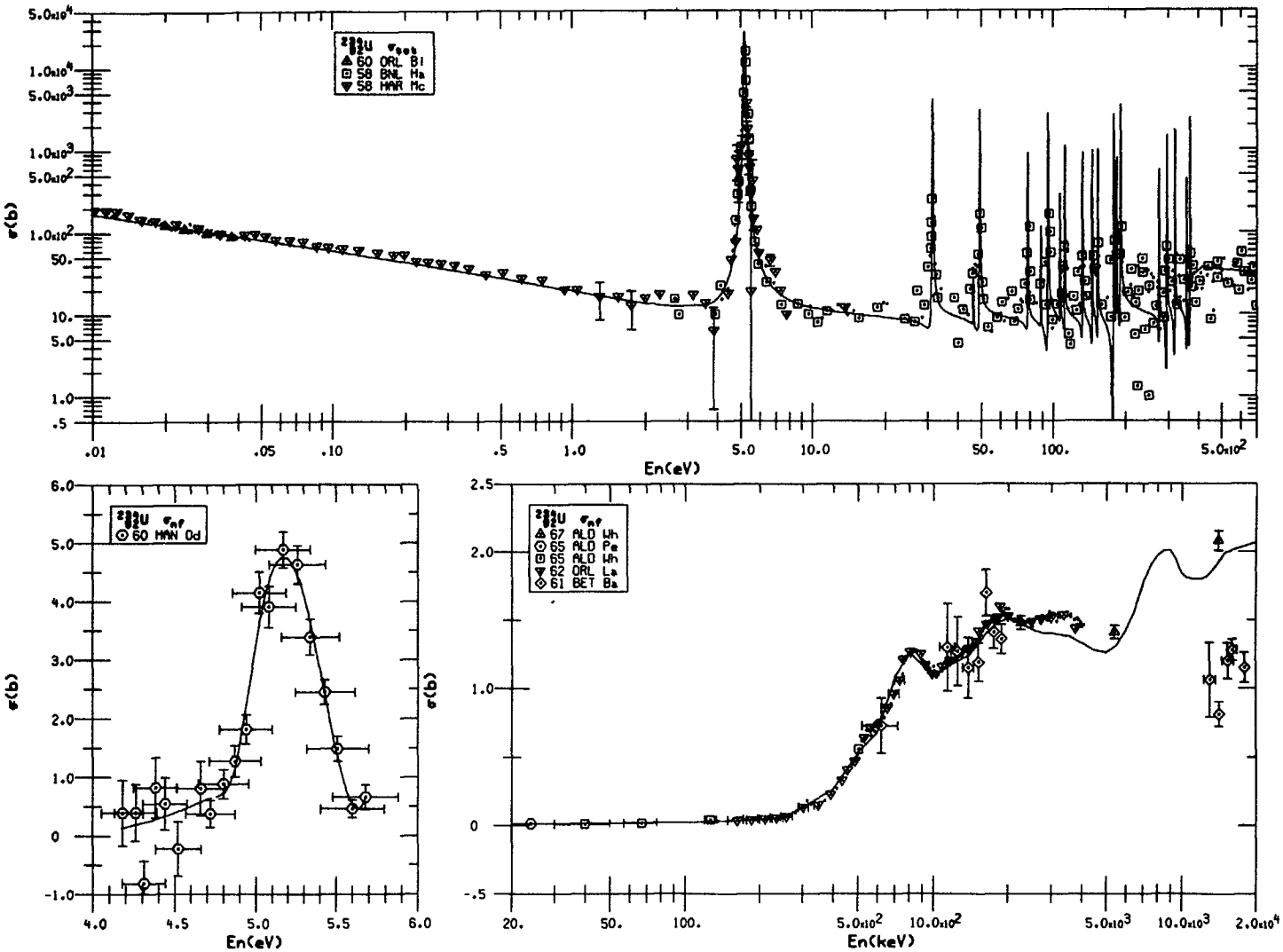
Yr	Lab	Author	Reference	Points	Range	Standard
			cont			
55	ANL	Bollinger,et.al.	PR,98,223(82),55	286	9.700 eV to 2.331 keV	
54	ANL	Hibdon,et.al.	ANL-5175,7,54	133	.670 keV to .155 MeV	abs
54	BNL	Seidl,et.al.	PR,95,476,54	128	16.9 eV to .225 keV	
54	HAR	Egelstaff	EGELSTAFF,54	52	.0019 eV to 2.500 eV	abs
53	WIS	Walt,et.al.	PR,89,1271,53	18	89.0 keV to 2.980 MeV	
52	LAS	Coon,et.al.	PR,88,562,52	1	5.890 b at 14.1 MeV	
51	COL	Havens Jr.+	AECD-3288,51	167	.0082 eV to 20.8 keV	
50	BRK	Hildebrand+	PR,80,842,50	1	5.030 b at 42.0 MeV	
			$^{232}_{90}$Th σ_{el}			
65	ANL	Smith,et.al.	EANDC US 62,65	16	.300 MeV to 1.500 MeV	C
65	ALD	Batchelor,et.al.	NP,65,236,65	4	2.000 MeV to 7.000 MeV	H
65	FEI	Kasakova,et.al.	65ANTWERP,,576,65	1	5.000 b at 2.000 MeV	
62	WIR	Hudson Jr,et.al.	PR,128,1271,62	1	3.130 b at 15.2 MeV	
61	CCP	Popov	SOV.PR.NP.,,224,61	1	3.700 b at 3.100 MeV	
54	WIS	Walt,et.al.	PR,93,1062,54	1	5.300 b at 1.000 MeV	
			$^{232}_{90}$Th σ_{inl}			
65	ALD	Batchelor,et.al.	NP,65,236,65	3	3.000 MeV to 7.000 MeV	
63	CCP	Glazkov	AE,14,400,63	4	.400 MeV to 1.000 MeV	
			$^{232}_{90}$Th σ_{sct}			
65	ORL	Rayburn,et.al.	NP,61,381,65	1	12.4 b at 1.440 eV	
57	ANL	Langsdorf Jr+	PR,107,1077,57	17	60.0 keV to 1.370 MeV	C
			$^{232}_{90}$Th σ_{non}			
65	ALD	Batchelor,et.al.	NP,65,236,65	3	3.000 MeV to 7.000 MeV	
63	ALD	Mc Taggart+	JNE,17,437,63	1	2.770 b at 14.0 MeV	
61	CCP	Popov	SOV.PR.NP.,,224,61	1	3.700 b at 3.100 MeV	
54	WIS	Walt,et.al.	PR,93,1062,54	1	1.800 b at 1.000 MeV	
			$^{232}_{90}$Th $\sigma_{n\gamma}$			
72	FEI	Chelnokov,et.al.	YFI-13,6,72	26	.200 keV to 34.6 keV	^{197}Au $\sigma_{n\gamma}$
71	LRL	Nagle,et.al.	71KNOX,2,259,71	30	.121 MeV to 2.730 MeV	^{235}U σ_{nf}
68	UPP	Lundgren	NUK,11,61,68	47	.0980 eV to 3.400 eV	
65	MUA	Chaubey,et.al.	NP,66,267,65	1	.480 b at 24.0 keV	^{127}I $\sigma_{n\gamma}$
64	ORL	Gibbons	GIBBONS,64	126	10.0 keV to 58.5 keV	
63	ANL	Stupegia,et.al.	JIN,25,627,63	22	.191 MeV to 1.170 MeV	^{235}U σ_{nf}
63	HAR	Moxon,et.al.	TRDWP/P-8,63	98	5.500 keV to .148 MeV	
63	CCP	Tolstikov,et.al.	AE,15,414,63	10	5.300 keV to .100 MeV	^{232}Th $\sigma_{n\gamma}$
62	LRL	Miskel,et.al.	PR,128,2717,62	26	32.0 keV to 3.970 MeV	^{235}U σ_{nf}
61	CCP	Stavissky+	AE,10,508,61	38	30.0 keV to 5.850 MeV	^{235}U σ_{nf}
60	CCP	Belanova	AE,8,549,60	1	.213 b at .220 MeV	
59	HAR	Hanna,et.al.	JNE,8,197,59	24	.100 MeV to 1.230 MeV	^{197}Au $\sigma_{n\gamma}$
59	ALD	Barry,et.al.	PPS,74,685,59	10	.300 MeV to 1.200 MeV	
58	ALD	Perkin,et.al.	PPS,72,505,58	1	5.200 mb at 14.5 MeV	^{27}Al σ_{na}
58	CCP	Leipunsky,et.al.	58GENEVA,15,50,58	1	.217 b at .200 MeV	^{127}I
58	CCP	Belanova	ZET,34,574,58	2	25.0 keV to .830 MeV	
57	SRL	Wade	DP-207,57	1	7.550 b at thermal	^{Fe} $\sigma_{n\gamma}$
57	ORL	Macklin,et.al.	PR,107,504,57	1	.500 b at 24.0 keV	^{127}I $\sigma_{n\gamma}$
57	SAC	Hubert,et.al.	TID-7547,39,57	1	7.600 b at thermal	
55	HAR	Crocker	JNE,1,234,55	1	7.310 b at thermal	^{197}Au $\sigma_{n\gamma}$
55	HAR	Egelstaff,et.al.	EGELSTAFF,55	1	7.200 b at thermal	

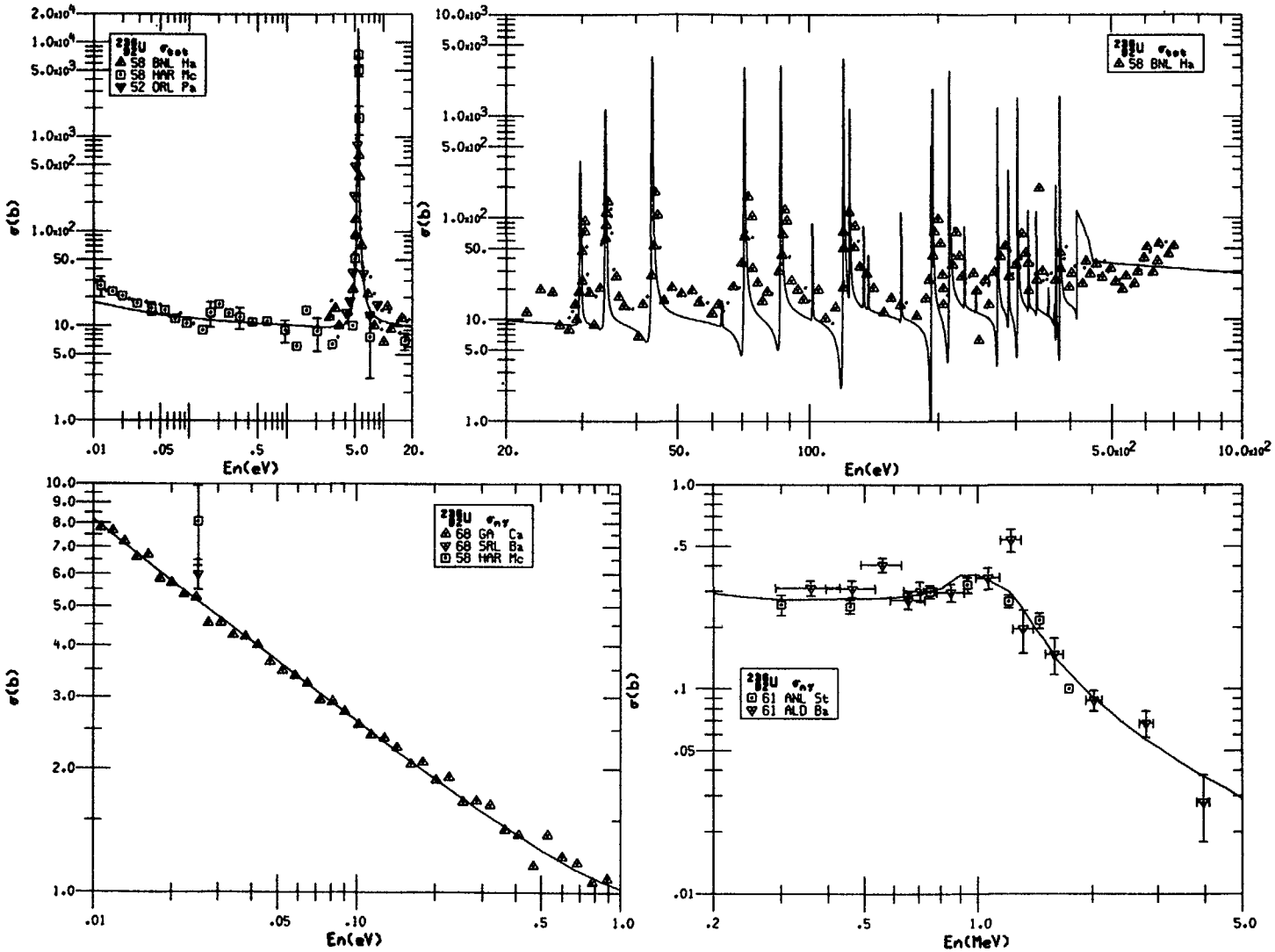

231Pa σtot
62 MTR Si
σ(b)
En(eV)
231Pa σnf
71 LAS Mu
44 LAS Wi
σ(b)
En(MeV)
231Pa σtot
67 MTR Si
σ(b)
En(eV)

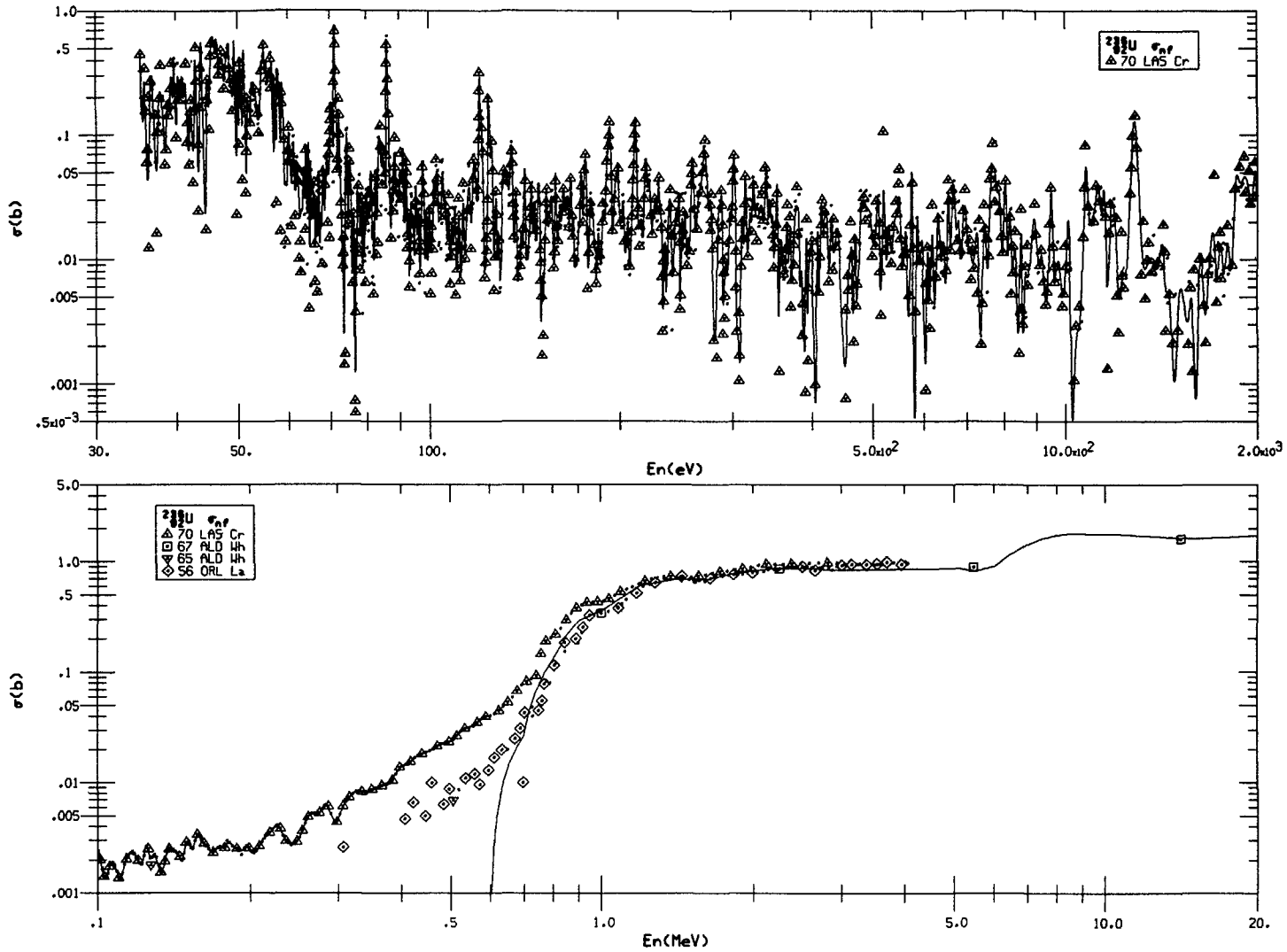
Yr	Lab	Author	Reference	Points	Range	Standard
			$^{233}_{91}$Pa σ_{tot}			
62	MTR	Simpson,et.al.	NSE,12,243,62	1094	.531 eV to 2.141 keV	abs
62	MTR	Simpson,et.al.	NSE,12,243,62	248	.0152 eV to 10.6 eV	
62	MTR	Simpson,et.al.	NSE,12,243,62	1	.211 kb at thermal	
			$^{233}_{91}$Pa $\sigma_{n\gamma}$			
62	MTR	Simpson,et.al.	NSE,12,243,62	1	.200 kb at thermal	
			$^{233}_{91}$Pa σ_{nf}			
71	LAS	Muir,et.al.	71KNOX,1,292,71	374	.100 MeV to 2.965 MeV	^{239}Pu σ_{nf}
69	KEN	Birgul,et.al.	RCA,11,108,69	1	1.430 b at 14.8 MeV	^{238}U σ_{nf}
64	CCP	Dubrovina,et.al.	DOK,157,561,64	46	.133 MeV to 1.730 MeV	
44	LAS	Williams	LA-150,44	15	.430 MeV to 3.000 MeV	^{235}U σ_{nf}
			$^{233}_{92}$U σ_{tot}			
67	MTR	Simpson,et.al.	NSE,28,133,67	847	.0110 eV to 10.7 keV	
67	MTR	Simpson,et.al.	NSE,28,133,67	1	55.0 b at thermal	
			$^{233}_{92}$U $\sigma_{n\gamma}$			
70	BET	Connor	WAPD-TM-837,70	1	39.5 b at thermal	
67	MTR	Simpson,et.al.	NSE,28,133,67	1	41.0 b at thermal	
62	ORL	Halperin,et.al.	ORNL-3320,1,62	1	42.0 b at thermal	

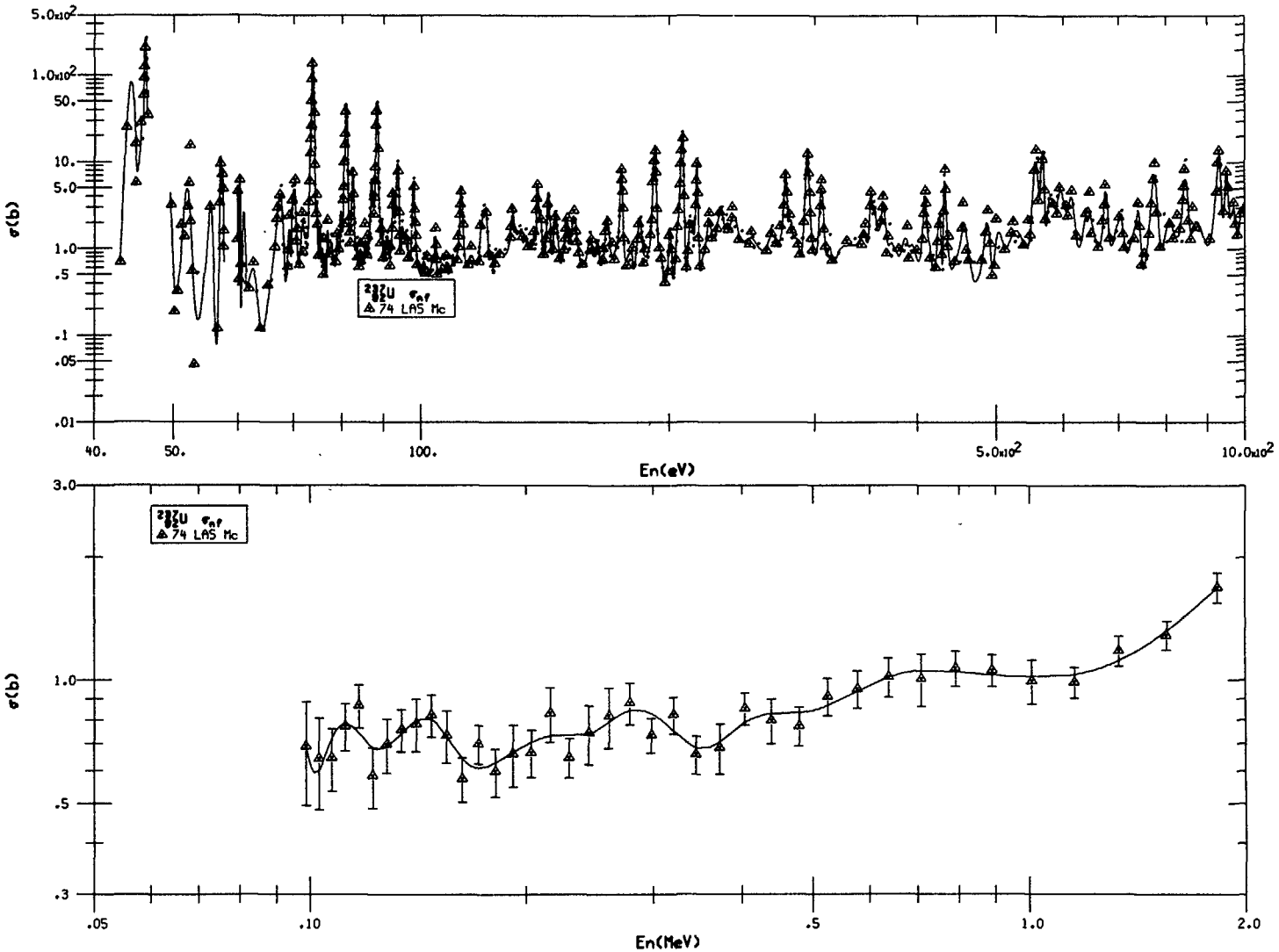



$^{233}_{92}U$ σ_{nf}
70 LAS Fa
68 LRL Au
64 HAR Ja
σ(b)
En(eV)
En(keV)


60 ORL Bl
58 BNL Ha
58 MAR Mc
σ(b)
En(eV)
60 MAN Od
67 ALO Wh
65 ALO Pe
65 ALO Wh
62 ORL La
61 BET Ba
En(keV)

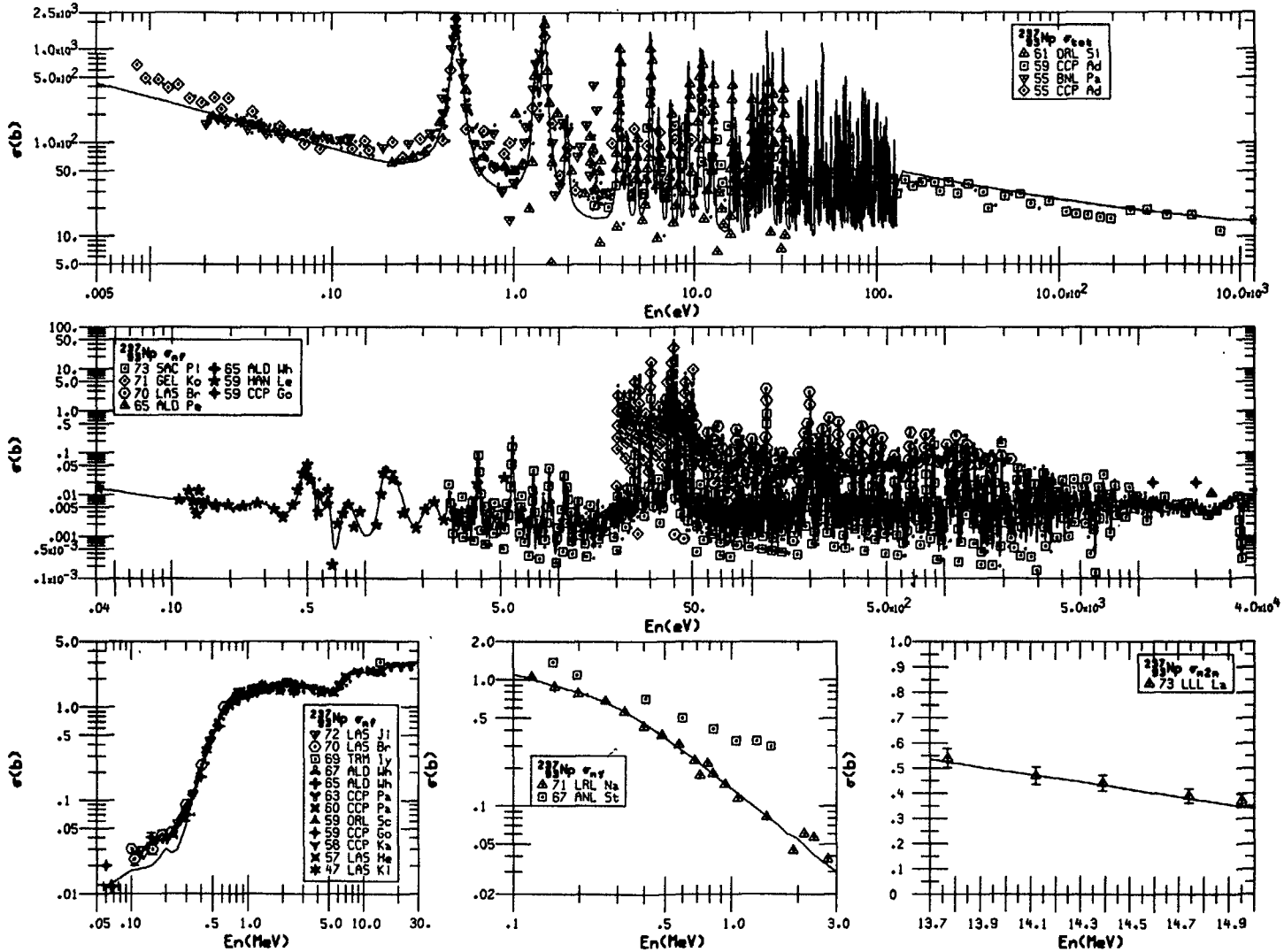

238U σtot
58 BNL Ha
58 HAR Mc
52 ORL Pa
σ(b)
En(eV)
238U σtot
58 BNL Ha
σ(b)
En(eV)
238U σnγ
68 GA Ca
68 SRL Ba
58 HAR Mc
σ(b)
En(eV)
238U σnγ
61 ANL St
61 ALD Ba
En(MeV)


238U σn,f
70 LAS Cr
σ(b)
En(eV)
238U σn,f
70 LAS Cr
67 ALD Fe
65 ALD Fe
56 ORL La
σ(b)
En(MeV)

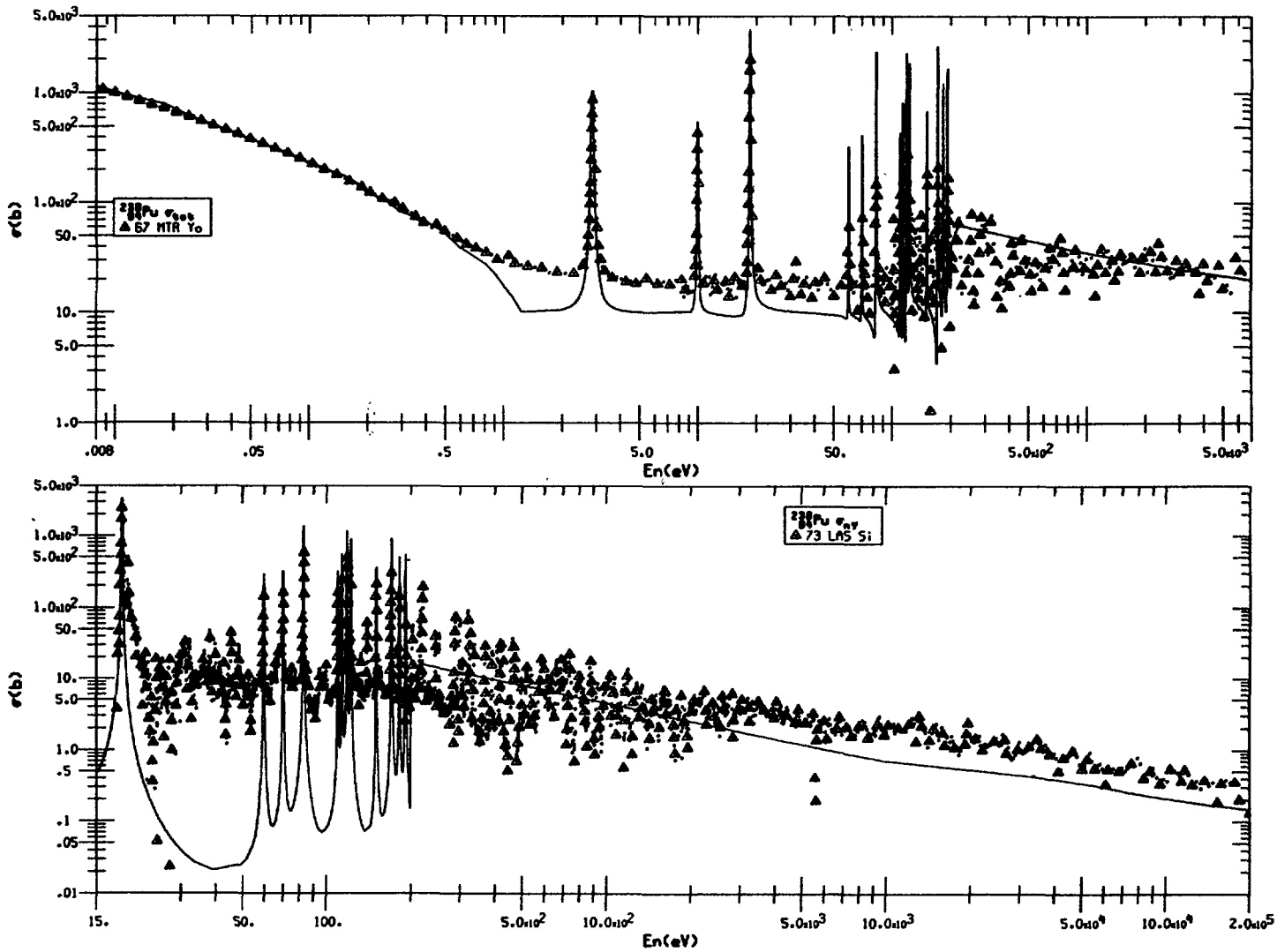

5.0x10²
1.0x10²
50.
10.
5.0
1.0
.5
.1
.05
.01
σ(b)
233U σnf
74 LAS Mc
40.
50.
100.
5.0x10²
10.0x10²
En(eV)
3.0
1.0
.5
.3
σ(b)
233U σnf
74 LAS Mc
.05
.10
.5
1.0
2.0
En(MeV)

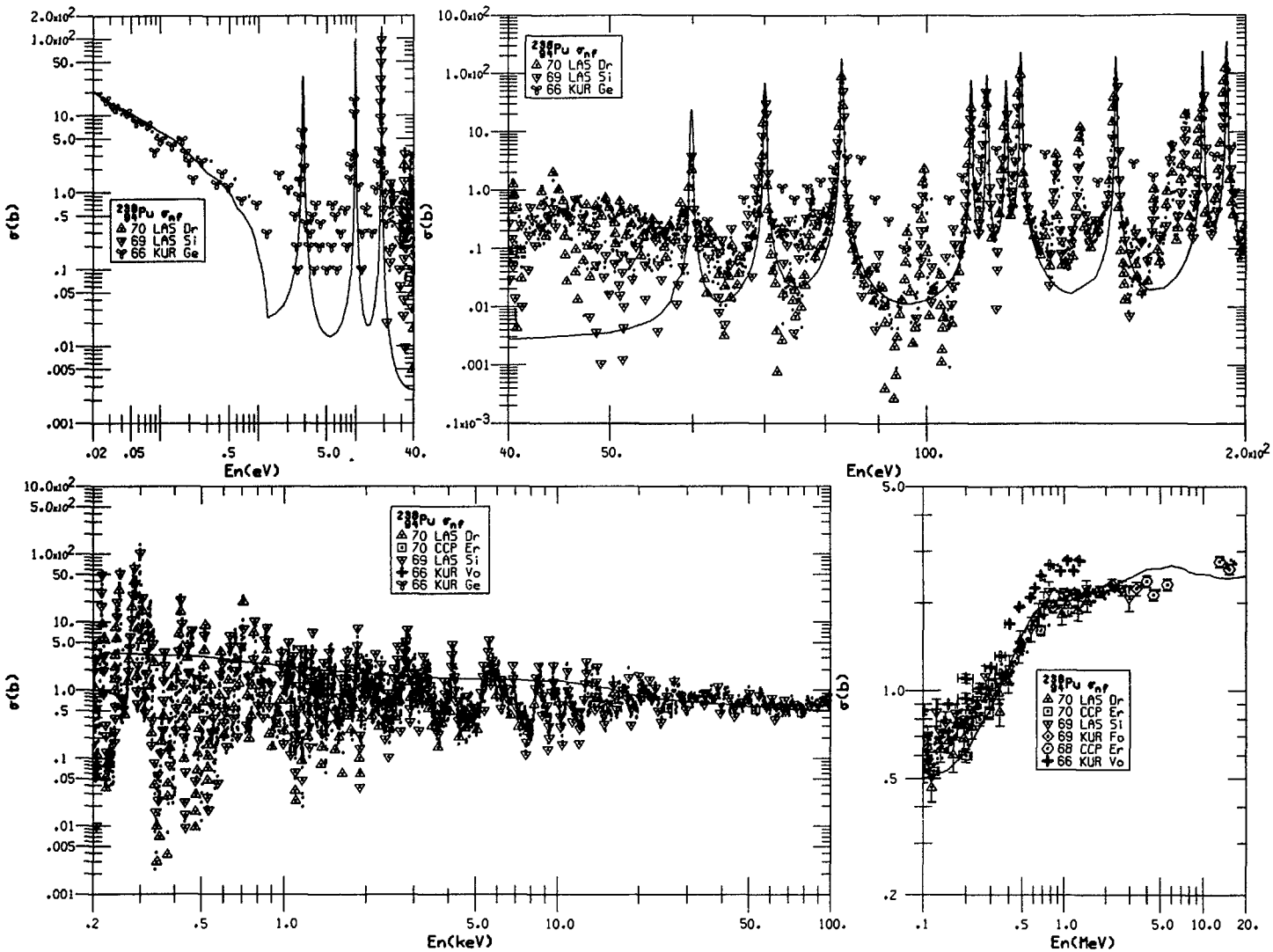
Yr	Lab	Author	Reference	Points	Range	Standard
			^{92}U $\sigma_{n\gamma}$			
63	FOA	Bergqvist	AF,23,425,63	8	18.0 keV to .300 MeV	Ag $\sigma_{n\gamma}$
61	BUC	Stefanescu,et.al	61BUCHAR,,553,61	1	7.830 b at thermal	B $\sigma_{n\gamma}$
			$^{233}_{92}U$ σ_{tot}			
67	MTR	Simpson,et.al.	NSE,29,415,67	931	.0101 eV to 9.292 keV	
			$^{233}_{92}U$ σ_{nf}			
70	KUR	Vorotnikov	YF,12,474,70	23	.130 MeV to 1.500 MeV	
70	LAS	Farrell	LA-4420,3,70	2199	40.1 eV to 21.3 keV	^{6}Li σ_{nt}
68	LRL	Auchampaugh+	NP/A,112,329,68	707	5.050 eV to 1.927 keV	^{232}U σ_{nf}
64	HAR	James	NP,55,517,64	3005	3.691 eV to .401 keV	^{235}U σ_{nf}
			$^{233}_{92}U$ ν			
73	CCP	Nurpeisov	AE,34,491,73	12	.320 MeV to 1.400 MeV	
72	FEI	Sergachev	YF,16,475,72	36	.100 MeV to 5.400 MeV	
72	FEI	Kolosov,et.al.	AE,32,(1),83,72	15	70.0 keV to 1.560 MeV	^{233}U ν
67	FEI	Kuznetsov,et.al.	AE,22,401,67	7	80.0 keV to .700 MeV	
64	FEI	Bljumkina+	NP,52,(4),648,64	7	80.0 keV to .700 MeV	
			$^{233}_{92}U$ σ_{tot}			
60	ORL	Block,et.al.	NSE,8,112,60	26	.0199 eV to .0444 eV	
58	BNL	Harvey,et.al.	PR,109,471,58	253	2.650 eV to .695 keV	
58	HAR	Mccallum	JNE,6,181,58	97	.0102 eV to 20.3 eV	
			$^{233}_{92}U$ σ_{nf}			
68	HAR	James	NP/A,118,68	217	4.000 eV to 20.0 keV	
67	ALD	White,et.al.	JNE,21,671,67	4	1.000 MeV to 14.1 MeV	^{235}U σ_{nf}
65	ALD	Perkin,et.al.	JNE,19,423,65	1	15.0 mb at 24.0 keV	
65	ALD	White,et.al.	65SAL2BURG,1,219,65	5	40.0 keV to .505 MeV	
62	ORL	Lamphere	NP,38,561,62	164	.136 MeV to 4.054 MeV	
61	BET	Babcock	BABCOCK,61	15	.350 MeV to 18.0 MeV	
60	HAN	Odegaarden	HW-64866,4,60	20	4.180 eV to 5.680 eV	

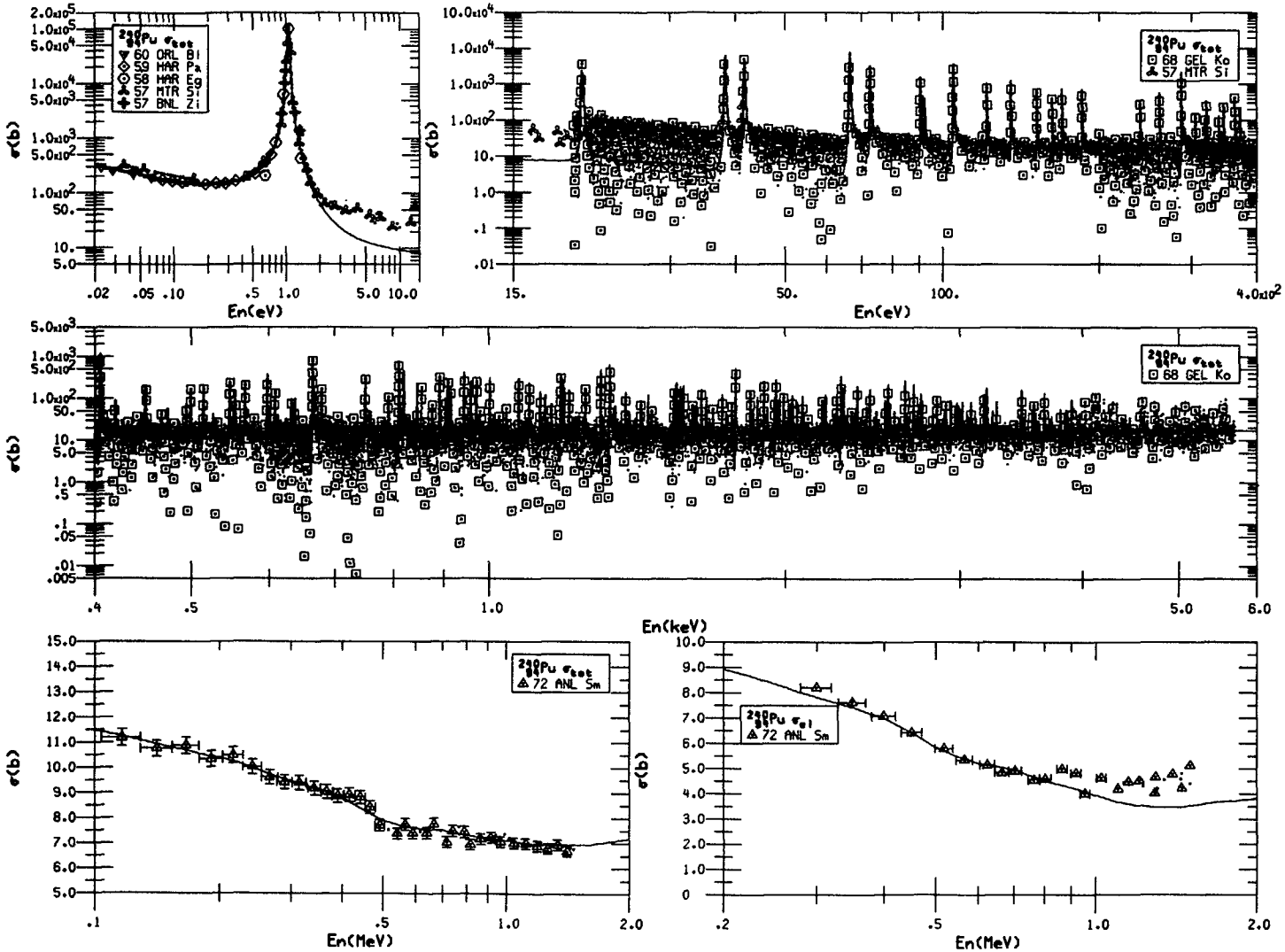
Yr	Lab	Author	Reference	Points	Range	Standard
			$^{234}_{92}U$ σ_{tot}			
58	BNL	Harvey,et.al.	PR,109,471,58	240	2.750 eV to .700 keV	
58	HAR	Mccallum	JNE,6,181,58	52	.0105 eV to 16.8 eV	
58	HAR	Mccallum	JNE,6,181,58	1	18.7 b at thermal	
52	ORL	Pavlicki,et.al.	ORNL-1477,52	18	4.100 eV to 9.500 eV	
			$^{234}_{92}U$ $\sigma_{n\gamma}$			
68	GA	Carlson,et.al.	GA-9057,68	40	.0107 eV to .891 eV	^{197}Au $\sigma_{n\gamma}$
68	SRL	Baumann,et.al.	NSE,32,265,68	1	6.000 b at thermal	
61	ANL	Stupegia,et.al.	JNE,15,200,61	8	.300 MeV to 1.732 MeV	
61	ALD	Barry,et.al.	PPS,78,801,61	14	.360 MeV to 3.970 MeV	
58	HAR	Mccallum	JNE,6,181,58	1	8.100 b at thermal	
			$^{235}_{92}U$ σ_{nf}			
70	LAS	Cramer,et.al.	LA-4420,74,70	2061	35.2 eV to 2.935 MeV	^{235}U σ_{nf}
67	ALD	White,et.al.	JNE,21,671,67	4	1.000 MeV to 14.1 MeV	^{235}U σ_{nf}
65	ALD	Perkin,et.al.	JNE,19,423,65	1	4.000 mb at 24.0 keV	
65	ALD	White,et.al.	65SAL2BURG,1,219,65	2	.127 MeV to .505 MeV	
56	ORL	Lamphere	PR,104,1654,56	101	.307 MeV to 4.050 MeV	^{235}U σ_{nf}
			$^{237}_{92}U$ σ_{nf}			
74	LAS	Mc Nally,et.al.	PR/C,9,717,74	1116	43.0 eV to 1.830 MeV	

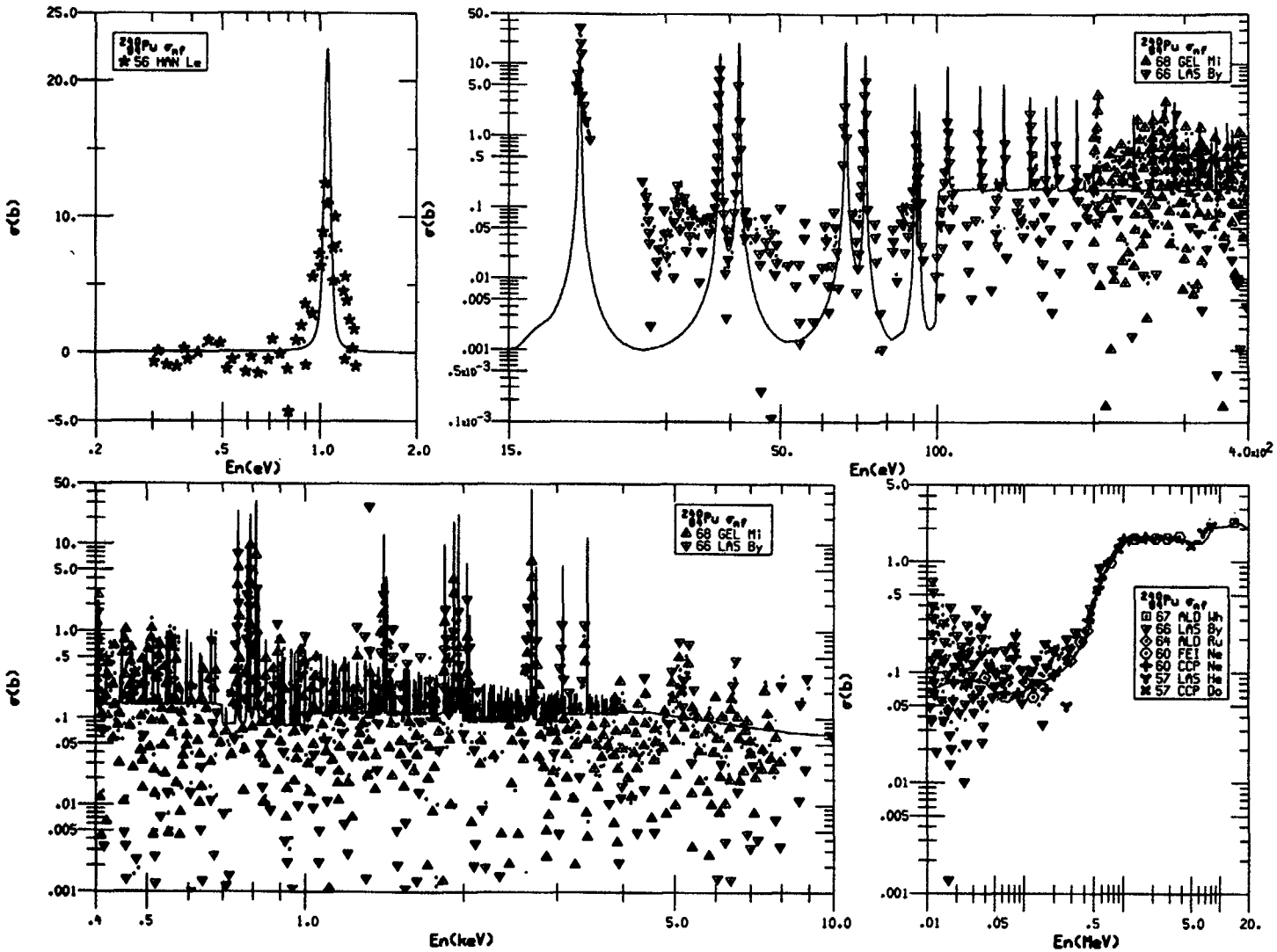

σ(b)
En(eV)
En(MeV)
237 93 Np σtot
61 ORL Si
59 CCP Ad
55 BNL Pa
55 CCP Ad
237 93 Np σnf
73 SAC Pi
71 GEL Ko
70 LAS Br
65 ALD Pe
65 ALD Wh
59 HAN Le
59 CCP Go
237 93 Np σnf
72 LAS Ji
70 LAS Br
69 TRM Ivy
67 ALD Wh
65 ALD Wh
63 CCP Pe
60 CCP Pe
59 ORL Pe
59 CCP Go
58 CCP Ka
57 LAS Ma
47 LAS Kl
237 93 Np σnf
71 LRL Na
67 ANL St
237 93 Np σn2n
73 LLL La

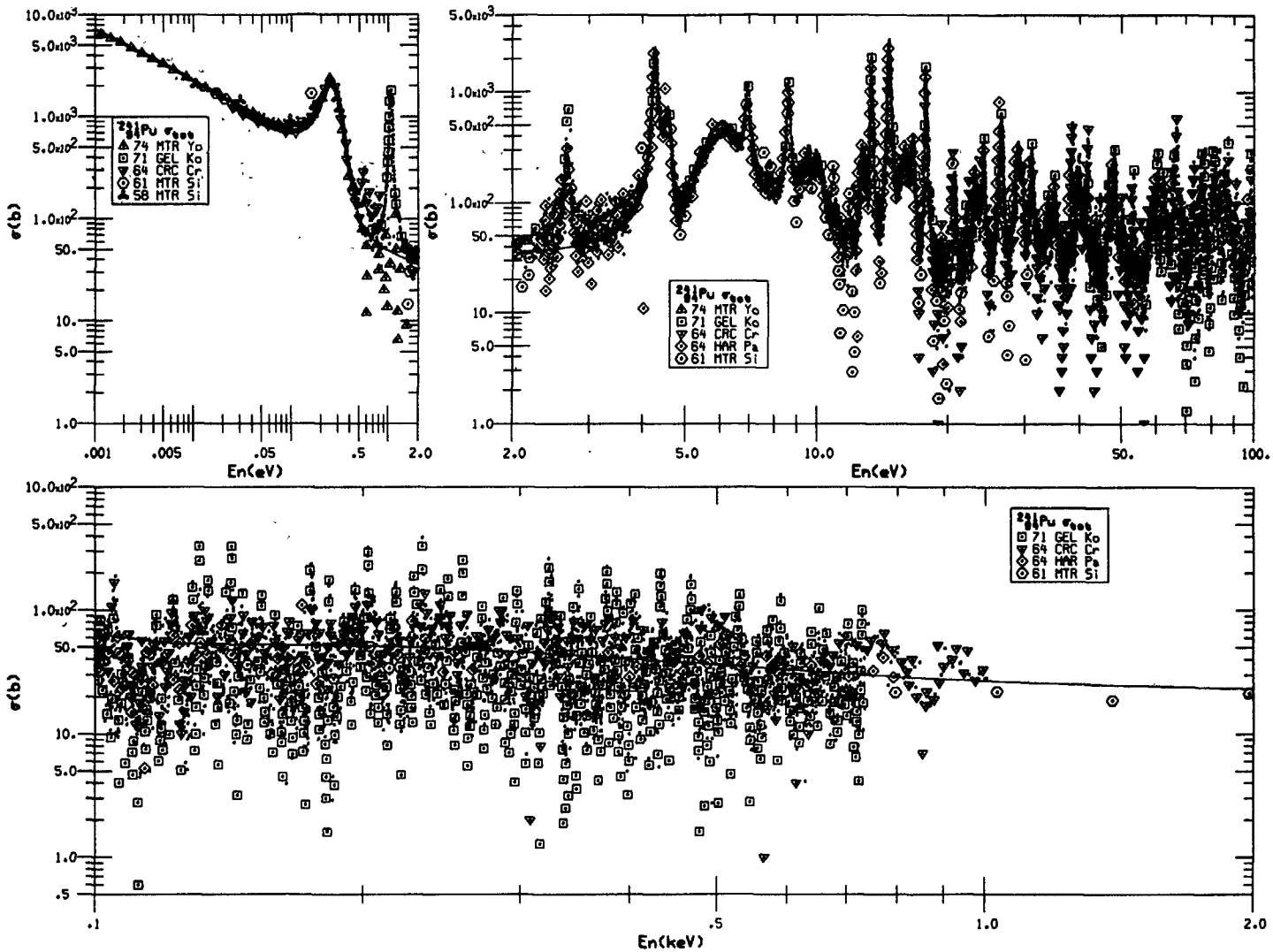
Yr	Lab	Author	Reference	Points	Range	Standard
			$^{237}_{93}Np$ σ_{tot}			
61	ORL	Slaughter,et.al.	BAP,6,70(X8),61	744	.216 eV to 41.3 eV	abs
59	CCP	Adaechuk,et.al.	AE,6,569,59	190	2.700 eV to 12.0 keV	abs
55	BNL	Palevsky,et.al.	PR,99,611(B11),55	312	.0202 eV to 2.876 eV	abs
55	CCP	Adaechuk,et.al.	55GENEVA,4,21 , ,55	88	.0084 eV to 9.700 eV	abs
			$^{237}_{93}Np$ σ_{nf}			
71	LRL	Nagle,et.al.	71KNOX,2,259,71	23	.121 MeV to 2.730 MeV	
67	ANL	Stupegia,et.al.	NSE,29,218,67	8	.152 MeV to 1.505 MeV	^{235}U σ_{nf}
			$^{237}_{93}Np$ σ_{n2n}			
73	LLL	Landrum,et.al.	PR/C,8,1938,73	5	13.8 MeV to 15.0 MeV	^{27}Al σ_{na}
61	ALD	Perkin,et.al.	JNE,14,69,61	1	.390 b at 14.0 MeV	^{27}Al σ_{na}
			$^{237}_{93}Np$ σ_{np}			
59	ALD	Coleman,et.al.	PPS,73,215,59	1	1.300 mb at 14.5 MeV	^{27}Al σ_{na}
			$^{237}_{93}Np$ σ_{nf}			
73	KTO	Kobayashi,et.al.	KOBAYASHI,73	7	3.500 MeV to 4.900 MeV	^{115}In $\sigma_{n,n'}$
73	SAC	Plattard	PLATTARD,73	3923	2.720 eV to 35.2 keV	^{10}B σ_{na}
72	LAS	Iacoletti+	NSE,48,412,72	102	.200 MeV to 7.657 MeV	
71	GEL	Kolar,et.al.	ZP,248,355,71	1171	20.1 eV to 52.1 eV	
70	LAS	Brown,et.al.	NP/A,156,609,70	2496	31.6 eV to 2.848 MeV	^{235}U σ_{nf}
69	TRM	Iyer,et.al.	69RDORKEE,2,289,69	1	2.980 b at 14.1 MeV	^{238}U σ_{nf}
67	ALD	White,et.al.	JNE,21,671,67	3	1.000 MeV to 14.1 MeV	^{235}U
65	ALD	Perkin,et.al.	JNE,19,423,65	1	11.0 mb at 24.0 keV	
65	ALD	White,et.al.	65SALZBURG,1,219,65	9	40.0 keV to .505 MeV	^{235}U
63	CCP	Pankratov	AE,14,177,63	29	2.500 MeV to 26.4 MeV	
60	CCP	Pankratov,et.al.	AE,9,399,60	17	9.600 MeV to 21.8 MeV	
59	HAN	Leonard Jr+	BAP,4,31(K1),59	200	.0412 eV to 5.250 eV	
59	ORL	Schmitt,et.al.	PR,116,1575,59	47	1.640 MeV to 7.430 MeV	
59	ORL	Schmitt,et.al.	PR,116,1575,59	74	.910 MeV to 7.430 MeV	
59	CCP	Gokhberg,et.al.	DOK,128,1157,59	25	12.0 keV to 1.500 MeV	
58	CCP	Protopopov+	AE,4,190,58	1	2.400 b at 14.6 MeV	
58	CCP	Kalinin,et.al.	58GENEVA,16,136,58	13	2.500 MeV to 8.300 MeV	
57	LAS	Henkel	LA-2122,57	52	.460 MeV to 14.0 MeV	^{237}Np σ_{nf}
47	LAS	Kiess	PR,72,88,47	16	.250 MeV to 3.000 MeV	

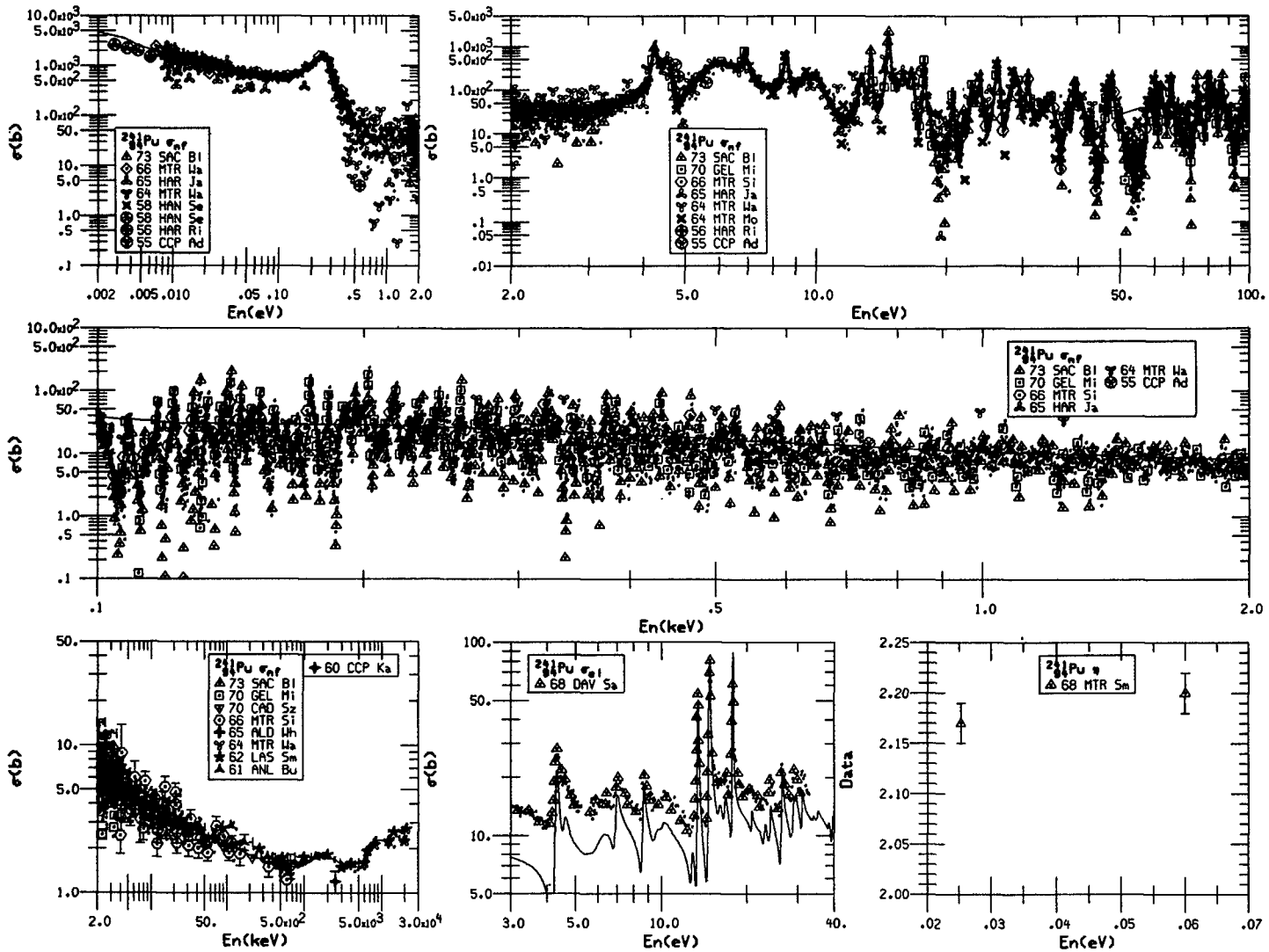


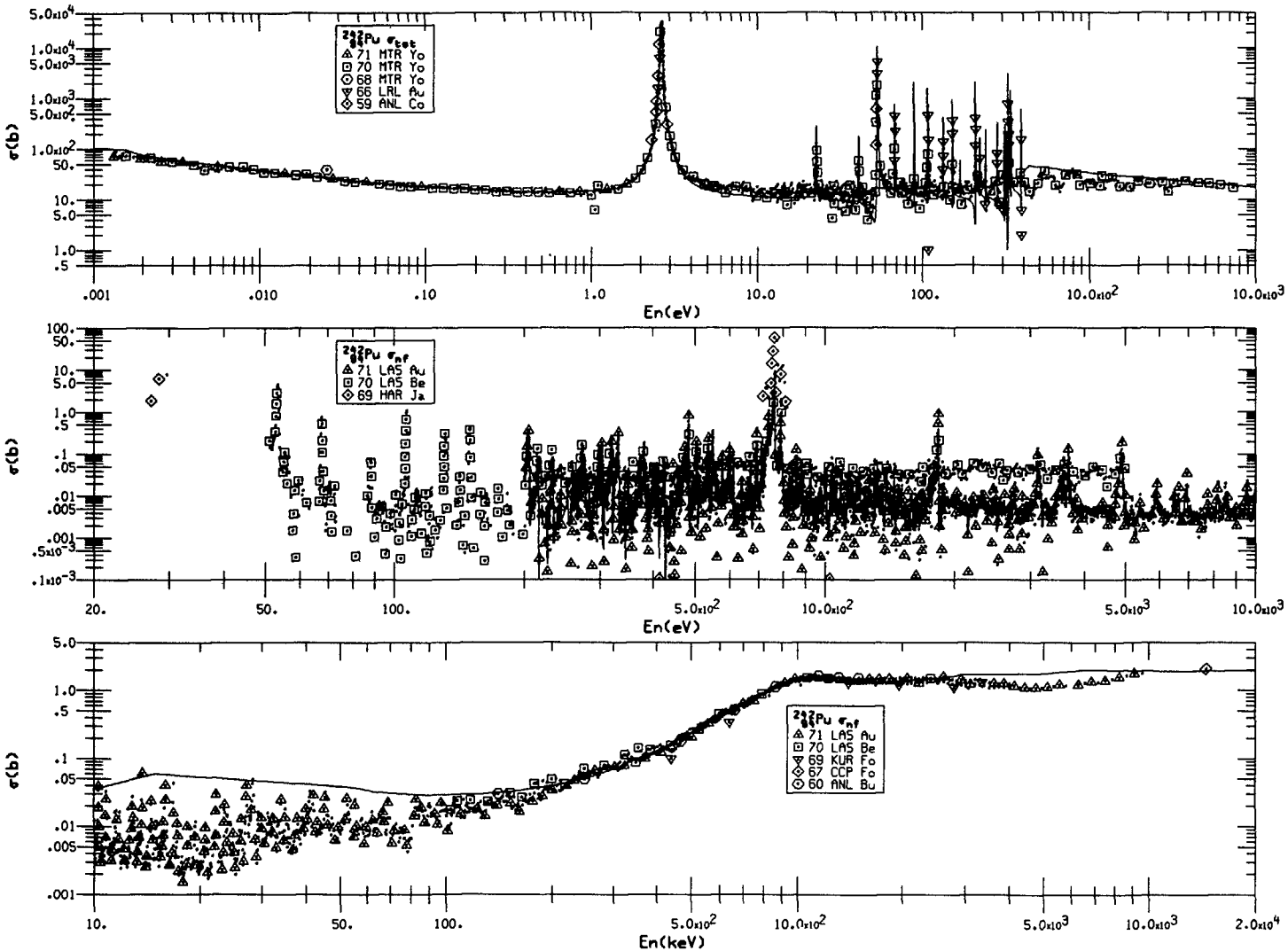

238Pu σnf
70 LAS Dr
69 LAS Si
66 KUR Ge
σ(b)
En(eV)
238Pu σnf
70 LAS Dr
69 LAS Si
66 KUR Ge
σ(b)
En(eV)
238Pu σnf
70 LAS Dr
70 CCP Er
69 LAS Si
66 KUR Vo
66 KUR Ge
σ(b)
En(keV)
238Pu σnf
70 LAS Dr
70 CCP Er
69 LAS Si
69 KUR Fo
68 CCP Er
66 KUR Vo
En(MeV)


239Pu σtot
60 ORL Bl
59 HAR Pa
58 HAR Eg
57 MTR Si
57 BNL Zi
σ(b)
En(eV)
239Pu σtot
68 GEL Ko
57 MTR Si
239Pu σtot
68 GEL Ko
En(keV)
239Pu σtot
72 ANL Sm
En(MeV)
239Pu σel
72 ANL Sm
En(MeV)


σ(b)
En(eV)
En(keV)
En(MeV)
56 HAN Le
68 GEL Mi
66 LAS By
67 MLD
66 LAS
64 MLD
60 FEI
60 CCP
57 LAS
57 CCP


σ(b)
En(eV)
En(keV)
74 MTR Yo
71 GEL Ko
64 CRC Cs
61 MTR Si
58 MTR Si
64 MAR Pa

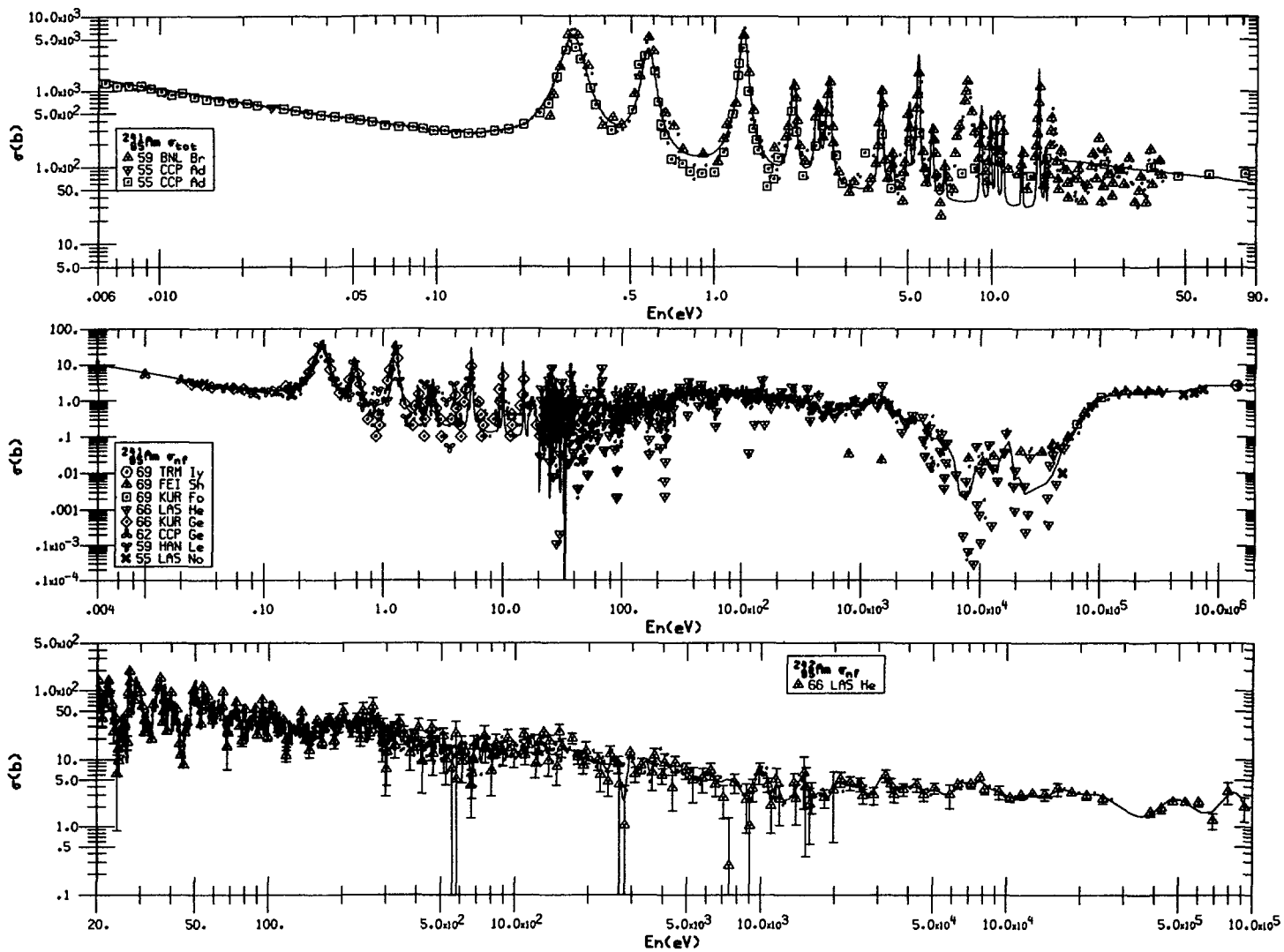

σ(b)
En(eV)
En(keV)
Data
60 CCP Ka
68 DAV Sa
68 MTR Sm

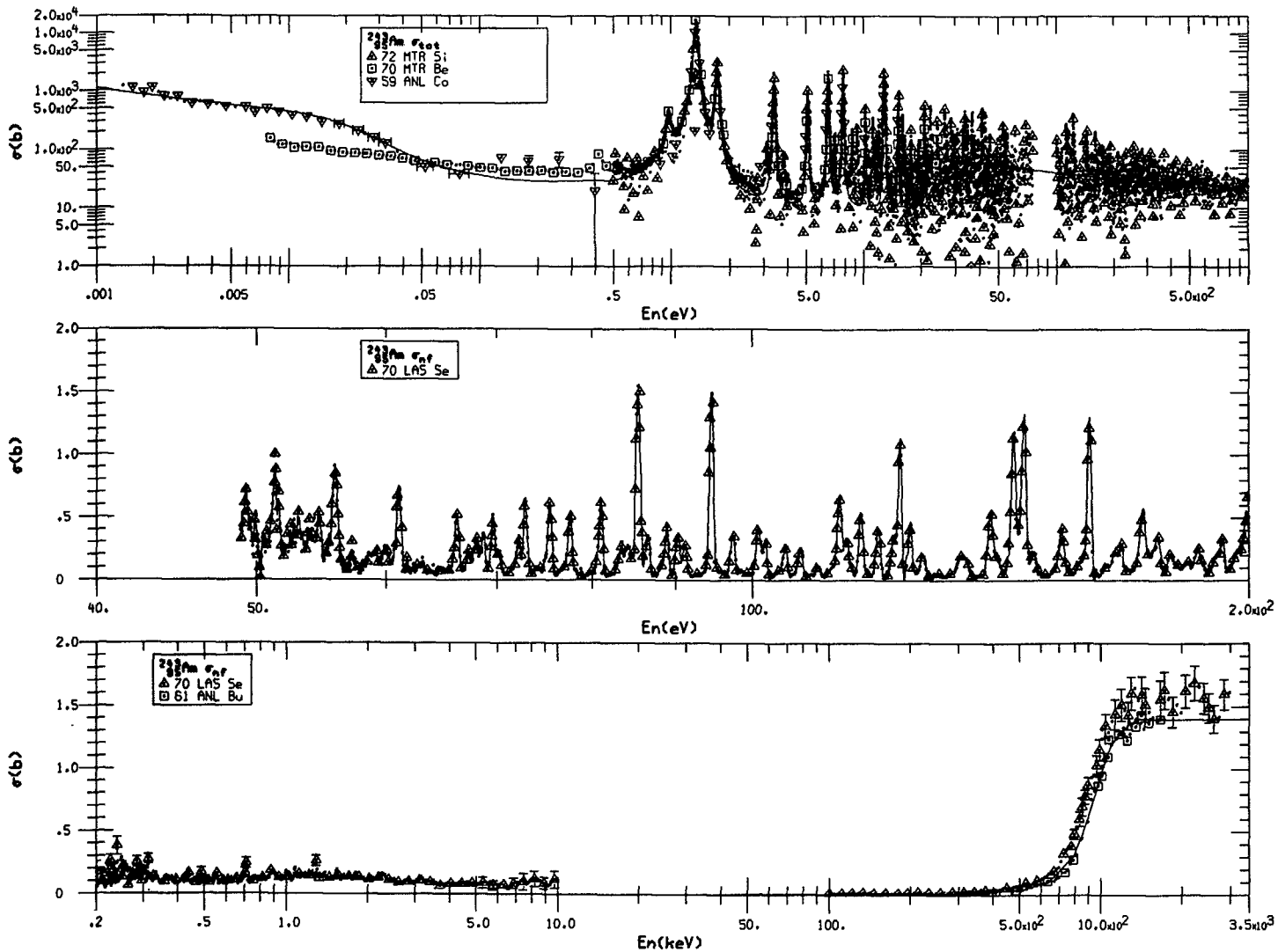

^{239}Pu σ_{tot}
71 MTR Yo
70 MTR Yo
68 MTR Yo
66 LRL Au
59 ANL Co
σ(b)
En(eV)
^{239}Pu σ_{nf}
71 LAS Au
70 LAS Be
69 HAR Ja
σ(b)
En(eV)
^{239}Pu σ_{nf}
71 LAS Au
70 LAS Be
69 KUR Fo
67 CCP Fo
60 ANL Bu
σ(b)
En(keV)

Yr	Lab	Author	Reference	Points	Range	Standard
			^{239}Pu σ_{tot}			
67	MTR	Young,et.al.	NSE,30,355,67	595	.0082 eV to 6.496 keV	abs
67	MTR	Young,et.al.	NSE,30,355,67	179	.100 keV to .200 keV	
62	BNL	Brunhart,et.al.	BAP,7,305(K16),62	1	.615 kb at thermal	
			^{239}Pu $\sigma_{n\gamma}$			
73	LAS	Silbert,et.al.	NSE,52,187,73	3505	17.8 eV to .199 MeV	
62	BNL	Brunhart,et.al.	BAP,7,305(K16),62	1	.588 kb at thermal	
57	CRC	Butler,et.al.	CJP,35,147,57	1	.410 kb at thermal	^{59}Co $\sigma_{n\gamma}$
			^{239}Pu σ_{nas}			
65	LAS	Stubbins	STUBBINS,65	1	3.760 b at 1.670 MeV	
			^{239}Pu σ_{nf}			
70	LAS	Drake,et.al.	LA-4420,101,70	2613	31.7 eV to 2.583 MeV	^{235}U σ_{nf}
70	CCP	Ermagambetov+	AE,29,(6),422,70	31	2.400 keV to 2.400 MeV	^{235}U σ_{nf}
69	LAS	Silbert	LA-4108-MS,69	6759	17.8 eV to 2.967 MeV	^{3}He σ_{np}
69	KUR	Fomushkin,et.al.	YF,10,(5),917,69	14	.440 MeV to 3.620 MeV	^{235}U σ_{nf}
68	CCP	Ermagambetov+	AE,25,527,68	20	.500 MeV to 16.9 MeV	^{235}U
67	LAS	Barton,et.al.	PR,162,1070,67	4	1.000 MeV to 14.9 MeV	^{235}U
67	CCP	Fomushkin,et.al.	YF,5,966,67	1	2.730 b at 14.5 MeV	^{238}U
66	KUR	Vorotnikov+	YF,3,(3),479,66	30	50.0 keV to 1.400 MeV	^{238}Pu σ_{nf}
66	KUR	Gerasimov	66PARIS,2,129,,66	233	.0238 eV to .420 keV	^{238}Pu σ_{nf}
			^{240}Pu σ_{tot}			
72	ANL	Smith,et.al.	NSE,47,19,72	55	.116 MeV to 1.467 MeV	abs
68	GEL	Kolar,et.al.	JNE,22,299,68	39295	19.7 eV to 5.705 keV	abs
60	ORL	Block,et.al.	NSE,8,112,60	41	.0205 eV to .152 eV	
59	HAR	Pattenden,et.al.	JNE,11,14,59	288	.0653 eV to .788 eV	
58	HAR	Egelstaff,et.al.	JNE,6,303,58	46	.627 eV to 1.372 eV	abs
57	MTR	Simpson,et.al.	BAP,2,219(S12),57	240	.0338 eV to 43.2 eV	abs
57	BNL	Zimmerman	ZIMMERMAN,57	52	.818 eV to 1.414 eV	
			^{240}Pu σ_{el}			
72	ANL	Smith,et.al.	NSE,47,19,72	30	.300 MeV to 1.500 MeV	C σ_{el}
			^{240}Pu $\sigma_{n\gamma}$			
70	CRC	Lounsbury,et.al.	70HELSINKI,1,287,70	1	.290 kb at thermal	
			^{240}Pu σ_{nf}			
68	GEC	Migneco,et.al.	NP/A,112,603,68	3290	.200 keV to 7.988 keV	^{239}Pu σ_{nf}
67	ALD	White,et.al.	JNE,21,671,67	3	1.000 MeV to 14.1 MeV	^{235}U σ_{nf}
66	LAS	Byers,et.al.	66WASH,2,903,,66	2374	20.0 eV to .978 MeV	
65	ALD	Perkin,et.al.	JNE,19,423,65	1	.106 b at 24.0 keV	
64	ALD	Ruddick,et.al.	JNE,18,561,64	13	61.0 keV to .485 MeV	
60	FEI	Nesterov,et.al.	AE,9,(1),16,60	73	40.0 keV to 3.790 MeV	^{239}Pu σ_{nf}
60	CCP	Kazarinova+	AE,8,139,60	2	2.500 MeV to 14.6 MeV	
60	CCP	Nesterov,et.al.	AE,9,16,60	76	40.0 keV to 4.200 MeV	^{239}Pu
57	LAS	Henkel,et.al.	AECD-4256,57	21	.270 MeV to 8.120 MeV	^{235}U
57	CCP	Dorofeev,et.al.	AE,2,10,57	2	.900 MeV to 5.000 MeV	
56	HAN	Leonard Jr+	BAP,1,248(C13),56	67	.305 eV to 1.298 eV	

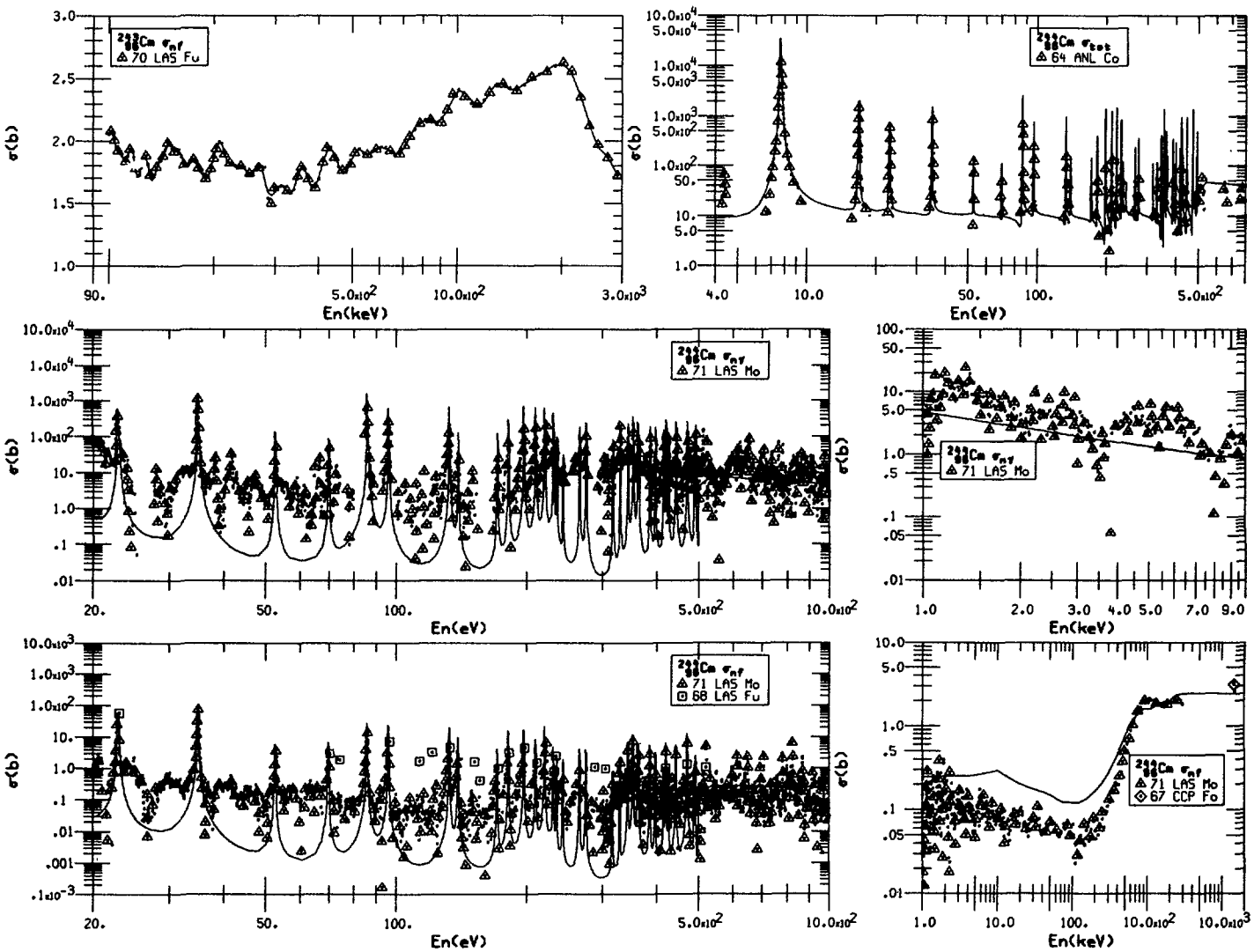
Yr	Lab	Author	Reference	Points	Range	Standard
			^{241}Pu σ_{tot}			
74	MTR	Young	YOUNG,74	374	.0010 eV to 3.554 eV	^{239}Pu ν
71	GEL	Kolar,et.al.	71KNOX,2,823,71	6996	.679 eV to .735 keV	abs
64	CRC	Craig,et.al.	CJP,42,2384,64	4438	.0247 eV to 1.005 keV	abs
64	HAR	Pattenden	PATTENDEN,64	3637	2.342 eV to .848 keV	
61	MTR	Simpson,et.al.	NSE,11,111,61	1797	.0160 eV to 1.979 keV	
58	MTR	Simpson,et.al.	BAP,3,176(F2),58	1	1.410 kb at thermal	
58	MTR	Simpson,et.al.	BAP,3,176(F2),58	44	.0105 eV to .540 eV	abs
			^{241}Pu σ_{el}			
68	DAV	Sauter,et.al.	PR,174,1413,68	299	3.040 eV to 32.8 eV	
			^{241}Pu σ_{nf}			
73	SAC	Bions	NSE,51,130,73	10340	1.596 eV to 29.8 keV	^{10}B $\sigma_{n\alpha}$
70	GEL	Migneco,et.al.	70HELSINKI,1,437,70	10430	2.722 eV to 4.043 keV	^{241}Pu σ_{nf}
70	CAD	Szabo,et.al.	70ANL,257,70	15	35.0 keV to .970 MeV	^{241}Pu σ_{nf}
66	MTR	Watanabe	IN-1012,66	256	.0063 eV to .497 eV	^{241}Pu σ_{nf}
66	MTR	Simpson,et.al.	66WAH,2,910,,66	2554	20.0 eV to .978 MeV	
66	ALD	White,et.al.	66PARIS,2,29,,66	1	1.028 kb at thermal	^{235}U σ_{nf}
66	ALD	White,et.al.	66PARIS,2,29,,66	1	1.022 kb at thermal	^{235}U σ_{nf}
66	ALD	White,et.al.	66PARIS,2,29,,66	1	.994 kb at thermal	^{235}U σ_{nf}
65	ALD	Perkin,et.al.	JNE,19,423,65	1	2.830 b at 24.0 keV	^{235}U ν
65	HAR	James	NP,65,353,65	1638	.0084 eV to 2.462 keV	
65	ALD	White,et.al.	65SALZBURG,1,219,65	4	40.0 keV to .505 MeV	^{235}U
64	MTR	Watanabe,et.al.	IDO-16995,64	223	.0236 eV to 1.882 eV	^{239}Pu
64	MTR	Watanabe,et.al.	IDO-16995,64	593	.245 eV to 2.854 keV	^{241}Pu σ_{nf}
64	MTR	Watanabe,et.al.	IDO-16995,64	15	3.118 eV to 7.264 eV	^{241}Pu σ_{nf}
64	MTR	Watanabe,et.al.	IDO-16995,64	1	.962 kb at thermal	^{241}Pu σ_{nf}
64	MTR	Moore,et.al.	PR/B,135,945,64	1018	2.247 eV to 99.5 eV	
62	MTR	Watanabe,et.al.	BAP,7,303(K4),62	1	.943 kb at thermal	
62	LAS	Smith,et.al.	PR,125,1329,62	86	.120 MeV to 21.0 MeV	^{235}U σ_{nf}
61	ANL	Butler,et.al.	PR,124,1129,61	82	.101 MeV to 1.825 MeV	^{235}U
60	CCP	Kazarinova+	AE,8,139,60	2	2.500 MeV to 14.6 MeV	
59	HAN	Leonard Jr+	HW-62727,19,59	1	.935 kb at thermal	^{235}U σ_{nf}
59	HAR	Raffle	AERE-R-2998,59	30	.0062 eV to .0975 eV	^{239}Pu
58	HAN	Seppi	SEPPI,58	62	.0257 eV to .938 eV	
58	CRC	Bigham,et.al.	58GENEVA,16,125,58	2	therm I to thermal	^{239}Pu σ_{nf}
58	HAN	Seppi,et.al.	HW-55879,3,58	20	.0025 eV to .0047 eV	^{241}Pu σ_{nf}
57	HAN	Leonard Jr+	HW-48893,98,57	1	.541 kb at .100 eV	^{197}Au $\sigma_{n\gamma}$
57	HAN	Leonard Jr+	HW-48893,98,57	1	.905 kb at thermal	^{197}Au $\sigma_{n\gamma}$
56	HAR	Richmond,et.al.	JNE,2,177,56	77	.0062 eV to 7.510 eV	^{241}Pu σ_{nf}
55	HAR	Raffle,et.al.	55GENEVA,4,187,55	1	.987 kb at thermal	^{239}Pu σ_{nf}
55	HAR	Egelstaff,et.al.	NRDC-84,9,55	1	.971 kb at thermal	^{239}Pu σ_{nf}
55	CCP	Adamchuk,et.al.	55GENEVA,4,216,55	157	.0117 eV to .743 keV	^{241}Pu σ_{nf}
			^{241}Pu α			
66	HAR	Cabell	66PARIS,2,3,,66	1	1355 b at thermal	
			^{241}Pu η			
68	MTR	Smith,et.al.	68WASH,1,589,68	2	therm I to .0600 eV	

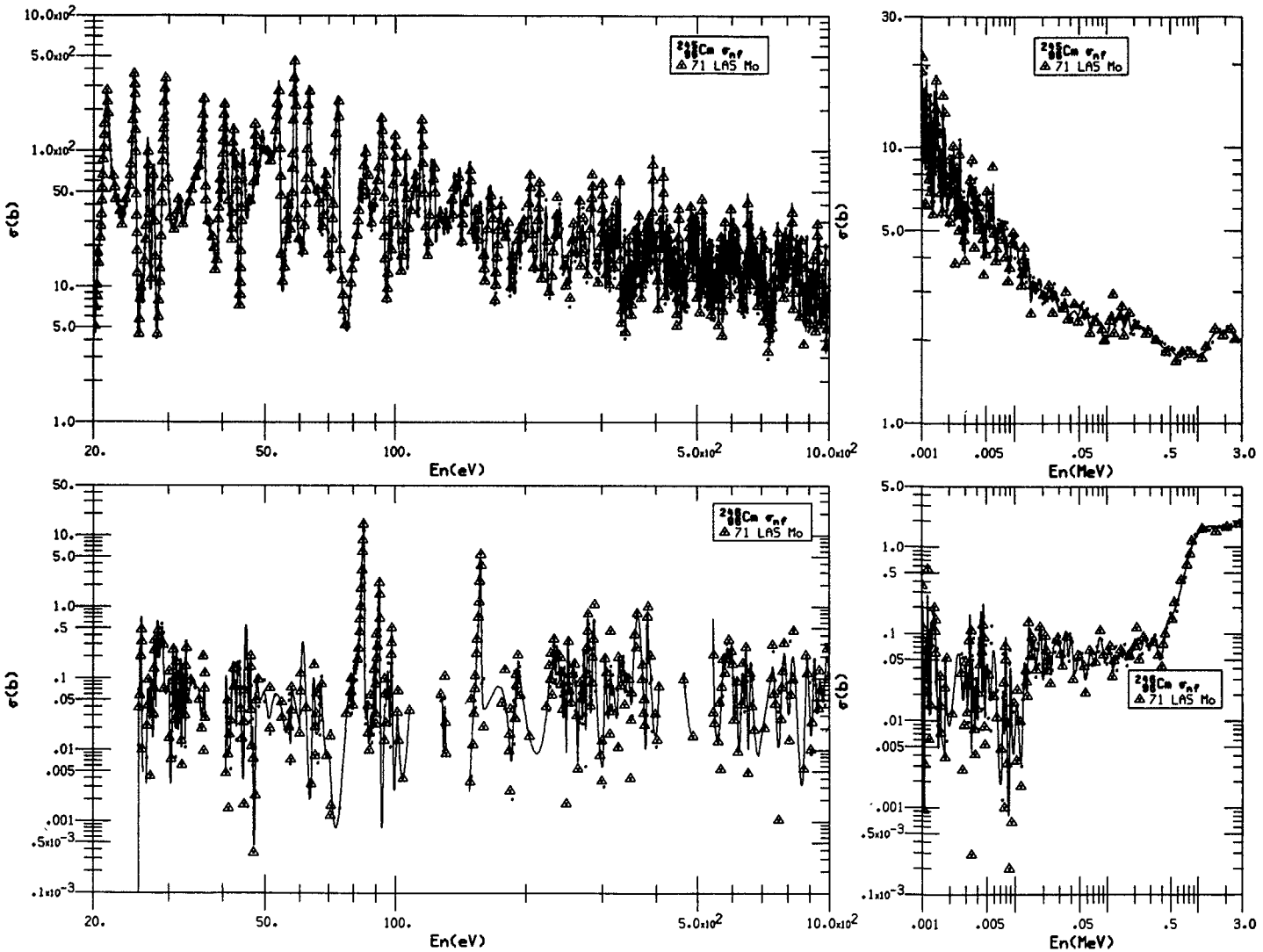
Yr	Lab	Author	Reference	Points	Range	Standard
			$^{239}_{94}Pu\ \sigma_{tot}$			
71	MTR	Young,et.al.	NSE,43,341,71	79	.0013 eV to 2.006 eV	abs
70	MTR	Young,et.al.	NSE,40,389,70	1017	.0013 eV to 7.726 keV	
68	MTR	Young,et.al.	IN-1132,68	1	39.0 b at thermal	
66	LRL	Auchampaugh+	PR,146,840,66	182	2.550 eV to .388 keV	abs
66	MTR	Young,et.al.	BAP,11,335(DBS),66	1	39.8 b at thermal	
59	ANL	Cote,et.al.	PR,114,505,59	50	2.336 eV to 54.7 eV	abs
			$^{239}_{94}Pu\ \sigma_{n\gamma}$			
57	CRC	Butler,et.al.	CJP,35,147,57	1	19.8 b at thermal	$^{59}Co\ \sigma_{n\gamma}$
56	ANL	Fields,et.al.	NSE,1,62,56	1	30.0 b at thermal	
			$^{239}_{94}Pu\ \sigma_{nf}$			
71	LAS	Auchampaugh+	NP/A,171,31,71	6601	.200 keV to 9.573 MeV	$^{6}Li\ \sigma_{nt}$
70	LAS	Bergen,et.al.	LA-4420,123,70	1697	51.2 eV to 2.963 MeV	$^{235}U\ \sigma_{nf}$
69	KUR	Fomushkin,et.al.	YF,10,(5),917,69	14	.440 MeV to 3.620 MeV	$^{235}U\ \sigma_{nf}$
69	HAR	James	NP/A,123,24,69	24	27.3 eV to .815 keV	
67	CCP	Fomushkin,et.al.	YF,5,966,67	1	2.060 b at 14.5 MeV	^{238}U
60	ANL	Butler	PR,117,1305,60	65	.141 MeV to 1.663 MeV	$^{235}U\ \sigma_{nf}$

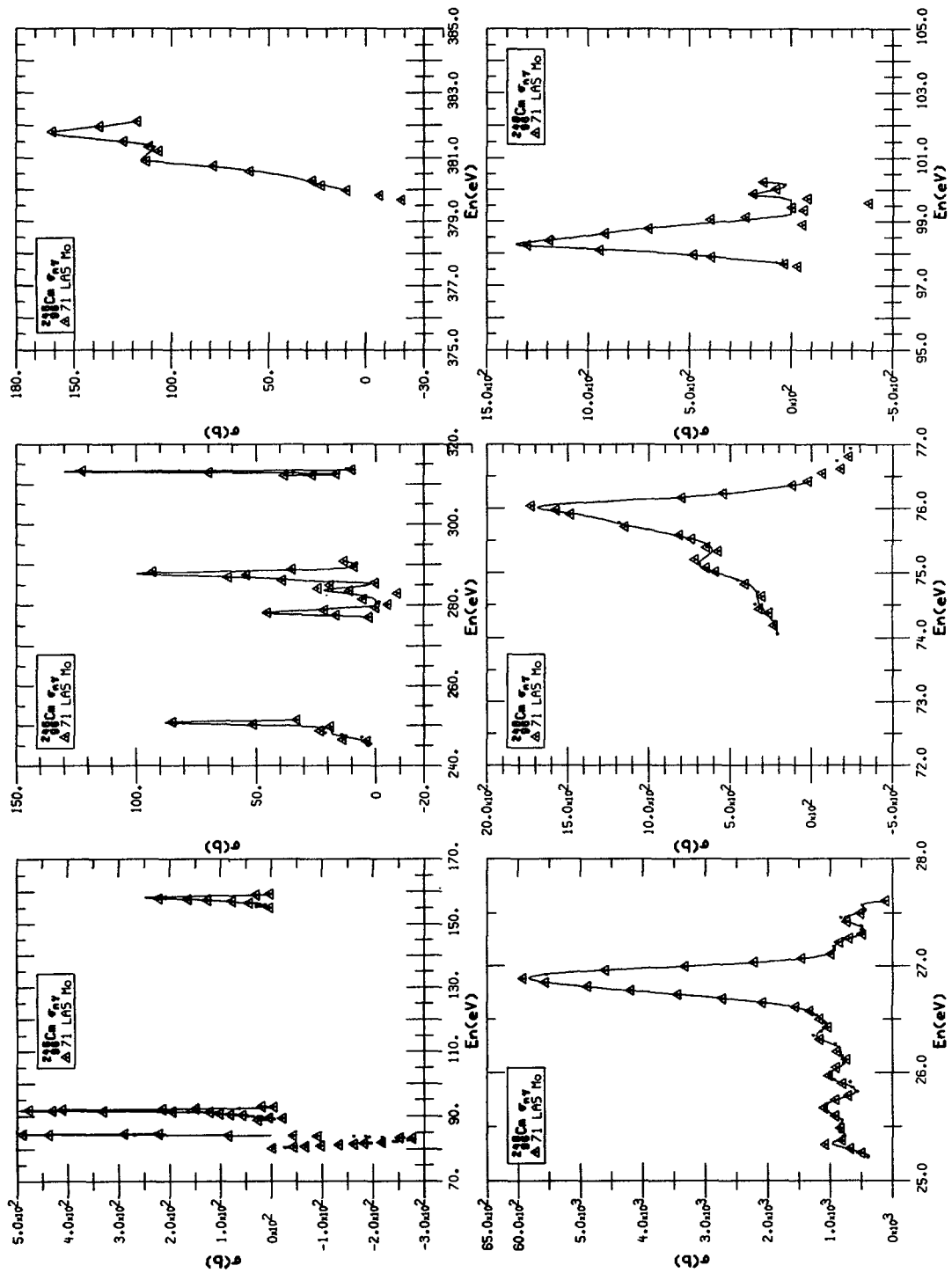


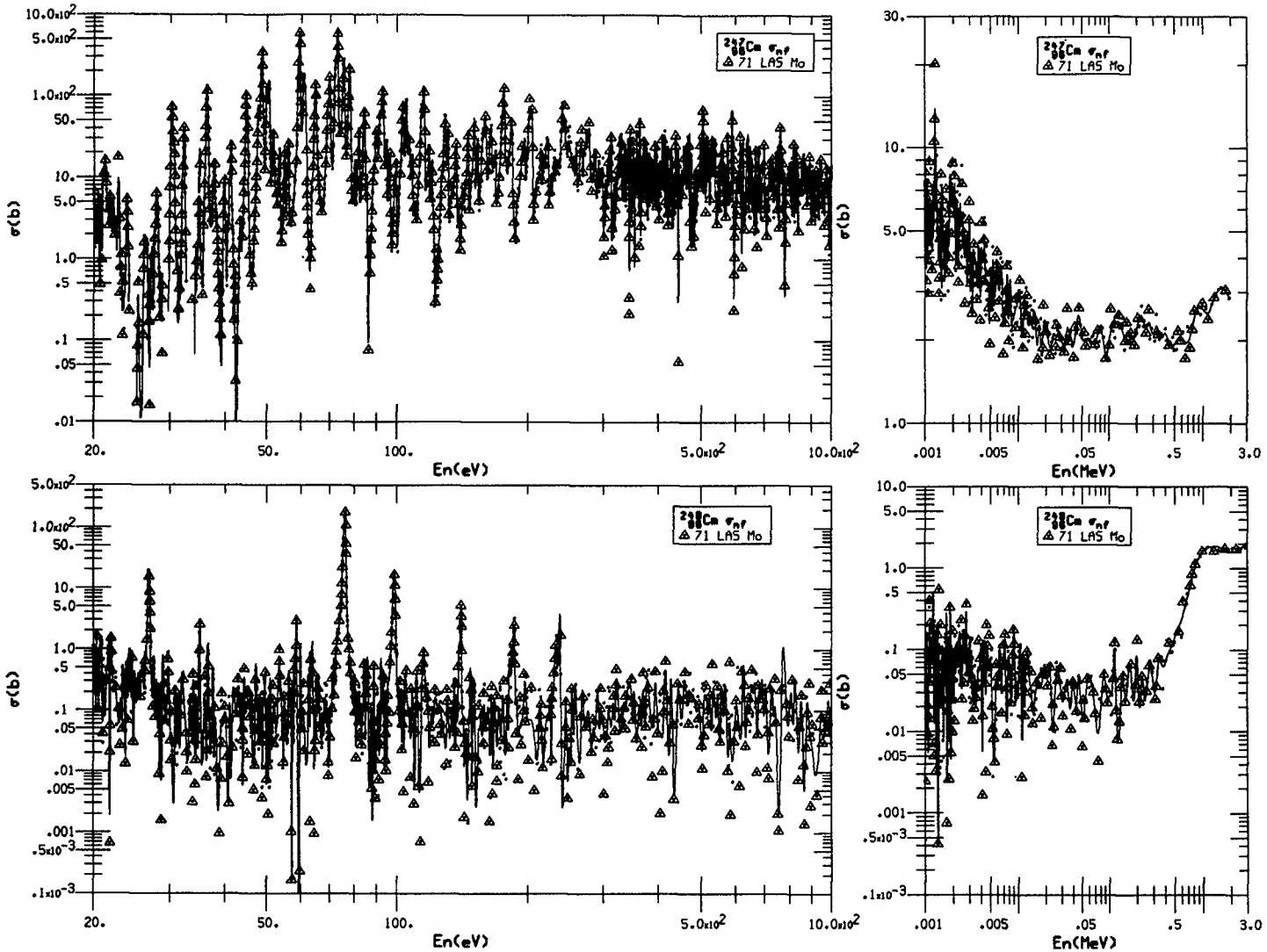

σ(b)
En(eV)
72 MTR Si
70 MTR Be
59 ANL Co
70 LAS Se
61 ANL Bu
En(keV)

Yr	Lab	Author	Reference	Points	Range			Standard
			$^{233}_{95}Am$ σ_{tot}					
59	BNL	Brussel,et.al.	BAP,4,34(M9),59	454	.255 eV	to	41.7 eV	abs
55	CCP	Adamchuk,et.al.	55GENEVA,4,216,55	1	.600 kb	at	thermal	
55	CCP	Adamchuk,et.al.	55GENEVA,4,216,55	199	.0061 eV	to	82.1 eV	abs
			$^{233}_{95}Am$ σ_{nf}					
69	TRM	Iyer,et.al.	69ROORKEE,2,289,69	1	2.700 b	at	14.1 MeV	
69	FEI	Shpak,et.al.	YF1-8,4,69	43	8.000 keV	to	3.300 MeV	^{239}Pu σ_{nf}
69	KUR	Fomushkin,et.al.	YF,10(5),917,69	14	.440 MeV	to	3.620 MeV	^{235}U σ_{nf}
67	CCP	Fomushkin,et.al.	YF,5,966,67	1	2.300 b	at	14.5 MeV	^{238}U
67	CCP	Fomushkin,et.al.	YF,5,966,67	1	2.530 b	at	14.5 MeV	^{238}U
66	KYU	Hyakutake	HYAKUTAKE,66	1	3.800 b	at	.0250 eV	^{235}U σ_{nf}
66	LAS	Hemmendinger	HEMMINDINGER,66	2463	20.0 eV	to	.978 MeV	
66	KUR	Gerasimov	66PARIS,2,129,66	588	.0231 eV	to	50.3 eV	^{241}Am σ_{nf}
65	LRL	Bowman,et.al.	PR/B,137,326,65	483	.0321 eV	to	5.226 keV	^{241}Am σ_{nf}
62	CCP	Gerasimov	IFG,8,75,62	15	.0040 eV	to	.420 eV	abs
60	CCP	Kazarinova+	AE,8,139,60	2	2.500 MeV	to	14.6 MeV	
59	HAN	Leonard Jr+	BAP,4,31(K1),59	110	.0309 eV	to	5.250 eV	
59	CCP	Protopopov+	AE,6,67,59	1	2.350 b	at	14.6 MeV	
55	LAS	Nobles,et.al.	PR,99,616(CA2),55	13	.490 MeV	to	7.340 MeV	^{235}U
			$^{233}_{95}Am$ σ_{nf}					
66	LAS	Hemmendinger	HEMMINDINGER,66	1860	20.0 eV	to	.929 MeV	
			$^{233}_{95}Am$ σ_{tot}					
72	MTR	Simpson,et.al.	ANCR-1060,72	6633	.500 eV	to	1.003 keV	abs
70	MTR	Berreth,et.al.	IN-1407,66,70	923	.0080 eV	to	25.6 eV	
59	ANL	Cote,et.al.	PR,114,505,59	194	.0014 eV	to	15.4 eV	abs
59	ANL	Cote,et.al.	PR,114,505,59	1	.190 kb	at	thermal	
59	ANL	Cote,et.al.	PR,114,505,59	6	.100 eV	to	.630 eV	abs
			$^{233}_{95}Am$ $\sigma_{n\gamma}$					
57	CRC	Butler,et.al.	CJP,35,147,57	1	68.5 b	at	thermal	^{59}Co $\sigma_{n\gamma}$
			$^{233}_{95}Am$ σ_{nf}					
70	LAS	Seeger	LA-4420,138,70	2199	48.9 eV	to	2.973 MeV	^{235}U σ_{nf}
67	CCP	Fomushkin,et.al.	YF,5,966,67	1	2.290 b	at	14.5 MeV	^{238}U
67	CCP	Fomushkin,et.al.	YF,5,966,67	1	2.150 b	at	14.5 MeV	^{238}U
61	ANL	Butler,et.al.	PR,124,1129,61	65	.300 MeV	to	1.670 MeV	^{235}U




^{238}Cu σ_{nf}
71 LAS Mo
σ(b)
En(eV)
En(MeV)

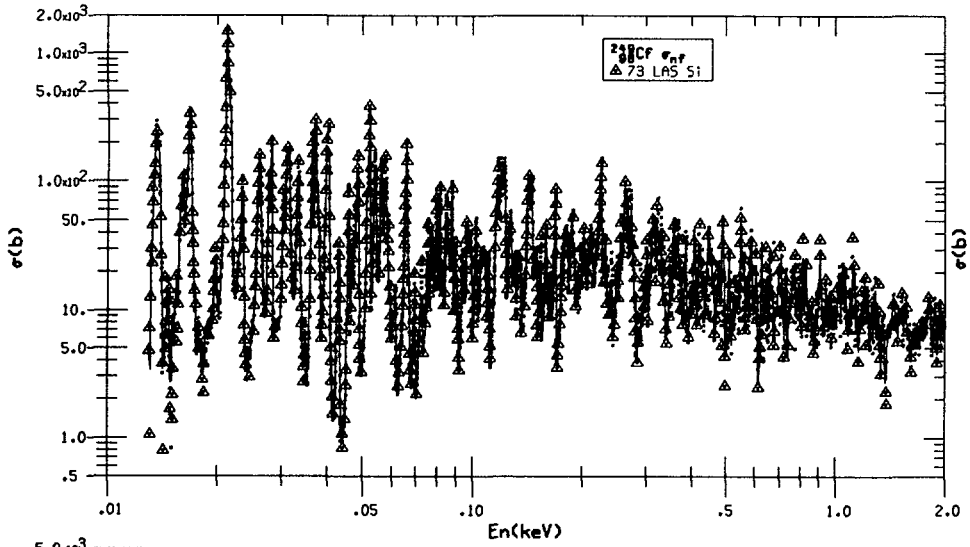

σ(b)
En(eV)
238U σn,γ
Δ71 LAS Mo

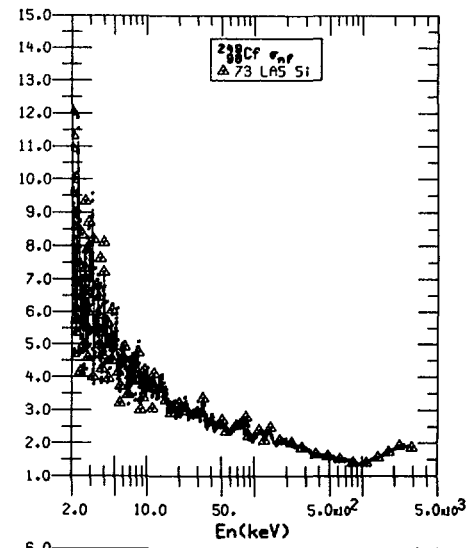

$^{247}_{96}$Cm σ_{nf}
△ 71 LAS Mo
σ(b)
En(eV)
En(MeV)
20.
50.
100.
.001
.005
.05
.5
3.0

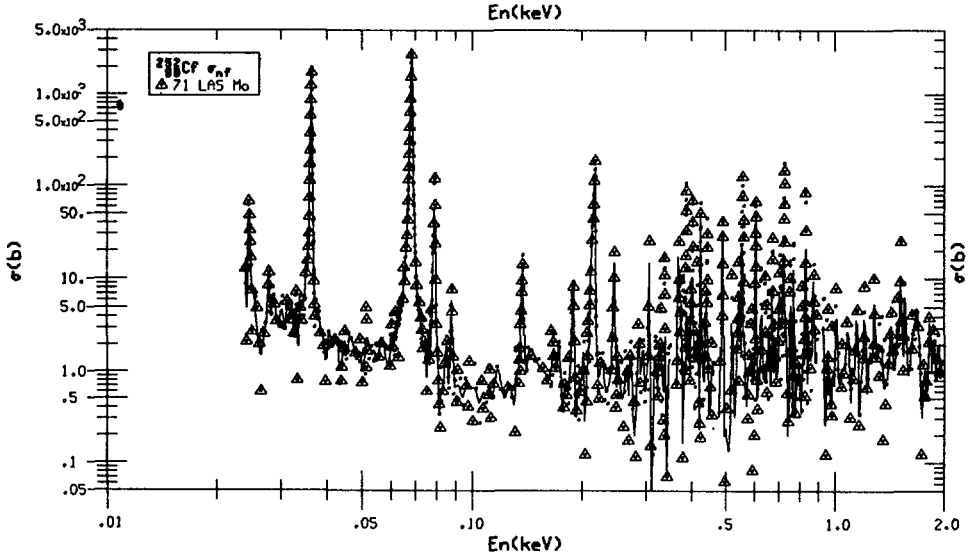
Yr	Lab	Author	Reference	Points	Range	Standard
			$^{233}_{96}Cm\ \sigma_{nf}$			
67	CCP	Fomushkin,et.al.	YF,5,966,67	1	3.030 b at 14.5 MeV	
			$^{233}_{96}Cm\ \sigma_{nf}$			
70	LAS	Fullwood,et.al.	LA-4420.157,70	134	99.7 keV to 2.910 MeV	$^{235}U\ \sigma_{nf}$
			$^{233}_{96}Cm\ \sigma_{tot}$			
72	MTR	Berreth,et.al.	NSE,49,145,72	1	23.0 b at thermal	
64	ANL	Cote,et.al.	PR/B,134,1047,64	328	4.273 eV to .780 keV	abs
			$^{233}_{96}Cm\ \sigma_{nγ}$			
71	LAS	Moore,et.al.	PR/C,3,1656,71	3791	20.0 eV to 9.951 keV	$^{6}Li\ \sigma_{nt}$
			$^{234}_{96}Cm\ \sigma_{nf}$			
72	SRL	Benjamin,et.al.	NSE,47,203,72	1	1.100 b at .0250 eV	
71	LAS	Moore,et.al.	PR/C,3,1656,71	4311	20.0 eV to 2.830 MeV	$^{6}Li\ \sigma_{nt}$
68	LAS	Fullwood,et.al.	68WASH,1,567,,68	43	23.0 eV to 1.409 keV	$^{6}Li\ \sigma_{nα}$
68	LAS	Koontz,et.al.	68WASH,1,597,,68	4	1.000 MeV to 14.9 MeV	^{235}U
67	CCP	Fomushkin,et.al.	YF,5,966,67	1	3.280 b at 14.5 MeV	
67	CCP	Fomushkin,et.al.	YF,5,966,67	1	3.100 b at 14.5 MeV	
			$^{234}_{96}Cm\ \sigma_{tot}$			
72	MTR	Berreth,et.al.	NSE,49,145,72	1	2.900 kb at thermal	
			$^{234}_{96}Cm\ \sigma_{nγ}$			
69	ORL	Halperin,et.al.	ORNL-4437,69	1	.340 kb at .0250 eV	
			$^{234}_{96}Cm\ \sigma_{nf}$			
72	SRL	Benjamin,et.al.	NSE,47,203,72	1	2.018 kb at .0250 eV	
71	LAS	Moore,et.al.	PR/C,3,1656,71	4301	20.0 eV to 2.831 MeV	$^{6}Li\ \sigma_{nt}$
70	ORL	Halperin,et.al.	ORNL-4581,37,70	1	1.920 kb at .0250 eV	$^{197}Au\ \sigma_{nγ}$
			$^{235}_{96}Cm\ \sigma_{nγ}$			
71	LAS	Moore,et.al.	PR/C,3,1656,71	157	80.1 eV to .382 keV	$^{6}Li\ \sigma_{nt}$
69	ORL	Halperin,et.al.	ORNL-4437,69	1	1.200 b at .0250 eV	
			$^{235}_{96}Cm\ \sigma_{nf}$			
72	SRL	Benjamin,et.al.	NSE,47,203,72	1	.170 b at .0250 eV	
71	LAS	Moore,et.al.	PR/C,3,1656,71	2797	20.0 eV to 2.830 MeV	$^{6}Li\ \sigma_{nt}$

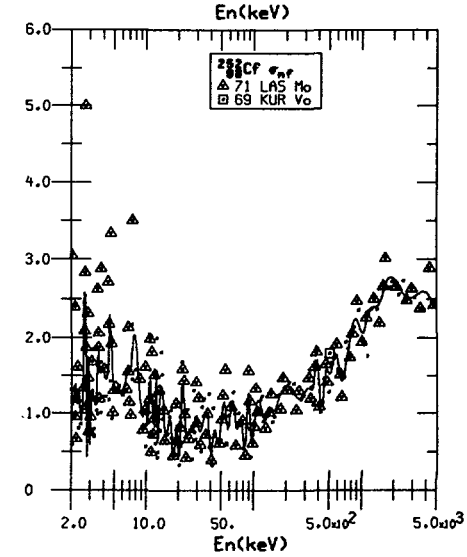
Yr	Lab	Author	Reference	Points	Range	Standard
			$^{237}_{96}Cm\ \sigma_{nf}$			
72	SRL	Benjamin,et.al.	NSE,47,203,72	1	82.0 b at .0250 eV	
71	LAS	Moore,et.al.	PR/C,3,1656,71	4296	20.0 eV to 1.908 MeV	$^{6}Li\ \sigma_{nt}$
70	ORL	Halperin,et.al.	ORNL-4581,37,70	1	.120 kb at .0250 eV	$^{197}Au\ \sigma_{nγ}$
			$^{243}_{96}Cm\ \sigma_{nγ}$			
71	LAS	Moore,et.al.	PR/C,3,1656,71	119	25.2 eV to .100 keV	$^{6}Li\ \sigma_{nt}$
			$^{243}_{96}Cm\ \sigma_{nf}$			
72	SRL	Benjamin,et.al.	NSE,47,203,72	1	.340 b at .0250 eV	
71	LAS	Moore,et.al.	PR/C,3,1656,71	2795	20.0 eV to 2.831 MeV	$^{6}Li\ \sigma_{nt}$

2.0
1.0
.5
.1
.05
.01
.005
.001
σ(b)
.5
1.0
1.5
2.0
En(MeV)
249Bk σnf
△ 69 KUR Vo









Yr Lab	Author	Reference	Points	Range	Standard
		^{249}Cf σ_{nf}			
71 ORL	Halperin,et.al.	ORNL-4706,47,71	1	.478 kb at .0250 eV	
		^{251}Cf σ_{nf}			
73 LAS	Silbert,et.al.	NSE,51,376,73	5170	13.0 eV to 2.897 MeV	
72 SRL	Benjamin,et.al.	NSE,47,203,72	1	1.660 kb at .0250 eV	
70 ORL	Halperin,et.al.	ORNL-4581,37,70	1	1.690 kb at .0250 eV	
		^{251}Cf σ_{nf}			
71 ORL	Halperin,et.al.	ORNL-4706,47,71	1	2.030 kb at .0250 eV	
		^{251}Cf σ_{nf}			
71 ORL	Halperin,et.al.	ORNL-4706,47,71	1	2.850 kb at .0250 eV	
		^{251}Cf σ_{nf}			
69 ORL	Halperin,et.al.	NSE 37,228,69	1	20.4 b at .0250 eV	
		^{252}Cf σ_{nf}			
71 ORL	Halperin,et.al.	ORNL-4706,71	1	32.0 b at .0250 eV	
71 LAS	Moore,et.al.	PR/C,4,273,71	1396	24.0 eV to 4.681 MeV	^{6}Li σ_{nt}
69 KUR	Vorotnikov+	YF,10,726,69	1	1.800 b at .500 MeV	

Yr Lab	Author	Reference	Points	Range	Standard
		^{238}U σ_{nf}			
69 KUR	Vorotnikov+	YF,10,726,69	11	.640 MeV to 5.000 MeV	

APPENDIX B

Nuclear and Atomic Radiation Data for Applications

There exist numerous applications which require knowledge of the nuclear and atomic radiations that follow the decay of radioisotopes. Tabulated here is a sample of such data sets which are available from the Nuclear Data Project (NDP). This particular sample was requested by the Environmental Sciences Division at ORNL for an intercomparative study of the effects of effluents from fossil-fueled and nuclear-fueled power generators.

The NDP Evaluated Nuclear Structure Data File (ENSDF) [39] contains evaluated nuclear structure information taken from the Nuclear Data Sheets as well as from other published data evaluations. The file is often updated with such important new data as may have become available since the most recent published evaluation. From the ENSDF data sets and existing computerized tabulations of relevant Z-dependent constants (e.g., fluorescence yields, electron binding energies, X-ray energies, etc.), the computer program MEDLIST prepares tabulations (or tapes) of the energies and intensities of all radiations involved in a specific decay, including their uncertainties. The listed radiations include Auger and conversion electrons, α-, β-, γ-, and X-rays. In the case of a β-transition, the average β-energy is also given, calculated from the β end-point energy and the forbiddenness of the β-transition. A low-intensity cutoff limit is built into the program and can be chosen to fit the users' needs. The MEDLIST program also calculates the equilibrium dose constant, Δ, which is described fully in Reference [30].

The next four pages contain a list of the radiations from ^{231}Pa + daughters [40]. The last page is an index to a selected list of radioactive decay data sets which have been prepared by the members of the Nuclear Data Project in response to user requests.

231PA A DECAY (3.248E4 Y 11) I(min)=0.10%

Radiation Type	Energy (keV)	Intensity (%)	Δ(g-rad/ μCi-h)
ce-L- 10	3.760 18	0.76 17	≈0
ce-L- 12	5.70 7	14 3	0.0017
ce-L- 14	7.520 21	33 11	0.0052
Auger-L	9.28	44 8	0.0086
ce-L- 15	10.11 3	19 5	0.0042
ce-M- 5	11.50 10	1.72 25	0.0004
ce-M- 7	13.898 5	30.3 13	0.0090
ce-NOP- 5	15.23 10	0.58 9	0.0002
ce-NOP- 7	17.631 5	9.3 9	0.0035
ce-L- 20	18.36 3	8.5 20	0.0033
ce-M- 10	18.598 5	0.20 5	≈0
ce-M- 12	20.54 6	3.2 7	0.0014
ce-M- 14	22.358 12	8 3	0.0039
ce-NOP-12	24.27 6	0.99 22	0.0005
ce-L- 25	24.32 3	1.8 5	0.0009
ce-M- 15	24.948 21	4.8 12	0.0026
ce-NOP-14	26.091 12	2.4 9	0.0013
ce-L- 26	26.53 3	0.14 3	≈0
ce-NOP-15	28.681 21	1.5 4	0.0009
ce-L- 28	32.90 3	1.5 4	0.0011
ce-M- 20	33.198 21	2.1 5	0.0015
ce-NOP-20	36.931 21	0.63 15	0.0005
ce-L- 31	37.35 4	4.3 10	0.0034
ce-M- 25	39.158 21	0.43 11	0.0004
ce-NOP-25	42.891 21	0.13 4	0.0001
ce-L- 33	43.83 4	3.3 8	0.0031
ce-M- 28	47.738 21	0.37 9	0.0004
ce-NOP-28	51.471 21	0.13 3	0.0001
ce-M- 31	52.19 3	1.2 3	0.0013
ce-L- 37	54.34 5	0.80 20	0.0009
ce-NOP-31	55.92 3	0.43 11	0.0005
ce-L- 38	57.52 4	0.39 10	0.0005
ce-M- 33	58.67 3	0.90 21	0.0011
ce-NOP-33	62.40 3	0.32 8	0.0004
ce-M- 37	69.18 4	0.21 6	0.0003
ce-L- 41	77.04 4	0.78 18	0.0013
ce-L- 42	81.08 5	0.24 7	0.0004
ce-L- 43	82.7 4	0.30 13	0.0005
ce-M- 41	91.88 3	0.21 5	0.0004
α 1	4631.0 20	0.10	0.0099
α 2	4642.0 20	0.10	0.0099
α 3	4680.0 20	1.5	0.150
α 4	4712.0 20	1	0.100
α 5	4736.0 20	8.4	0.847
α 6	4851.0 20	1.4	0.145
α 7	4933.0 20	3	0.315
α 8	4950	22.8	2.40
α 9	4974.0 20	0.4	0.0424
α 10	4984.0 20	1.4	0.149
α 11	5011.0 20	25.4	2.71
α 12	5028	20	2.14
α 13	5030.5 20	2.5	0.268
α 14	5057.3 20	11	1.18

6 weak α's omitted (ΣIα = 0.07%)

231PA A DECAY (3.248E4 Y 11) I(min)=0.10%

Radiation Type	Energy (keV)	Intensity (%)	Δ(g-rad/ μCi-h)
X-ray L	12.7	46 8	0.0123
γ 14	27.360 10	9.3 19	0.0054
γ 16	31.00 5	0.9 5	0.0006
γ 17	31.54 5	0.70 20	0.0005
γ 20	38.200 20	0.15 4	0.0001
γ 21	39.57 4	0.14 7	0.0001
γ 22	39.970 20	1.20 20	0.0010
γ 23	42.48 5	0.60 10	0.0005
γ 24	43.05 5	0.70 20	0.0006
γ 26	46.370 20	0.21 5	0.0002
γ 30	56.76 4	0.60 10	0.0007
γ 34	70.50 5	0.70 10	0.0011
γ 48	242.20 10	0.80 10	0.0041
γ 52	255.80 7	0.101 21	0.0006
γ 54	260.22 8	0.17 4	0.0010
γ 57	283.67 6	1.6 4	0.0097
γ 59	300.08 6	2.3 5	0.0147
γ 60	302.67 6	2.3 5	0.0148
γ 61	302.67 6	2.3 5	0.0148
γ 62	310.15 10	0.14 5	0.0009
γ 64	318.1 7	0.20 10	0.0014
γ 66	330	1.3 4	0.0091
γ 67	340.81 7	0.16 4	0.0012
γ 70	357.16 7	0.17 4	0.0013

71 weak γ's omitted (ΣIγ = 1.34%)

227AC B- DECAY (21.773 Y 3) I(min)=0.10%

Radiation Type	Energy (keV)	Intensity (%)	Δ(g-rad/ μCi-h)
ce-L- 3	4.03 20	0.78 16	≈0
ce-M- 1	4.12 10	33 7	0.0029
ce-NOP- 1	7.97 10	10.8 23	0.0018
Auger-L	9.48	0.37 9	≈0
ce-M- 2	10.02 10	6.7 15	0.0014
ce-NOP- 2	13.87 10	2.2 5	0.0007
ce-M- 3	19.32 20	0.17 4	≈0
β- 1 max	19.1 20		
avg	4.8 5	10	0.0010
β- 2 max	34.3 20		
avg	8.7 6	35	0.0065
β- 3 max	43.6 20		
avg	11.1 6	54	0.0128
total β- avg	9.6 7	99	0.0203
X-ray L	13	0.41 9	0.0001

3 weak γ's omitted (ΣIγ = 0.06%)

227TH A DECAY (18.718 D 5) I(min)=0.10%

Radiation Type	Energy (keV)	Intensity (%)	Δ(g-rad/ μCi-h)
ce-L- 5	1.03 10	1.2 5	≈0
ce-M- 2	3.18 20	2.6 6	0.0002
ce-NOP- 2	6.79 20	0.85 17	0.0001
Auger-L	9	43 9	0.0083
ce-K- 54	9.28 6	0.22 5	≈0
ce-K- 52	9.278 8	0.16 4	≈0
ce-L- 8	10.34 10	0.53 11	0.0001
ce-L- 9	10.67 3	41 15	0.0093
ce-L- 10	12.383 11	15 5	0.0041
ce-M- 5	15.45 10	0.30 11	≈0
ce-L- 15	24.56 10	0.18 5	≈0
ce-M- 8	24.76 10	0.13 3	≈0
ce-L- 16	24.88 10	0.7 3	0.0003
ce-M- 9	25.09 3	11 4	0.0057
ce-L- 18	25.19 3	0.35 18	0.0002
ce-M- 10	26.798 11	4.1 13	0.0023
ce-NOP- 9	28.70 3	3.5 13	0.0022
ce-L- 20	29.06 10	2.3 6	0.0014
ce-NOP-10	30.412 11	1.3 5	0.0009
ce-L- 21	30.64 10	0.11 9	≈0
ce-L- 22	30.96 10	4.5 10	0.0030
ce-L- 23	31.64 10	0.7 4	0.0005
ce-L- 25	34.9633 15	0.119 24	≈0
ce-L- 27	37.31 10	0.19 4	0.0002
ce-M- 16	39.30 10	0.17 9	0.0001
ce-L- 28	42.27 10	5.7 19	0.0052
ce-L- 31	43.5 3	0.11 6	0.0001
ce-M- 20	43.48 10	0.62 14	0.0006
ce-M- 22	45.38 10	1.10 23	0.0011
ce-M- 23	46.06 10	0.17 9	0.0002
ce-NOP-20	47.09 10	0.21 5	0.0002
ce-NOP-22	48.99 10	0.37 8	0.0004
ce-L- 34	49.56 10	0.53 17	0.0006
ce-L- 37	54.46 10	0.50 13	0.0006
ce-M- 28	56.69 10	1.5 5	0.0019
ce-NOP-28	60.30 10	0.56 18	0.0007
ce-L- 39	60.53 6	0.32 7	0.0004
ce-M- 34	63.98 10	0.13 5	0.0002
Auger-K	65.9	0.16 10	0.0002
ce-M- 37	68.88 10	0.14 4	0.0002
ce-L- 41	74.76 6	0.14 3	0.0002
ce-L- 42	75.76 10	0.11 3	0.0002
ce-L- 46	81.06 10	0.60 14	0.0010
ce-L- 54	93.96 6	3.0 7	0.0059
ce-L- 53	93.96 6	0.60 24	0.0012
ce-M- 46	95.48 10	0.16 4	0.0003
ce-K- 76	101.08 10	0.26 8	0.0006
ce-M- 53	108.38 6	0.16 7	0.0004
ce-K- 86	130.98 20	0.53 13	0.0015
ce-K- 87	132.08 8	0.55 12	0.0016
ce-K- 92	146.48 20	0.13 4	0.0004
ce-K- 93	148.63 15	0.11 3	0.0003
ce-K- 95	152.33 5	0.67 15	0.0022
ce-K-105	177.48 7	0.36 8	0.0014
ce-K-108	182.23 6	0.89 19	0.0035
ce-K-112	192.68 10	0.27 6	0.0011
ce-K-115	200.52 13	0.72 17	0.0031
ce-K-118	208.74 13	0.26 6	0.0011
ce-L- 87	216.76 8	0.104 22	0.0005
ce-K-126	230.48 14	0.35 8	0.0017
ce-L- 95	237.01 5	0.77 17	0.0039
ce-M- 95	251.93 5	0.21 5	0.0011
ce-L-108	266.91 6	0.18 4	0.0010
ce-L-115	285.20 13	0.11 3	0.0007

227TH A DECAY (18.718 D 5) I(min)=0.10%

Radiation Type	Energy (keV)	Intensity (%)	Δ(g-rad/ μCi-h)
α 1	5585.9 16	0.176 6	0.0209
α 2	5600.6 18	0.170 17	0.0203
α 3	5613.3 16	0.216 8	0.0258
α 4	5668.0 15	2.06 12	0.249
α 5	5693.0 16	1.50 10	0.182
α 6	5700.8 16	3.63 20	0.441
α 7	5709.0 16	8.2 3	0.997
α 8	5713.2 16	4.89 20	0.595
α 9	5757.06 15	20.3 10	2.49
α 10	5762.3 15	0.228 10	0.0280
α 11	5795.5 15	0.311 5	0.0384
α 12	5807.5 15	1.270 20	0.157
α 13	5866.6 15	2.42 10	0.302
α 14	5909.9 15	0.174 8	0.0219
α 15	5916.0 15	0.78 3	0.0983
α 16	5959.7 15	3.00 15	0.381
α 17	5977.92 10	23.4 10	2.98
α 18	6008.81 15	2.90 15	0.371
α 19	6038.21 15	24.5 10	3.15
22 weak α's omitted (ΣIα = 0.19%)			
γ 2	8.00 20	0.14 3	≈0
γ 3	8	0.14 3	≈0
X-ray L	12.3	43 9	0.0113
γ 5	20.27 10	0.20 8	≈0
γ 9	29.91 3	0.10 4	≈0
γ 15	43.80 10	0.23 6	0.0002
γ 14	43.8 5	0.23 6	0.0002
γ 21	49.88 10	0.20 16	0.0002
γ 22	50.20 10	8.5 18	0.0091
γ 30	62.50 20	0.24 6	0.0003
γ 39	79.77 6	2.1 5	0.0036
X-ray $K\alpha_2$	85.430 10	1.78 12	0.0032
X-ray $K\alpha_1$	88.470 10	2.93 19	0.0055
γ 41	94.00 6	1.4 3	0.0028
X-ray Kβ	100	1.34 9	0.0029
γ 53	113.20 6	0.15 6	0.0004
γ 54	113.20 6	0.74 16	0.0018
γ 52	113.2	0.56 12	0.0014
γ 55	117.20 7	0.17 4	0.0004
γ 62	141.30 7	0.13 3	0.0004
γ 75	204.30 10	0.23 6	0.0010
γ 76	205.00 10	0.15 5	0.0007
γ 77	206.10 10	0.23 5	0.0010
γ 79	210.65 10	1.13 24	0.0051
γ 86	234.90 20	0.45 11	0.0023
γ 87	236.00 8	11.2 24	0.0563
γ 91	250.20 10	0.37 11	0.0020
γ 92	250.40 20	0.13 4	0.0007
γ 93	252.55 15	0.11 3	0.0006
γ 94	254.70 20	0.80 19	0.0043
γ 95	256.25 5	6.8 15	0.0371
γ 96	262.80 10	0.100 23	0.0006
γ101	273.00 10	0.49 11	0.0028
γ105	281.40 7	0.61 13	0.0037
γ108	286.15 6	1.6 4	0.0096
γ112	296.60 10	0.43 10	0.0027
γ113	300.00 10	0.20 9	0.0013
γ114	300.30 20	0.20 9	0.0013
γ115	304.44 13	1.05 25	0.0068
γ118	312.66 13	0.48 11	0.0032
γ120	314.86 11	0.46 11	0.0031
γ125	329.90 10	2.8 6	0.0193
γ126	334.40 14	1.00 23	0.0071
γ128	342.46 6	0.35 9	0.0026
γ130	350.50 10	0.110 25	0.0008
193 weak γ's omitted (ΣIγ = 1.71%)			

223RA A DECAY (11.434 D 2) I(min)=0.10%

Radiation Type	Energy (keV)	Intensity (%)	Δ(g-rad/ μCi-h)
Auger-L	8.7	26 3	0.0048
ce-K- 11	23.91 7	7.24 18	0.0037
ce-K- 15	45.80 5	12.6 4	0.0123
ce-K- 16	55.79 4	18.2 5	0.0216
Auger-K	62.7	1.4 8	0.0019
ce-L- 11	104.26 8	1.36 4	0.0030
ce-M- 11	117.83 6	0.322 8	0.0008
ce-NOP-11	121.21 6	0.115 3	0.0003
ce-L- 15	126.15 6	2.29 7	0.0062
ce-L- 16	136.14 5	3.29 9	0.0095
ce-M- 15	139.72 4	0.561 16	0.0017
ce-NOP-15	143.10 4	1.90 6	0.0058
ce-M- 16	149.71 3	0.783 20	0.0025
ce-NOP-16	153.09 3	0.272 7	0.0009
ce-K- 34	171.01 4	9.1 3	0.0330
ce-K- 38	225.49 5	1.57 5	0.0075
ce-K- 41	239.92 7	1.01 4	0.0052
ce-L- 34	251.36 5	1.66 5	0.0089
ce-M- 34	264.93 3	0.392 11	0.0022
ce-NOP-34	268.31 3	0.135 4	0.0008
ce-L- 38	305.84 6	0.285 9	0.0019
ce-L- 41	320.27 8	0.182 6	0.0012
ce-K- 57	346.54 6	0.197 10	0.0015
α 1		0.25	≈0
α 2	5287.3 10	0.15	0.0169
α 3	5338.7 10	≈0.13	≈0.0148
α 4	5365.6 10	≈0.13	≈0.0149
α 5	5433.6 5	2.27 20	0.263
α 6	5501.6 10	1.00 15	0.117
α 7	5540.0 10	9.2 3	1.08
α 8	5606.9 3	24.2 4	2.89
α 9	5716.4 3	52.5 8	6.39
α 10	5747.2 4	9.5 6	1.16
α 11	5857.5 10	0.32 4	0.0399
α 12	5871.6 10	0.85 4	0.106
16 weak α's omitted (ΣIα = 0.31%)			
X-ray L	11.7	23 3	0.0057
γ 5	33.6 5	0.10 3	≈0
X-ray $K\alpha_2$	81.070 20	14.2 4	0.0245
X-ray $K\alpha_1$	83.780 20	23.6 6	0.0421
X-ray Kβ	94.9	10.7 3	0.0216
γ 11	122.31 6	1.190 20	0.0031
γ 15	144.20 4	3.26 7	0.0100
γ 16	154.19 3	5.59 10	0.0184
γ 17	158.62 4	0.688 13	0.0023
γ 22	179.67 12	0.153 12	0.0006
γ 34	269.41 3	13.6 3	0.0780
γ 38	323.89 4	3.90 9	0.0269
γ 39	328.50 6	0.198 10	0.0014
γ 41	338.32 6	2.78 7	0.0200
γ 42	342.90 7	0.20 3	0.0015
γ 43	346.8 3	0.17	0.0013
γ 47	371.84 8	0.490 12	0.0039
γ 57	444.94 5	1.27 6	0.0120
53 weak γ's omitted (ΣIγ = 1.06%)			

219RN A DECAY (3.96 S 1) I(min)=0.10%

Radiation Type	Energy (keV)	Intensity (%)	Δ(g-rad/ μCi-h)
Auger-L	8.33	1.18 11	0.0002
ce-K- 1	37.59 10	0.36 3	0.0003
ce-K- 2	178.09 5	1.16 10	0.0044
ce-L- 2	254.26 5	0.68 4	0.0037
ce-K- 3	308.69 20	0.228 12	0.0015
α 1	6424.9 10	7.5 5	1.03
α 2	6553.2 10	11.5 5	1.61
α 3	6819.3 3	81.0 10	11.77
X-ray L	11	0.89 10	0.0002
X-ray $K\alpha_2$	76.862 5	0.49 4	0.0008
X-ray $K\alpha_1$	79.290 5	0.83 6	0.0014
X-ray Kβ	89.8	0.371 24	0.0007
γ 1	130.70 10	0.105 6	0.0003
γ 2	271.20 5	10.0 5	0.0578
γ 3	401.80 20	6.5 4	0.0558

215PO A DECAY (0.001780 S 4) I(min)=0.10%

TWO WEAK ALPHA GROUPS (<0.1%) OMITTED
215PO ALSO DECAYS VIA A 0.00023% B- BRANCH TO 215AT

Radiation Type	Energy (keV)	Intensity (%)	Δ(g-rad/ μCi-h)
α 1	7386.4 8	100	15.73

211PB B- DECAY (36.1 M 2) I(min)=0.10%

Radiation Type	Energy (keV)	Intensity (%)	Δ(g-rad/ μCi-h)
β- 1 max	539 6		
avg	161.0 20	6.10 20	0.0209
β- 2 max	966 6		
avg	315 3	1.80 20	0.0121
β- 3 max	1371 6		
avg	473 3	91.40 20	0.921
total β- avg	451 3	99.3 4	0.954
γ 1	404.84 4	3.68 8	0.0317
γ 2	426.99 4	1.70 6	0.0154
γ 3	831.83 4	3.50 10	0.0620

211BI A DECAY (2.14 M 2) I(min)=0.10%

Radiation Type	Energy (keV)	Intensity (%)	Δ (g-rad/ μCi-h)
Auger-L	7.78	1.28 10	0.0002
ce-K- 1	265.47 10	2.63 5	0.0149
α 1	6278.8 6	16.CO 10	2.14
α 2	6623.1 6	84.CC 10	11.85
X-ray L	10.3	0.85 10	0.0002
X-ray $K\alpha_2$	70.8319 9	0.739 21	0.0011
X-ray $K\alpha_1$	72.8715 9	1.25 4	0.0019
X-ray Kβ	82.6	0.553 17	0.0010
γ 1	351.00 10	12.77 8	0.0955

207TL B- DECAY (4.77 M 2) I(min)=0.10%

Radiation Type	Energy (keV)	Intensity (%)	Δ (g-rad/ μCi-h)
β- 1 max	1421 6		
avg	494 3	99.76 4	1.05

Selected Index of Decay Data Sets (ENSDF)

September 1975

^{3}H β- decay
^{11}C β+ decay
^{13}N β+ decay
^{14}C β- decay
^{15}O β+ decay
^{16}N β- decay
^{18}F β+ decay
^{22}Na β+ decay
^{24}Na β- decay
^{26}Al β+ decay (7.16x10^{5} y)
^{27}Mg β- decay
^{28}Mg β- decay
^{28}Al β- decay
^{32}Si β- decay
^{32}P β- decay
^{33}P β- decay
^{35}S β- decay
^{38}Cl β- decay
^{41}Ar β- decay
^{42}K β- decay
^{43}K β- decay
^{44}Ti ε decay
^{44}Sc β+ decay
^{45}Ca β- decay
^{46}Sc IT decay (18.67 s)
^{46}Sc β- decay
^{47}Ca β- decay
^{47}Sc β- decay
^{48}V ε decay
^{51}Cr ε decay
^{52}Mn ε decay (5.67 d)
^{52}Fe β+ decay
^{52}Mn β+ decay (21.1 m)
^{52}Mn IT decay (21.1 m)
^{54}Mn ε decay
^{55}Fe ε decay
^{56}Mn β- decay
^{57}Co ε decay
^{58}Co ε decay
^{59}Fe β- decay
^{60}Co β- decay
^{63}Ni β- decay
^{64}Cu β+ decay
^{64}Cu β- decay
^{65}Zn β+ decay
^{67}Cu β- decay
^{67}Ga ε decay
^{68}Ga ε decay
^{68}Ge β+ decay
^{69}Zn IT decay (13.76 h)
^{69}Zn β- decay (13.76 h)
^{69}Zn β- decay (57 m)
^{72}Ga β- decay
^{72}As ε decay
^{73}As ε decay
^{73}Se ε decay (7.1 h)
^{74}As β- decay
^{74}As ε decay
^{75}Se ε-decay
^{76}As β- decay
^{77}As β- decay
^{77}Br β+ decay
^{79}Kr β+ decay
^{81}Kr ε decay
^{82}Br β- decay (35.30 h)
^{84}Rb β- decay
^{84}Rb β+ decay
^{85}Kr β- decay (4.48 h)
^{85}Kr IT decay (4.48 h)
^{85}Kr β- decay (10.72 y)
^{85}Sr ε decay
^{86}Rb β- decay
^{86}Y β+ decay
^{86}Zr ε decay
^{87}Kr β- decay
^{87}Y ε decay
^{87}Sr ε decay (2.805 h)
^{87}Sr IT decay (2.805 h)
^{88}Kr β- decay
^{88}Rb β- decay
^{88}Y ε decay
^{89}Kr β-decay
^{89}Rb β-decay
^{89}Sr β-decay
^{90}Sr β- decay
^{90}Y β- decay
^{94}Nb IT decay (6.26 m)
^{94}Nb β- decay (6.26 m)
^{94}Nb β- decay (2.03x10^{4} y)
^{97}Nb IT decay
^{97}Zr β- decay
^{97}Nb β- decay
^{97}Ru ε decay
^{99}Mo β-decay
^{99}Tc IT decay (6.02 h)
^{103}Ru β- decay
^{103}Rh IT decay (56.12 m)
^{103}Pd ε decay
^{109}Pd β- decay (13.46 h)
^{109}Cd ε decay
^{110}Ag β- decay (24.6 s)
^{110}Ag β- decay (252 d)
^{110}Ag IT decay (252 d)
^{110}Ag ε decay (24.6 s)
^{111}Ag β- decay
^{111}In ε decay
^{113}Sn ε decay
^{113}In IT decay (1.658 h)
^{115}In IT decay (4.3 h)
^{115}In β- decay (4.3 h)
^{116}In β- decay (54.15 m)
^{117}Sb ε decay
^{117}Sn IT decay (14.0 d)
^{119}Sn IT decay (245 d)
^{121}Te ε decay (154 d)
^{121}Te ε decay (17 d)
^{121}Te IT decay (154 d)
^{122}Sb ε decay
^{122}Sb β- decay
^{123}I ε decay
^{124}Sb β- decay (60.20 d)
^{124}I β+ decay
^{125}I ε decay
^{126}I β- decay
^{126}I ε decay
^{127}Xe ε decay
^{129}Te IT decay (33.6 d)
^{129}Te β- decay (33.6 d)
^{129}Te β- decay (69.6 m)
^{129}Xe IT decay (8.0 d)
^{129}I β- decay
^{130}I β- decay (12.36 h)
^{131}I β- decay
^{131m}Xe IT decay
^{133}I β- decay
^{133}Xe IT decay (2.19 d)
^{133}Ba ε decay (10.5 y)
^{133}Xe β- decay (5.245 d)
^{133}Ba IT decay (38.9 h)
^{134}Te β- decay
^{134}I β- decay
^{134}Cs β- decay
^{135}I β- decay
^{135}Xe IT decay (15.29 m)
^{135}Xe β- decay (9.172 h)
^{136}Cs β- decay
^{138}Xe β- decay
^{138}Cs β- decay (32.2 m)
^{141}Ce β- decay
^{145}Pm ε decay
^{175}Yb β- decay
^{177}Lu β- decay
^{181}Hf β- decay
^{181}W ε decay
^{185}W β-decay
^{187}W β- decay
^{191}Os β- decay
^{194}Ir β- decay
^{197}Pt β- decay (20.0 h)
^{198}Au β- decay
^{199}Au β- decay
^{201}Tl ε decay
^{203}Hg β- decay
^{203}Pb ε decay
^{207}Bi ε decay
^{239}Np β- decay

75-9905
Translated from French

Review Paper No. A2

Importance of Transactinide Nuclear Data for the Physics of Fast and Thermal Reactor Cores

J.Y. Barre and J. Bouchard
Commissariat à l'énergie atomique, France

Abstract

The importance of transactinium isotope nuclear data in the physics of fast and thermal reactors relating directly to core parameters and to fuel composition calculations is reviewed. The influence of transactinides on reactivity loss and internal breeding gain and power in fast reactors is alanysed. The accuracies required in transactinide nuclear data for core parameter calculations, and the sensitivity of the isotopic compositions of fast reactor fuel due to variations in transactinide cross sections are evaluated. Specific nuclear data accuracies are recommended.

Introduction

The problems relating to the knowledge of transactinide nuclear data of relevance to the physics of fast and thermal reactor cores fall into two main categories:

1. Problems relating directly to the parameters of the core itself (change in isotopic composition, reactivity, power);

2. Problems relating to the fuel composition calculations necessary for other parts of the fuel cycle (fuel handling, fuel transport, fuel reprocessing, waste management, fuel fabrication using recycled fuel - subjects considered in greater detail in other review papers presented at this meeting [7]).

The case of fast and thermal reactors is considered separately for each category of problem, especially the first one.

In the case of fast reactors, we consider only sodium-cooled plants using PuO_2-UO_2 as fuel. In the case of thermal reactors, the emphasis is on light-water reactors with or without plutonium recycling, but HTRs are also considered.

A clear distinction is made between the higher plutonium isotopes (^{240}Pu, ^{241}Pu, ^{242}Pu) and ^{236}U, in respect of which systematic study programmes are already under way for the different basic reactor types, and the other transactinium isotopes (the other plutonium isotopes and the isotopes of americium and curium), which have been studied far less.

N.B. In this paper, all uncertainties correspond to the equivalent of a standard deviation (67 % change of occurring). An accuracy of ± 50 % corresponds to an uncertainty factor of 2.

1. PROBLEMS RELATING DIRECTLY TO THE CORE PARAMETERS

1.1. General remarks

Until a few years ago, most reactor calculations covered only the principal isotopes of thorium, uranium and plutonium. It is only recently that - with the development of nuclear power plants, high burn-up rates and the study of more complex fuel cycles - it has become necessary to take into account a larger number of isotopes, extending from ^{232}U to ^{245}Cm.

The importance of these transactinides varies greatly, depending on what one is concerned with: reactivity, radiation protection, etc.

It is important to have an idea of the long-term trend as regards the production of transactinides so that one can determine whether the nuclear data relating to them will need to be increasingly accurate and whether they may become a major problem in some cases.

The relative concentrations of these isotopes, which generally increase rapidly at the start of the fuel cycle, reach fairly low saturation levels, and the importance of the isotopes remains less than that of the principal fissile or fertile isotopes - at least in the case of the fuel cycles which are being studied at present.

The saturation process is well known in the case of ^{240}Pu. Another example is ^{238}Pu - an interesting isotope owing to its alpha activity and to the fact that it cannot be eliminated in the course of reprocessing.

The formation of ^{238}Pu is important during the first uranium fuel cycle of a light-water reactor. For a burn-up of 35 GWd/ton the concentration of ^{238}Pu in the total plutonium is close to 1.5%. Three situations are then possible:

(a) No plutonium recycling - the concentration of 1.5% remains a maximum;

(b) Plutonium recycling in a light-water reactor - the concentration remains between 1% and 1.5%, this being attributable to the reduced formation of ^{238}Pu from ^{236}U, which largely compensates for the increase due to the (n,2n) reactions associated with ^{239}Pu and to the alpha decay of ^{242}Cm;

(c) Use of the plutonium in fast reactors - the ^{238}Pu concentration in the total plutonium decreases rapidly, stabilizing around 0.5%; this is due to a production/disappearance ratio for this isotope which is less favourable under fast-spectrum conditions than under thermal-spectrum conditions and to the fact that the plutonium in the blankets contains virtually no ^{238}Pu.

The example of ^{238}Pu is no exception. Nevertheless, while it is important to know that the relative concentrations of these transactinium isotopes are usually fairly low, one should bear in mind that the total quantities produced are increasing.

The principal growth and decay sequence employed in core calculations is shown in Fig. 1.

The nuclear data necessary for reactor calculations, for determining the core parameters and for predicting fuel compositions are:

- The neutron cross-sections ($\sigma(n,\gamma)$, $\sigma(n,f)$, $\sigma(n,2n)$);
- The mean numbers of neutrons per fission ($\bar{\nu}$);
- Certain radioactive half-lives (^{239}Np, ^{241}Pu, ^{242}Cm, ...).

1.2. Neutronics

1.2.1. Thermal reactors

We consider mainly the case of light-water reactors without or with plutonium recycling. Comparisons are made with HTR systems, and the conclusions are not appreciably different. Transactinium isotopes are less important in reactors which use natural or very slightly enriched uranium, and we do not give any results for cores of this type.

Table I gives, by way of example, the isotopic concentrations of the transactinides in irradiated PWR fuel for a burn-up of 30 GWd/ton.

These concentrations are very dependent on several parameters unrelated to nuclear data - for example,

- Initial composition of the fuel,
- Core characteristics,
- Refuelling mode,
- Power history.

The values in Table I should therefore be regarded only as orders of magnitude.

Table I

Isotopic concentrations of transactinides in a PWR fuel for a mean burn-up of 30 000 MWd/ton

(isotopic masses in g/ton of oxide fuel)

Isotope	Uranium fuel	Pu recycling
^{232}U	10^{-4}	8×10^{-5}
^{234}U	126	90
^{236}U	4240	2985
^{237}Np	398	316
^{239}Np	6	6
^{236}Pu	7×10^{-4}	6×10^{-4}
^{238}Pu	107	172
^{240}Pu	1695	3220
^{241}Pu	1047	2030
^{242}Pu	252	572
^{241}Am	30	84
^{242m}Am	1.3	4
^{243}Am	48	186
^{242}Cm	9.4	26
^{243}Cm	0.05	0.2
^{244}Cm	9,3	85
^{245}Cm	0.02	0.3

If one excludes ^{236}U and the higher plutonium isotopes, the highest concentrations are of ^{237}Np, ^{234}U, ^{238}Pu, ^{241}Am and ^{243}Am. In the case of a uranium fuel, we can neglect all the other isotopes, even if the burn-up is higher.

In a situation with plutonium recycling, the uranium and neptunium isotopes are less important, while the principal curium isotopes (^{242}Cm and ^{244}Cm) and ^{242m}Am can no longer be neglected.

The reactivity consequences are analysed in section 1.3.1.

1.2.2. Fast reactors

For the purposes of this paper, we have chosen as example of a fast reactor a sodium-cooled, conventional 1200 MW(e) power plant using PuO_2-UO_2. The fuel for this plant is assumed to be from a PWR without plutonium recycling. Given the time between reprocessing and fuelling, the following isotopic compositions at the time of start-up are assumed (Table II):

^{238}Pu : 1%
^{239}Pu : 58%
^{240}Pu : 24%
^{241}Pu : 12%
^{242}Pu : 5%
^{241}Am : 2% of the total plutonium

The total plutonium enrichment is of the order of 15% and the flux $\simeq 6 \times 10^{15}$ n.cm^{-2}s^{-1}.

The parameters of the isotopic composition changes in the core which were studied relate to the loss of reactivity per cycle and the internal breeding gain.

The cycle duration is taken to be one year (load factor $\simeq 0.8$), corresponding to a burn-up of about 30 000 MWd/ton. The variations in atomic composition (atoms/cm^3) of the principal transactinides are given in Table II for two burn-ups: 30 000 MWd/ton and 120 000 MWd/ton. There is a general accumulation of ^{237}Np, ^{242m}Am, ^{243}Am and ^{244}Cm and a saturation or reduction of ^{238}Pu, ^{241}Am and ^{242}Cm. The concentration of the various transactinides always remains lower - by a factor of at least 5 - than that of ^{241}Pu or ^{242}Pu. These atomic compositions should be regarded as orders of magnitude - functions of the assumptions made.

Table II

Order of magnitude of the atomic compositions of the principal transactinides as a function of burn-up; 1200 MW(e) fast reactor

(N atoms/cm^3 x 10^{24})

Higher Pu isotopes	Initial state	30 000 MWd/ton	120 000 MWd/ton
^{240}Pu	2.9 -04	3.1 -04	3.6 -04
^{241}Pu	1.5 -04	1.1 -04	6.3 -05
^{242}Pu	6.2 -05	6.5 -05	6.1 -05

Transactinides	Initial state	30 000 MWd/ton	120 000 MWd/ton
^{232}U	0.	9. -13	3. -11
^{237}U	0.	4. -08	4. -08
^{237}Np	5. -08	1.1 -06	3.7 -06
^{239}Np*	0.	3. -06	3. -06
^{236}Pu	0.	1.4 -11	1.1 -10
^{237}Pu	0.	2.1 -10	1.6 -10
^{238}Pu	1.2 -05	1.0 -05	0.8 -05
^{241}Am	2.1 -05	1.9 -05	1.3 -05
^{242m}Am	0.	8. -07	1.3 -06
^{243}Am	0.	5. -06	1.3 -05
^{242}Cm	0.	9. -07	8. -07
^{243}Cm	0.	4. -08	1.7 -07
^{244}Cm	0.	5. -07	5.4 -06

* Isotope with a short half-life which has an effect primarily on the reactivity variation when the plant is started up again.

The detailed contribution of the principal transactinides other than the higher plutonium isotopes to the reactivity loss per cycle ($\simeq -2\%\ \Delta K/K$) is shown in Table III for the first cycle and for an equilibrium situation (around 120 000 MWd/ton). While the overall contribution is zero, because of mutual cancelling, the individual effects remain non-negligible, particularly in the case of ^{242m}Am, ^{243}Am, ^{242}Cm, ^{238}Pu and ^{241}Am. The required accuracy in the quantity $(\nu\sigma_F - \sigma_a)$, which has a bearing on reactivity, varies between $\pm$ 30% and $\pm$ 50% for these isotopes.

The large variations - depending on the cycle considered - in the contributions of each isotope to this parameter should be noted.

Certain isotopes with an intermediate half-life (a few days) can modify the reactivity loss after re-startup before reaching an equilibrium concentration: that is the case with ^{239}Np, which contributes $\simeq +0.12\ \frac{\Delta K}{K}$ after a shutdown. An accuracy of the order of $\pm$ 20% in $(\nu\sigma_F - \sigma_a)$ is also desirable in the case of this isotope for purposes of power plant control.

By comparison, for the higher plutonium isotopes the desirable accuracies in $(\nu\sigma_F - \sigma_a)$ for the reactivity loss per cycle are: $\pm$ 30% for ^{240}Pu; $\pm$ 3% for ^{241}Pu; $\pm$ 100% for ^{242}Pu.

The influence of the transactinides on the internal breeding gain (IBG) is also slight, except in the case of ^{241}Am (see Table IV), for which the capture cross-section must be known to within $\pm$ 20%. For ^{242m}Am, it is therefore necessary to know $(\nu\sigma_F - \sigma_a)$, and especially the fission cross-section, to within $\pm$ 20%. In the case of the other isotopes, an order of magnitude ($\pm$ 50%) is sufficient. In the case of the higher plutonium isotopes, the necessary accuracies for the IBG are: $\pm$ 5% for the capture of ^{240}Pu and $\pm$ 3% for the fission of ^{241}Pu.

1.3. Reactivity

1.3.1. Thermal reactors

In Table V we classify the transactinides according to their reactivity effect in a light-water reactor.

It will be seen that, from this point of view, the curium isotopes are completely negligible in the case of uranium fuel and that their effect remains so slight in the case of plutonium recycling that there is no particular demand for data on these isotopes.

As regards the other elements, the accuracy requirements for reactivity calculations are expressed in the second part of Table V. So as to cover - given these requirements - all the situations which can reasonably

Table III

Influence of transactinides on the reactivity loss per cycle or on the reactivity - fast reactor

$\Delta K/K$, in milliniles (10^{-5} $\Delta K/K$)

	Reactivity loss per cycle (1 year ≃ 30 000 MWd/ton)		Reactivity	
	First cycle	Fourth cycle	Initial state	120 000 MWd/ton
^{232}U	0.	0.	0.	0.
^{237}U	0.	0.	0.	0.
^{237}Np	-16.	-10.	-1.	-51.
^{239}Np	-67.	+ 2.	0.	-59.
^{236}Pu	0.	0	0.	0.
^{237}Pu	0.	0	0.	0.
^{238}Pu	-54.	-11.	308.	207.
^{241}Am	49	+44	-473	-292.
^{242m}Am	107	+ 1	0.	174
^{243}Am	-88	-42	0.	-256
^{242}Cm	62	- 7	0.	53
^{243}Cm	1.	1	0.	5.
^{244}Cm	2.	7.	0	20.
TOTAL	-4	-15	-166	-200
^{241}Pu*	-2700	-520	10600	4603

* Given for purposes of comparison.

N.B. Total reactivity loss per cycle ≃ - 2000 millinile - all effects less than 1 millinile are taken to be zero.

Table IV

Influence of transactinides on internal breeding gain, power and total absorption in a fast reactor

	Effect of IBG (absolute)	Percentage of the total power	Percentage of the total absorption
^{232}U	0.	0	0
^{237}Np	0.005	0	0.1
^{239}Np	0.	0	0.1
^{236}Pu	0.	0	0
^{238}Pu	- 0.003	0.5	0.3
^{241}Am	0.023	0.2	0.6
^{242m}Am	- 0.007	0.3	0.1
^{243}Am	0.004	0.1	0.5
^{242}Cm	- 0.001	0.1	0
^{243}Cm	0.	0	0
^{244}Cm	0.	0.1	0
TOTAL	+ 0.021	1.3	1.7
Higher Pu Isotopes			
^{240}Pu	.112	6.3	6.4
^{241}Pu	- .143	7.8	4.0
^{242}Pu	- .001	0.7	1.1

Burn-up $\simeq$ 120 000 MWd/ton

be envisaged at the present time, we have taken into account the possibility of successive recyclings of the plutonium in light-water reactors, which leads to a greater demand for ^{242}Pu, and a thorium cycle in high-temperature reactors. The latter is also the reason for the requirements stated for ^{231}Pa and ^{233}Pa and for the necessary accuracies in the case of ^{232}U and ^{234}U. Lastly, plutonium recycling in an HTR does not lead to different conclusions[2].

1.3.2. Fast reactors

The influence of transactinides on the reactivity balance, which one needs to know in order to predict the critical enrichments either at start-up or at a burn-up of 120 000 MWd/ton is shown in Table III. The percentages of the total absorption for each isotope are given in Table IV for 120 000 MWd/ton. In both cases the effect is slight, owing to mutual cancelling; the individual effects remain non-negligible but of an order of magnitude different from that in the case of the higher plutonium isotopes (e.g. ^{241}Pu).

The resulting accuracy requirements are stated in Table VI.

1.4. Other parameters

1.4.1. Thermal reactors

In a thermal reactor, ^{241}Pu is the only transactinide considered here which makes a significant contribution to the power output: about 2-3% of the total for a reactor using 5-10% enriched uranium in the case of plutonium recycling. In order to calculate this contribution, however, it is not necessary to have data more accurate than for reactivity effect calculations. The contributions to power output of ^{240}Pu and ^{242m}Am fission are always well below 1%, even in the most extreme cases. There is accordingly no particular demand.

In connection with these fission rates, the effective fractions of delayed neutrons corresponding to these isotopes are also secondary and no special requirement for kinetic calculations is stated.

Capture in ^{240}Pu is one of the important reactions for spectrum calculations, above all because of the 1 eV resonance. However, so far there appears to be no particular lack of data here.

1.4.2. Fast reactors

The contribution of transactinides to the total power due to fission is shown in Table IV; it is very low. There are accordingly no particular requirements in the case of transactinides as regards either the energy released through fission or the delayed neutron parameters.

Table V

A. Reactivity effects of transactinides in a light-water reactor

Effect on δ K/K	Uranium fuel	Plutonium recycling
$> 10^{-2}$	^{236}U, ^{240}Pu, ^{241}Pu	^{236}U, ^{240}Pu, ^{241}Pu
10^{-3} à 10^{-2}	^{234}U, ^{237}Np, ^{242}Pu	^{234}U, ^{237}Np, ^{242}Pu, ^{241}Am, ^{243}Am
10^{-4} à 10^{-3}	^{238}Pu, ^{241}Am, ^{243}Am, ^{242m}Am	^{238}Pu, ^{242m}Am
10^{-5} à 10^{-4}	/	^{242}Cm, ^{243}Cm, ^{244}Cm

B. Accuracies necessary in the case of thermal reactor cross-sections related to the reactivity effects

Precision	Isotopes	Cross-sections
< 5 %	^{234}U, ^{236}U, ^{240}Pu, ^{241}Pu, ^{242}Pu ^{241}Pu	$\sigma(n, \gamma)$ $\sigma(n, f)$, $\bar{\nu}$
10 %	^{231}Pa, ^{233}Pa, ^{237}Np, ^{241}Am, ^{243}Am	$\sigma(n, \gamma)$
30 %	^{232}U, ^{238}Pu ^{242m}Am	$\sigma(n, \gamma)$ $\sigma(n,f)$, $\bar{\nu}$

Table VI

Desirable precisions in the case of fast reactor cross-sections for reactivity calculation problems

($\pm$ in %)

Higher Pu isotopes	$\nu\sigma_F - \sigma_a$	σ_c	σ_F	ν
240 Pu	8	5	2	1
241 Pu	2	8	1.5	0.5
242 Pu	50	8	4	4
Other transactinides				
237 Np	50	30	50	50
239 Np	50	50	-	-
238 Pu	15	20	7	4
241 Am	10	5	15	10
242 m Am	30	100	15	10
243 Am	20	10	30	25
242 Cm	50	50	50	30

1.5. Conclusions

The accuracies required in transactinide nuclear data for determining core parameters are summarized in Table VII for thermal and fast reactors.

It should be emphasized that these accuracies relate to cross-sections calculated on the basis of typical power reactor cross-sections.

As regards the isotopes belonging to the first category, the required accuracies have already been or soon will be achieved, thanks in particular to the integral measurement programmes which have been completed or are still under way. This is also true for some isotopes in the second part of Table VII.

Several examples can be cited:

- Analyses of irradiated fuels from light-water reactors (several countries),
- Programmes relating to higher plutonium isotopes in fast reactors (CEA),
- ^{241}Am capture and the fission of several transactinides measured in ZEBRA (UK),
- Programme relating to transactinides for plutonium recycling in light-water reactors (EURATOM).

A brief description of the programme on higher plutonium isotopes in fast reactors is given in Annex I, where an example is given of what is expected in such programmes.

2. REQUIREMENTS FOR PREDICTING FUEL COMPOSITIONS

The reactor physics calculations necessary for determining core parameters also enable us to predict the composition of irradiated fuels and are therefore important for the correct assessment of problems of other parts of the fuel cycle, radiation protection, transportation, reprocessing, etc.

In this part of our paper we first give the results of two sensitivity calculations which should enable one to link requirements as regards a knowledge of isotopic compositions with demands in the field of neutron cross-sections. We then put forward some conclusions with a view to finding whether the most precise cross-section requirements, and hence the effort we must put into the corresponding neutron calculations, relate to the core parameters or the composition of the unloaded fuels.

Table VII

Accuracies required in transactinide nuclear data for core parameter calculations ($\pm$ in %)

Isotopes	$\sigma(n,\gamma)$		$\sigma(n,f)$		$\bar{\nu}$
	Thermal	Fast	Thermal	Fast	
234 U	5	-	-	-	-
236 U	4	-	-	-	-
240 Pu	1	5	-	2	1
241 Pu	3	8	1	1.5	0.5
242 Pu	5	8	-	4	4
231 Pa	10	-	-	-	-
233 Pa	10	-	-	-	-
232 U	30	-	-	-	-
237 Np	10	30	-	50	50
239 Np	-	20	-	50	50
238 Pu	30	20	-	7	4
241 Am	10	5	-	15	10
242m Am	50	50	30	15	10
243 Am	10	10	-	30	25
242 Cm	50	50	-	25	15
244 Cm	50	-	-	-	-

N.B. Thermal = Spectra of thermal reactors (LWRs or HTRs)
Fast = Spectra of fast reactors.

2.1. Sensitivity studies

The results of two studies are reported here: the first relates to a PWR with enriched uranium fuel (Table VIII) and the second to a fast reactor with the same characteristics as the one studied in the first part of the paper (Table IX).

2.2. Requirements for predicting isotopic compositions

These requirements relate to problems of the fuel cycle outside the reactor core; they are analysed in greater detail in another paper[7]. Our aim is merely to give some orders of magnitude so as to see whether the corresponding demands for accuracy in the cross-sections are more or less important than those which derive from study of the core parameters.

2.2.1. After-power

Most of the after-power derives from the fission products, the contribution of the heavy nuclei being limited to a maximum of about 20% in the case of a fast reactor fuel and relatively long cooling time. This contribution derives essentially from the alpha particles of the curium isotopes and requires a knowledge of the ^{242}Cm and ^{244}Cm concentrations to within about 50%.

2.2.2. Neutron emission

The neutron emission of transactinides is important in connection with radiation protection, especially for the calculation of lead castles for transportation. Here also the main contribution derives from ^{242}Cm and ^{244}Cm. In Table X, some values are given for light-water and fast reactor fuels.

For radiation protection calculations, an accuracy of 20% for the neutron source is a good approximation. This presupposes a knowledge of the curium concentrations with a maximum uncertainty of $\pm$ 15% if we take into account the other sources of error - i.e. spontaneous fission half-lives and (α,n) reactions.

2.2.3. Problems connected with uranium and plutonium recycling

For plutonium recycling one requires information on:

- Concentrations of the principal isotopes; they are generally well known from core calculations and can be checked after reprocessing;

Modification of the cross-sections			Effect on the isotopic compositions (in %)											
Isotope	Reaction	Variation (%)	^{237}Np	^{236}Pu	^{238}Pu	^{240}Pu	^{241}Pu	^{242}Pu	^{241}Am	^{242m}Am	^{243}Am	^{242}Cm	^{243}Cm	^{244}Cm	^{245}Cm
^{237}Np	C	+ 10	- 3	- 2	+ 7										
^{238}Pu	C	+ 10			- 2										
^{240}Pu	C	+ 10				- 6	+ 4	+ 5	+ 5	+ 5	+ 5	+ 5	+ 5	+ 5	+ 5
^{241}Pu	C	+ 10					- 2	+ 9	- 1	- 1	+ 9	- 1	- 1	+ 9	+ 9
^{241}Pu	F	+ 10					- 4	- 4	- 3	- 3	- 3	- 3	- 3	- 3	- 3
^{242}Pu	C	+ 10						- 2			+ 8			+ 8	+ 8
^{241}Am	C	+ 10							- 5	+ 4		+ 5	+ 5		
^{242m}Am	C	+ 20								- 2					
^{242m}Am	F	+ 20								- 10					
^{243}Am	C	+ 20									- 5			+ 18	+ 18
^{242}Cm	C	+ 20											+ 20		
^{244}Cm	C	+ 20													+ 20

C = capture; F = fission

Table VIII Sensitivity of the isotopic compositions of a PWR fuel to variations in the cross-sections of the transactinides

Isotope	Reaction+	$\frac{\Delta\sigma}{\sigma}$ %	^{236}Pu	^{237}Np	^{238}Pu	^{240}Pu	^{241}Pu	^{242}Pu	^{241}Am	^{242m}Am	^{243}Am	^{242}Cm	^{243}Cm	^{244}Cm	^{245}Cm
^{237}Np	C	20	- 3	- 4	+ 5										
	F	10													
^{238}Pu	C	20			- 4										
	F	20			- 10										
^{240}Pu	C	10				- 1	7	3	5	4	1	4	3		
	F	10				- 1	- 1								
^{241}Pu	C	10					- 1	6	- 1	- 1	4	- 1		3	3
	F	10					- 5	- 2	- 3	- 3	- 1	- 3	- 3	- 1	- 1
^{242}Pu	C	20						- 1			+ 8			11	12
	F	10													
^{241}Am	C	20							- 7	+ 17	+ 1	+ 17	+ 17	-	-
	F	20							- 1	- 1		- 1	- 1		
^{242m}Am	C	50								- 9	1			1	1
	F	50								- 13					
^{243}Am	C	30									- 4			14	17
	F	30									- 1			- 1	
^{242}Cm	C	50										- 1	51		
	F	50										- 1	- 1		
^{244}Cm	C	50												- 15	32
	F	50												- 1	- 1
C = capture; F = fission						Effect on the isotopic compositions (in %)									

Table IX Sensitivity of the isotopic compositions of a fast reactor fuel to variations in the cross-sections of the transactinides

- ^{241}Am formation associated with the presence of ^{241}Pu. (The same conclusion as regards the ^{241}Pu concentration, so that there remains only the half-life. From this point of view, knowledge to within $\pm$ 5% of the americium concentration is quite sufficient.)
- The ^{238}Pu concentration; prediction of this is important because ^{238}Pu gives rise to most of the neutron emission of the plutonium oxide. An accuracy of 20% sufficient for predicting the handling conditions;
- The ^{236}Pu concentration; when it is not completely negligible, the ^{236}Pu concentration is important because of the various photons emitted in the ^{228}Th decay chain. Knowledge to within a factor of 2 is sufficient for this type of prediction.

In the case of uranium reutilization the ^{232}U concentration is important if the material is going to pass through an isotopic enrichment plant; this is so for the same reasons as in the case of ^{236}Pu. The same accuracy is desirable.

2.2.4. Problems connected with waste

Besides the after-power and neutron emission problems associated with transactinides, which are important parameters for waste storage, for long-term risk evaluation one needs a knowledge of the concentrations of the long-lived isotopes (above all ^{241}Am, ^{237}Np and the isotopes of curium). At this level, the required accuracy is of the order of a factor of 2.

2.3. Conclusions and expressions of requirements

From the sensitivity studies reported in section 3.1, and taking into account the aims mentioned in section 3.2, we obtain a list of neutron cross-section requirements (see Table XI).

To these requirements one must add (again for the purpose of calculating the final isotopic compositions):

- Knowledge of the branching ratios of ^{241}Am capture; the important parameter is the fraction leading directly to ^{242}Cm (without passing through the metastable state), which must be known to within $\pm$ 10%;

Table X

Distribution of neutron sources in the irradiated fuels of light-water and fast reactors

Isotopes	Thermal reactors (30 GWd/ton)			Fast reactors					
				30 GWd/ton			120 GWd/ton		
	Total	Sp.f.	(α, n)	Total	Sp.f.	(α, n)	Total	Sp.f.	(α, n)
238 Pu	0.8	0.1	0.7	1.4	0	1.4	0.5	0	0.5
240 Pu	0.8	0.7	0.1	1.3	0.9	0.4	0.6	0.4	0.2
241 Am	0.1	0	0.1	0.5	0	0.5	0	0	0
242 Cm	65	53	12	81	66	15	29	24	5
244 Cm	33	32.5	0.5	15	15	0	69	68	1
T O T	100	86	14	100	66	34	100	86	14

Sp.f. = Spontaneous fissions

(α,n) = (α,n) reaction in oxygen

Table XI

Necessary accuracies of transactinide nuclear data for a knowledge of the isotopic composition of fuel

(± %)

Isotopes	$\sigma(n,\gamma)$	$\sigma(n,f)$
^{238}Pu	50	50
^{242}Pu	15	
^{241}Am	20	
^{243}Am	15	
^{242}Cm	50	
^{243}Cm	50	50
^{244}Cm	50	
^{245}Cm	50	50

- Knowledge of the half-life of ^{241}Pu; the present situation should be fairly satisfactory since an accuracy of 5% is sufficient;
- Certain (n,2n) reactions; especially reactions in ^{238}U, ^{237}Np and ^{239}Pu, for which accuracies of $\pm$ 20%, $\pm$ 50% and $\pm$ 50% respectively would be needed.

3. CONCLUSIONS

The transactinide nuclear data of greatest importance for the core physics of thermal and fast reactors are the cross-sections for capture, fission and certain (n,2n) reactions.

The accuracy requirements are related directly to the necessary accuracies in respect of the core parameters (reactivity) and to the necessary accuracies in respect of the isotopic compositions of the fuel when outside the core (during transport, reprocessing, fabrication, etc.). For most of the isotopes, the first criterion is the more limiting one.

The desired accuracies for the higher plutonium isotopes (^{240}Pu, ^{241}Pu, ^{242}Pu) and for ^{234}U and ^{236}U (Tables VII and XI) have already been or soon will be achieved as a result of integral experiment programmes.

As regards the other transactinides, the desired accuracies (Tables VII and XI) are on the whole not very restrictive, and it should be possible to achieve them through more thorough evaluations. In the case of certain data, it is also possible to use the results of integral experiment programmes (e.g. ^{241}Am). Lastly, analysis of the isotopic compositions of irradiated fuels provides an overall check on the calculations.

It should also be noted that, as regards the problems of the isotopic compositions of fuel when outside the reactor, the accuracies so far achieved represent an order of magnitude on which improvements are likely to be made with experience. It is to be hoped that this meeting will produce better definitions of the accuracies necessary for this type of problem.

ACKNOWLEDGEMENTS

The authors thank all those who - through their contributions - made the writing of this review paper possible.

CONTRIBUTIONS

[1] I. OTAKE, Transplutonium elements in fast reactors, F UJI ELECTRIC CO. LTD, Japan (1975).

[2] C.F. GRIGGS, M.J. HALSALL, Transplutonium elements in thermal reactors, Winfrith, UKAEA (1972).

[3] J.L. CRANDALL, Tons of curium and pounds of Californium D.P.MS. 68-100, Savannah River Laboratory. Presented at the ANS international meeting Washington (1968).

[4] E.J. HENNELLY, Nuclear data for actinide recycle, Savannah River Laboratory, Conference on nuclear cross section and technology, Washington (1975).

[5] C.K. PAULSON, E.J. HENNELLY, Cross section measurement of Pu 236 formation in Pu 238 by 237 Np(n,2n) reactions, NSE 55-24-27 (1974).

BIBLIOGRAPHY

[6] TELLIER, H., Multigroup cross-sections for radiative capture and fission of ^{242m}Am (in French), CEA Note N 1821 (September 1975).

[7] Review papers A3, A4, A5, A6, A7 and A8 presented at this meeting.

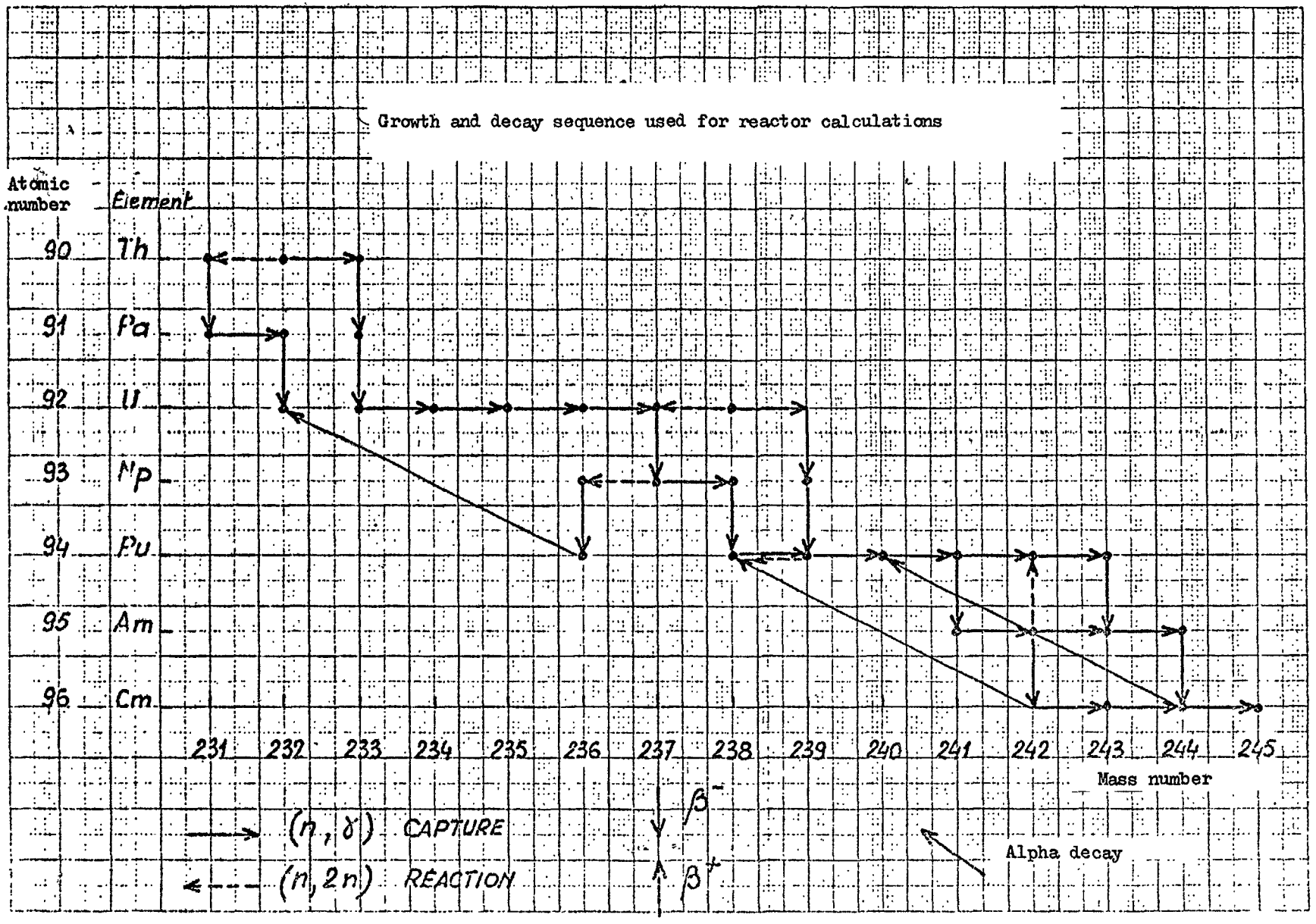

Growth and decay sequence used for reactor calculations
Atomic number
Element
90 Th
91 Pa
92 U
93 Np
94 Pu
95 Am
96 Cm
231 232 233 234 235 236 237 238 239 240 241 242 243 244 245
Mass number
(n, γ) CAPTURE
(n, 2n) REACTION
β⁻
β⁺
Alpha decay

Fig. 1

ANNEX 1

Summary of the programme relating to the higher plutonium isotopes (^{240}Pu, ^{241}Pu, ^{242}Pu) for fast reactors

As regards the higher plutonium isotopes, the desired accuracies will be achieved by the CEA on the basis of the programme of integral experiments which has been completed and the results of which are at present being analysed. Four types of experiment were involved:

- Study of the net reactivity effect of the plutonium fuel as a function of the isotopic compositions of the plutonium in a series of variable spectrum lattices: measurement of $\nu\sigma_F - \sigma_a$;
- Oscillations of plutonium samples with variable isotopic concentrations (for example, between 1% and 50% of ^{240}Pu): measurement of $\nu\sigma_F - \sigma_a$;
- Irradiation of pure samples of the higher plutonium isotopes in Rapsodie and Phénix: measurement of σ_c;
- Study of the fission rate ratios of the higher plutonium isotopes by fission chamber measurements in different lattices in critical experiments: measurement of σ_F.

By way of example, Figs 2-4 show fission rate ratios measured and calculated for ^{240}Pu, ^{241}Pu and ^{242}Pu in different lattices. After adjustment of the evaluated data to the experimental values, the cross-sections will be known to within $\pm$ 2% for the entire fast reactor spectrum range.

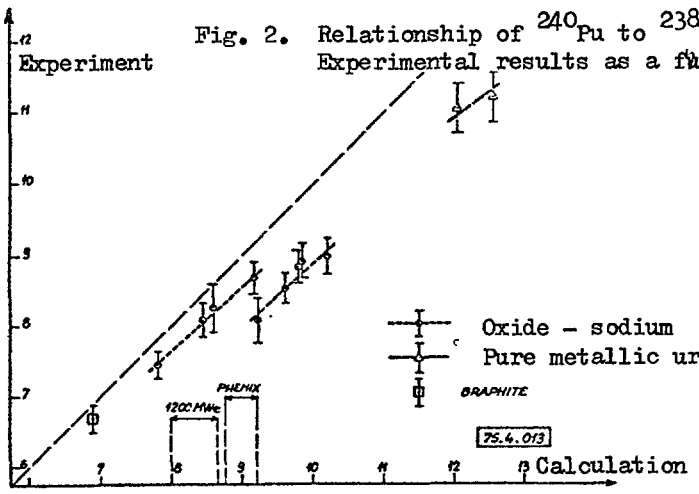


Fig. 2. Relationship of ^{240}Pu to ^{238}U fission rates
Experimental results as a function of calculated results

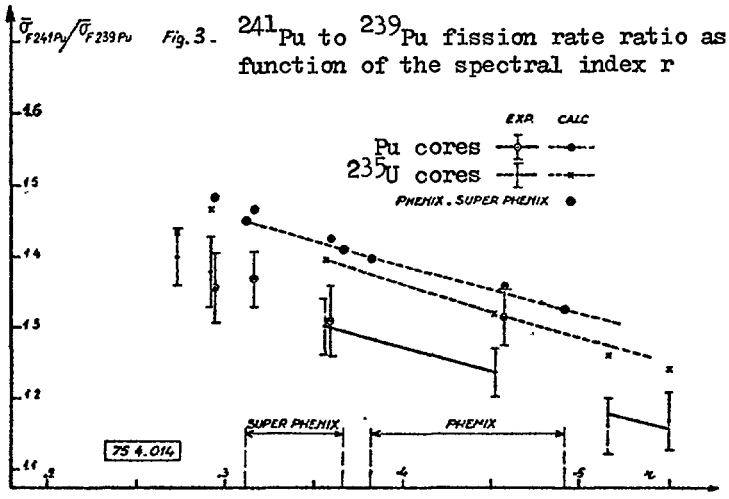


Fig. 3. ^{241}Pu to ^{239}Pu fission rate ratio as a function of the spectral index r

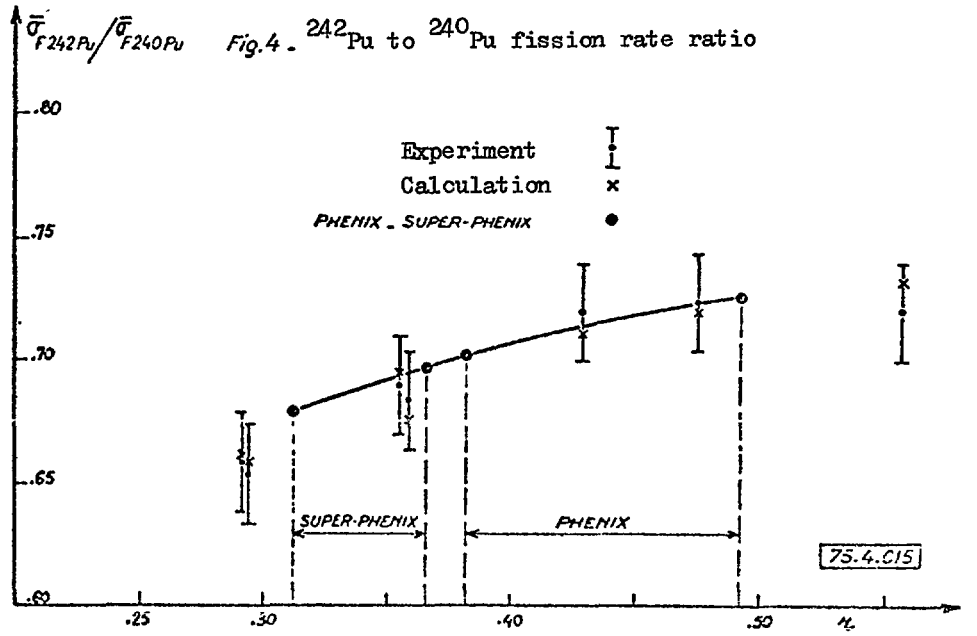


Fig. 4. ^{242}Pu to ^{240}Pu fission rate ratio

Review Paper No. A3

Transactinium Isotope Build-up and Decay in Reactor Fuel and Related Sensitivities to Cross Section Changes

by

H. Kuesters and M. Lalovic

Kernforschungszentrum Karlsruhe
Institut fuer Neutronenphysik und Reaktortechnik

Abstract

The importance of nuclear data for the assessment of transactinium isotope build-up and decay in typical thermal and fast reactors is presented. Consideration is given to associated hazards from radioactive materials during shipping, reprocessing and waste storage. The effect of nuclear data uncertainties on the prediction of transactinium isotope formation and on the level of radioactivity after shutdown is evaluated. Results of transactinium isotope nuclear data sensitivity studies, performed with the ORIGEN code for typical 1000 MWe LWRs, HTGRs and a 2000 MWe LMFBR, are presented.

1. Introduction

The requirements for the accuracy of nuclear data in design applications for thermal and fast reactors are rather well established. Though the requests are not completely met at present, there seems to be no major difficulty in predicting the physics characteristics of fresh or even burnt cores with respect to criticality, power distribution, reactivity and control effects.

These investigations up to now did not deal deeply with the build-up and decay of special transactinium isotopes such as Np, Am and Cm. The reason for this is a rather simple one: it is anticipated that the low concentration of higher actinides does not have a great influence on core performance as far as thermal reactors without Pu-recycling are concerned; even in fast reactors with plutonium fuel the amount of Pu241 and Pu242 is just a few percent of the total plutonium present in a equilibrium cycle with reprocessed fast reactor fuel. In the last years the higher actinides became more important in view of the large amount of radioactive materials to be handled in an increasing

---

Contributors: Dierckx (EUR), Ezure (J), Johansson (S), Lutz (G), Olsson (S), Ottewitte (CH).

reactor population in which use is made of recycled Pu /1/. Mainly the long lived α- emitters are of predominant concern because of the associated radiation hazard. New ideas were born to transmute the α- emitters in reactors or accelerators to less radioactive products. It became obvious that related cross-sections in most cases were not available or were partially missing; generally it was felt that the poor knowledge on the accuracy of the data might lead to major uncertainties in predicting appropriate shielding requirements for shipping spent fuel, in predicting the amount of decay heat after reactor shutdown, and in reliably predicting the associated radiation hazard of stored, radioactive waste. Moreover, the occurrance of spontaneous fission events was thought to lead to uncertainties in the determination of shutdown- and fuel reloading reactivitiy; as far as transmutation of isotopes is considered, the uncertainties of nuclear data even may not allow firm conclusions on the various routes of presently proposed procedures.

This study is aimed to give a first indication on the importance of nuclear data for the transactinium isotopes build-up and decay in reactors and for the associated hazards of the radioactive materials during shipping, reprocessing and waste storage.

The sensitivity of decay heat production to nuclear data uncertainties is also discussed. In addition the dependence of special transactinide build-up on the design properties and the performance of the various reactor types will be given. Uncertainties related to the methods used in describing the fuel cycle are analysed. Results from postirradiation analyses and from integral experiments in fast zero power assemblies are discussed in order to show the present differencies between theoretical predictions and experimental assessment. This investigation is performed for typical reactors of present interest as LWRs, HTGRs, LMFBRs.

2. Nuclear data in use for the description of transactinium isotopes.

Most of the investigations, dealing with longtime aspects of reactor fuel, are based on data which originate from measurements in the early sixties. Only a few special measure-

ments and evaluations have been made /2/, often reference is made to ENDFB/2, the Table of Isotopes /3/ or BNL 325/2. Only some investigations have been performed using modified ENDFB/3 data and BNL 325, 3rd Edition /5,6/. Reactor industry often uses adjusted cross sections or group constants. As a general rule, the data in question are not very reliable and in some cases, as for instance the capture of Am241, they are estimated to be uncertain to about a factor of 2. This situation is easily understood by quoting two reasons:

- not a high priority was given from reactor physics applications for special data measurements.

- measurements on the radioactive materials are extremely difficult.

A general review of the data status is being given in other papers to this meeting /4/ and will not be discussed further in this paper. However, one aspect should be mentioned: for many cases of practical interest data are not at hand and therefore the users are requested to implement rather rough estimates or simplifications for special cross sections in the transactinium area. This situation seems to be the main reason for complaints from users to data producers.

3. Methods used to determine fuel cycle aspects.

In order to be able to deduce requests for nuclear data of special transactinium isotopes, one has to consider the accuracy of theoretical methods, and, if predictions are to be compared with experimental results, the accuracy of measurements also. The latter aspect will be discussed to some extent in chapter 5 of this paper.

3.1 General Remarks

Quite generally, the methods presently available are in fact capable to deal with the object to describing the behaviour of actinides in a reactor system. The sophistication is not always used by the various groups, depending on the objective of the special investigations.

3.2 Thermal Reactors

For thermal reactors the starting point normally is a cell calculation, dealing with many neutron groups in the thermal region and applying for instance THERMOS to determine the space and energy dependent neutron spectra, i.e. using neutron transport theory. For the ephithermal resonance region either effective resonance integrals are used or special spectrum codes deal with resolved and unresolved resonances directly.

The whole core calculations then normally are done in a few group diffusion theory, using the fine group neutron spectra and the flux distribution in space for group collapsing. In dealing with the long term behaviour of reactor fuel, this sophistication is not used for each time step, but rather at some special time intersections, when fuel shuffling, burn up or build up of isotopes have caused a noticeable change in nuclide concentration, by this causing a change in resonance selfshielding effects and in neutron spectrum distribution; therefore the effective cross sections have to be redetermined at these time steps. As a rule, the resonance selfshielding of the burnt isotopes decreases with increasing burn up, the resonance shielding for the converted isotopes increases with increasing burn up.

In order to get an impression of the magnitude of the changes in effective cross sections during burn up, for the PWR-Obrigheim the effective absorption cross section decreases by about 10 % for Pu240 from 2 MWd/t up to 20000 MWd/t_1 /7/. Whether a 1/E-weighting spectrum is used for collapsing to coarse energy-group mesh or a detailed many group system-spectrum, causes for 2000 MWd/t burn up about 5 % change in the effective σ_a between 1.1 eV and 6.5 eV; including mutual self-shielding of various resonance isotopes the differencies in σ_a may add up to about 30 %. The influence of the U238 resonance on the cross sections of Pu240 is generally smaller and amounts to about 2 %. A further illustration is given in Fig. 1, where the influence of various Pu-contents in a LWR pin according to various burn-up, states on neutron spectrum in the vicinity of the U238 resonance is shown. In Tab. 1, the energy dependence of the production rates of the main isotopes at the end of the first cycle of the Obrigheim PWR is given, illustrating that for Pu240 the range 0-1.01 eV (including the high Pu240 resonance),

for Pu241 the epithermal range up to 50 eV is especially. important.

	10,5MeV -10keV	10keV -46,5eV	46,5eV -1,01eV	1,01eV -0eV
Pu239	10,7	29,7	37,2	22,3
Pu240	0,1	2,4	4,7	92,7
Pu241	0,1	1,1	61,1	37,7

Tab.1: Production rates of Pu239, Pu240 and Pu241 as function of energy for the PWR Obrigheim (ca. 13000 MWd/t)

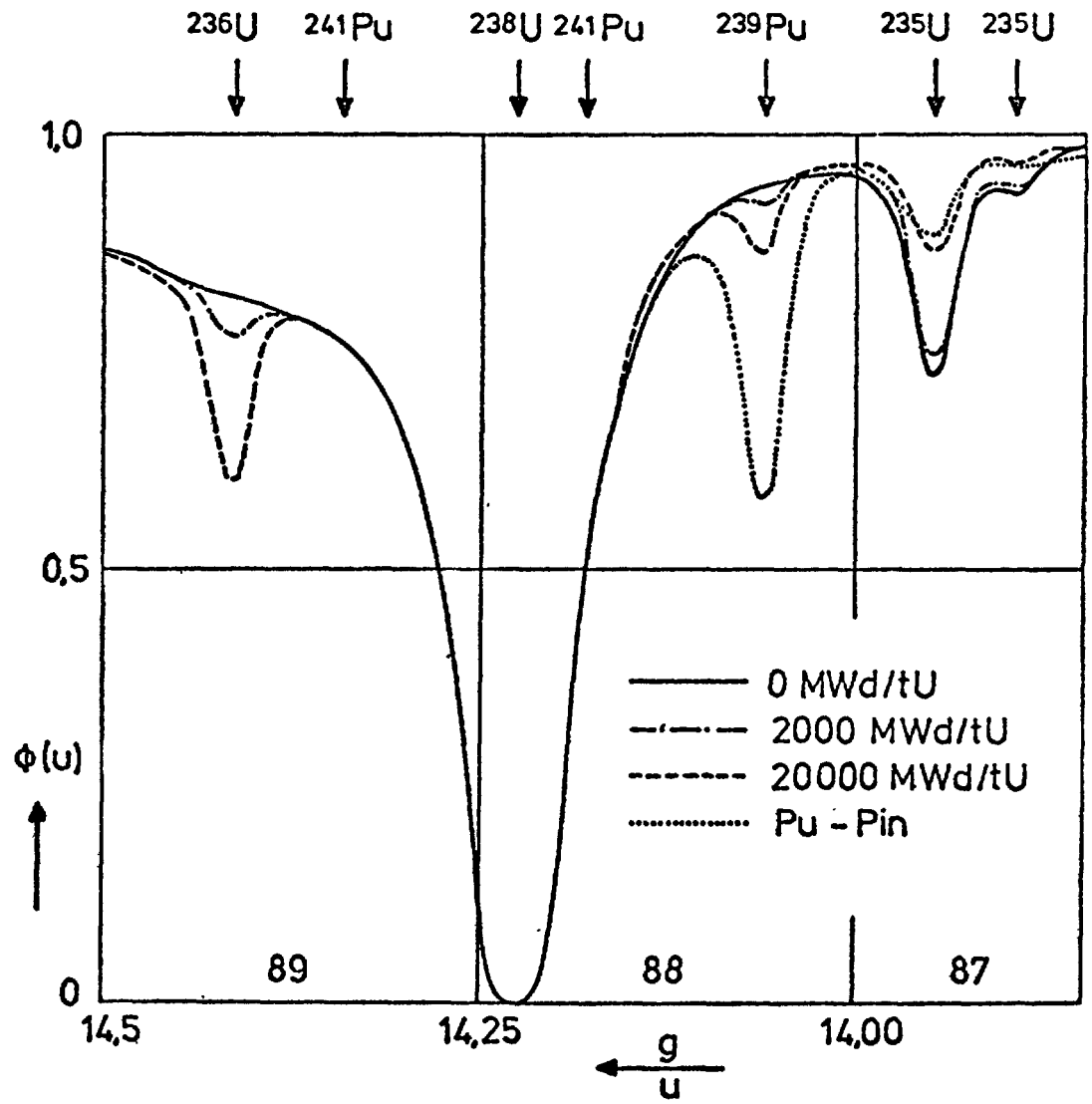


Fig.1: Resonance spectra in a PWR pin for various Pu concentrations, around the 6,67 eV resonance o- U 238.

In addition, one has to be careful in choosing the coarse energy group structure in the whole core calculations. For instance, during the analysis of the Garigliano BWR /13/ two and five group calculations were performed and compared with experiment by determined nuclide concentrations. Two groups were obtained by collapsing the 3 fast neutron groups and the two thermal groups of the 5 group set to one fast and one thermal group, respectively. The results are given in Tab. 2.

Isotope	Five groups	Two groups
U235	- 4	- 6
U236	- 7	- 7
U238	+ 0.07	+ 0.2
Pu239	+ 4	- 25
Pu240	+ 3	+ 8
Pu241	± 10	± 9
Pu242	+ 25.5	± 8.5

Tab.2: Average deviations of isotopic contents in [%] obtained from two-group and five-group calculations, compared to experiment.

The five-group calculations improve the agreement with experiment except for Pu241 and Pu242, indicating that either simplifications in the collapsing procedure were made or that even 5 groups are insufficient.

Other methodical questions are also of importance on the burn-up behaviour of LWR fuel: the spatial location of the fuel pins near control blades or control rods, or adjacent to watergaps and reflectors require special attention, if burn up predictions of these fuel elements are requested to an accuracy of better than 20 %. As an example, in /13/ the inadequate representation of the control blade in the Garigliano BWR causes about 10 % deviation in the burn up of neighbouring fuel elements. In the analysis of the Italian TRINO-BWR fuel that effect is made responsible for a local 18 % deviation from experiment.

In concluding this section, the errors in predicting the transactinide concentrations in LWRs during burn up may turn

out to be 20 % and even more if simplified methods are being used in describing geometry and the detailed neutron spectrum.

In chapter 5 we will demonstrate to what accuracy present methods and data are capable to interpret post irradiation experiments.

3.3 Fast Reactors

From the point of view of theoretical methods, the fast reactor is somewhat simpler to calculate because of the larger neutron mean free paths, so that cell heterogeneity is not very dominant. Furthermore, due to the larger internal conversion of fertile to fissile material, the burn up dependence of the effective cross sections of the various nuclides is not as much pronounced as in thermal reactors. Conventional designs of fast reactors do have an appreciable amount of nuclide conversion in the first few rows of the radial blankets. More modern designs do investigate now, for achieving a better breeding performance and a reduced coolant void reactivity, ring core designs. For a proper prediction of actinides of these cores at least 2 dimensional time dependent diffusion calculations have to be performed.

It should be mentioned here that very often fundamental mode calculations are performed in the frame of α-waste forecasts in a developing reactor population, for instance by using the Oak Ridge Code ORIGEN /8/. This code is unable to describe properly the in-reactor material behaviour with time. Even if nuclear data are adjusted to represent average actinide concentrations of a more defined calculation, the resulting deviations are of the order -60 to +30 %, for important nuclides, as is shown in Tab. 3 /9/ (note, that the U236 concentration deviates by a factor of 7).

It should be mentioned that similar scatter is observed by adjusting ORIGEN-data to a PWR data assay.

ORIGEN is mainly used for waste hazard projections; but we should bear in mind this order of uncertainty, which has to be compared with the consequences of uncertain nuclear data. This will be done in chapter 6 of this paper.

Nuclide	Deviation of nuclide concentrations
U236	factor 7
Np237	- 60 %
Pu238	- 20 %
Pu240	6 %
Pu 241	10 %
Pu242	- 30 %
Am241	- 20 %
Am242	-
Am243	- 25 %
Cm242	-
Cm243	-
Cm244	30 %

Tab.3: Deviation of nuclide concentrations with "adjusted" data in ORIGEN compared with a more refined calculation for a 1000 MWe LMFBR.

4. Build-up of Transactinides in Different Reactor Types and Dependence on Reactor Operation.

In this chapter we present the production of transactinides in 1000 MWe power plants. The data are collected from ref. /6/ and /10/. In /6/ the special aspect is considered, whether it is worthwile to recycle the transactinides together with Pu in a BWR, or whether these nuclides should be separated from the heavy metal batches. The assumptions for the Pu-recycling and uranium cases in /6/ and /10/ are not directly comparable, but for the purpose of demonstrating the large differences in actinide build-up in various operating schemes, the data are very instructive. Also for the fast reactor one can find different figures for the various nuclides, depending on fuel management schemes and assumed burn-up, thus all the numbers in Tab. 4 are indicative only. Generally it can be stated, that it is by no means satisfactory for practical application, to predict actinide production or the amount of actinides to be reprocessed yearly, for idealized reactor types with some fuel management scheme.

This is so, because the differences in the heavy radionuclide concentrations amount to a factor of 10 for various types and operation schemes.

Note that the LWR with Pu- and high actinide recycling has to be regarded as the worst case with respect to its potential for radiation hazard. This hazard is expressed in Table 4 by the yearly produced radioactivity in Ci. Again it can been seen that the differences amount up to a factor of 10.

The main radioactivity stems from the α-decay of Cm244, and Pu238 (5:1), β-decay of Pu241, Np239 and Am242 in ($\approx$1200:11:1) and γ radiation of Am241, Np239 ($\approx$50:6). Thus the large amount of these isotopes in a Pu-recycled LWR will cause problems in shielding requirements for shipping spent fuel and for reprocessing units. The recycling of the higher actinides will very probably yield an even worse situation. In a Pu-recycling strategy, this is the only possible way at present, because the seperation of these strong α-emitters very probably will not technically feasible for the next 2 decades.

In concluding this section, we find differences in actinide content and radiation up to about a factor of 10 depending on reactor type and various mode of operation. Again, this has to be kept in mind when data uncertainties are discussed in chapter 6.

5. Theoretical analysis of irradiation experiments on fuel from power reactors and of integral experiments in fast zero power reactors

This chapter will deal with the present capability to predict actinide build up in LWR reactors. In addition, the interpretation of reaction rate ratios for actinide isotopes, measured in fast zero power reactors, will be discussed.

5.1 Analysis of irradiation experiments

In the last few years many irradiation experiments have been performed to determine burn up and nuclide concentration with LWR fuel. Theoretical predictions of these quantities have been compared with experimental results. Mainly the analysis was

Isotope	Uranium-Fueled Water-Reactor		Uranium-Plutonium Water-Reactor		All Actinides Recycled Only Pu Recycled	Fast Breeder Reactor	
	[kg/y]	[Ci/y]	[kg/y]	[Ci/y]		[kg/y]	[Ci/y]
Pu236	0.02	$8.5 \cdot 10^3$	0.08	$4 \cdot 10^4$		$2\ 10^{-3}$	9
Pu238	4.5	$7.5 \cdot 10^4$	30	$5 \cdot 10^5$		15	$3 \cdot 10^5$
Pu239	145	$9 \cdot 10^3$	538	$3 \cdot 10^4$		1318	$8 \cdot 10^4$
Pu240	59	$1.3 \cdot 10^4$	494	$1 \cdot 10^5$		441	10^5
Pu241	27	$3 \cdot 10^6$	291	$3 \cdot 10^7$		120	10^7
Pu242	9	37	219	800		75	300
Am241	2	$6 \cdot 10^3$	32	10^5	1.9	11	$4 \cdot 10^4$
Am242M	0.01	$1 \cdot 10^2$	0.3	$3 \cdot 10^3$	2.2	0.2	$2 \cdot 10^3$
Am243	2.5	$5 \cdot 10^2$	43	$8 \cdot 10^3$	1.9	5.6	$1 \cdot 10^3$
Cm242	0.09	$3 \cdot 10^5$	0.9	$3 \cdot 10^6$	1.85	0.3	10^6
Cm243	0.02	10^2	0.02	$9 \cdot 10^2$		0.02	$8 \cdot 10^2$
Cm244	0.83	$7 \cdot 10^4$	8.8	$7 \cdot 10^5$	6.3	0.3	$3 \cdot 10^4$

Tab.4: The yearly production of heavy actinides [kg/y] and the associated activity [Ci/y]
The reactors in question are 1000 MWe power plants.

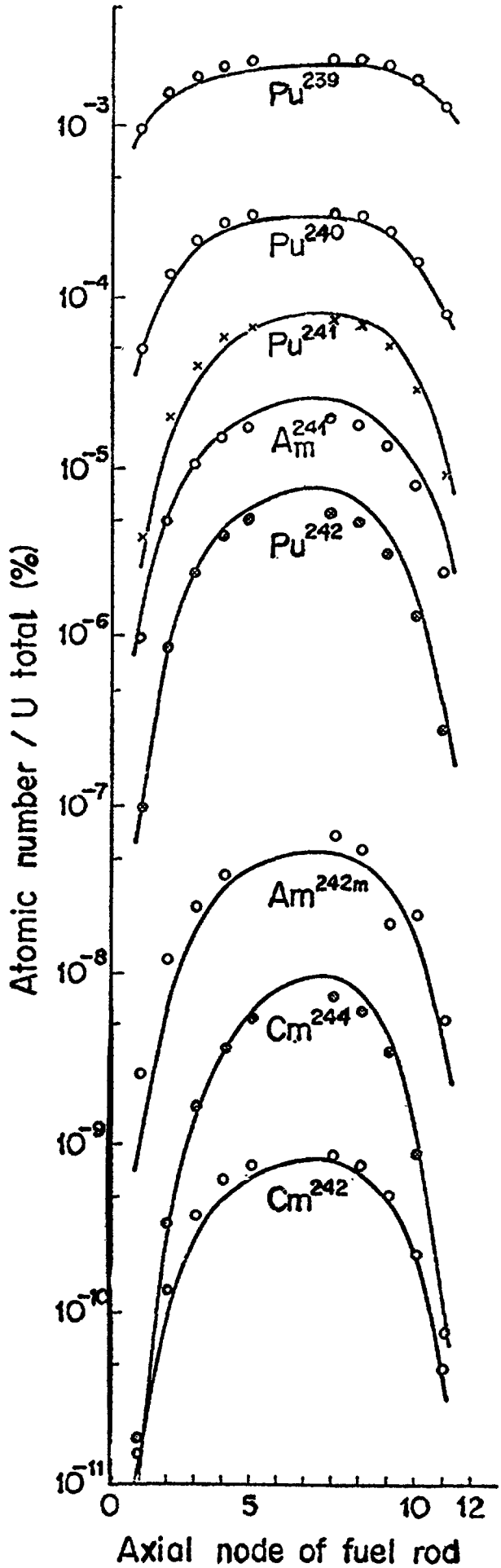


Fig.2: Comparison of experimental and theoretical (——) axial distributionsof actinides in JPDR-1.

concentrated on the determination of the U235, U236, Pu239, Pu240, Pu241 and Pu242 contents, very few experiments give information on the other actinide concentrations in spent fuel.

As a first example, the axial distribution of actinides in the 12.5 MWe BWR Japanese Power Reactor JPDR-1 for medium burn up is shown in Fig. 2 (note the logarithmic scale) /11/. The whole core calculations were performed using a 3 dimensional diffusion theory modal method of the FLARE type, nuclear data were taken from the Japanese Data Library . Transport theory (THERMOS in 30 groups) was applied for the calculation of the cell-flux distribution in the thermal energy region. It can be seen that no drastic differences show up in the comparison. While the main Pu isotope distributions are being predicted satisfactorily, the predicted concentrations Pu242, Am241, Am242M, Cm242 and Cm244 deviate by about 20 to 30 % from experiment.

A similar deviation is obtained in the analysis of the German 60MWth test reactor VAK (BWR) /12/: U238: ± 3 %, Pu239: ± 7 %, Pu240: ± 11 %, Pu241: ± 17 %, Pu242: ± 17 %. The results are regarded as satisfactory (1971) by the authors.

For the analysis of the Italian TRINO-BWR fuel /14/ an interesting cross check was made on the experimental techniques for burn-up determination between the Karlsruhe Transuranium Institute and the Ispra Center. Both laboratories give results, which are in close agreement: on the average 1 - 4 %. Differences of about 1 % between the Nd148 and Cs137 techniques are attributed to uncertainties in the fission yield data. The burn-up is predicted theoretically to better than 2,3 % on the average, the isotopic contents of U235, U236 and U238 to better than or about 3 %, the Pu nuclides to about 10 - 15 % with some exceptions.

The analysis of the US-Saxton II-PWR core was performed with modified ENDFB/3 data /15/.
The comparison between calculations (C) and experiments (E) for the main chain U and Pu isotopes and for Np, Pu, Am and Cm isotope concentrations at end of life (22000 MWd/t) are given in Tab. 5 and Tab. 6, respectively. In these tables also the relative uncertainty of the measurements is given. The largest experimental uncertainty is connected with the determination of U234 (30 %) and Pu238 (24 %).

Tab.5: Comparison between measurement and calculation of the main chain U- and Pu-isotopes in Saxton Core II mixed-oxide fuel

Parameter	Percent Relative Uncertainty in Measurement (a)	Percent Difference between Calculation and Measurement
		Pellet
U234 Atom Percent	29.4	2.9
U235 Atom Percent	0.9	0.8
U236 Atom Percent	5.6	5.2
U238 Atom Percent	0.01	0.00
Pu238 Atom Percent	2.3	-24.6
Pu239 Atom Percent	0.03	0.86
Pu240 Atom Percent	0.2	2.2
Pu241 Atom Percent	0.3	3.6
Pu242 Atom Percent	0.9	0.4
Pu239/U238 Atom Ratio	0.7	- 4.8
Pu/U Mass Ratio	0.7	- 3.9

Notes: (a) Two standards deviation precision.

It is seen that the content in Np237, Pu238 and the Curium isotopes are underestimated by about 30 - 40 %.

In conclusion of this section, the theoretical analysis of LWR fuel elements results in a deviation of the actinide contents to experiment of up to about 40 %; this figure is related to Cm244. which together with Pu238 constitutes the strongest α particle source in LWRs.

5.2 Theoretical interpretion of actinide reaction rate ratios in fast zero power assemblies

Fission rate ratios have been determined in the fast zero power reactors ZEBRA 14 /16/ and SNEAK /17/. The results are given in Tab. 7.

Tab.6: Comparison between measurement and Leopard-HIC calculation of Np, Pu, Am, and Cm Isotopes in Saxton Core II mixed-oxide fuel

Isotope	Measured Parameter	Percent[(a)] Relative Uncertainty in Measurement	Percent[(b)] Difference between Calculation and Measurement
Np237	Np237 dpm/g U	± 15	- 34
Pu236	Pu236/Pu239	± 24	8
Pu238	Pu238/Pu239	± 4	- 28
Am241	Am241/Pu239	± 24	- 4
Am243	Am243/Pu239	measured data not resolved	-
Cm242	Cm242/Pu239	± 10	- 24
Cm244	Cm244/Pu239	± 20	- 38

Notes: (a) Two standard deviations. Referenced to core end-of-life.

(b) (Calc-Meas)/Meas x 100 percent; LEOPARD-HIC axial Zone 6 calculation at about 20000 MWD/MTM.

Isotope	σ_f/σ_f (Pu239) in ZEBRA 14	σ_f/σ_f (Pu239) in SNEAK 9C-2	$1+\alpha$ in SNEAK 9C-2
U238	1.04 ± 4 %	0.95 ± 2,2 %	0.98 ± 5 %
Pu240	1.003 ± 5 %	0.94 ± 1,5 %	1.27 ± 4 %
Pu241	1.05 ± 3 %	1.05 ± 1.5 %	1.03 ± 10 %
Pu242	1.23 ± 5 %		
Am241	1.26 ± 4 %	1.40 ± 1.8 %	1.95 ± 4 %
Am243	0.88 ± 4 %		
Cm244	1.35 ± 8 %		

Tab.7: C/E values for fission rate ratios of actinides in fast zero power reactors:

Because both the experimental and theoretical assessment has been done with some sophistication, most of the deviations are due to data uncertainties, related to the group sets used in the UK and Germany.

There is a clear indication that the fission data of Pu242, Am241, Am243 and Cm244 are uncertain to between 10 to 40 %.

In SNEAK 9 C also 1 + α for Am241 has been measured. The theoretical value differs by about a factor of 2 from the experimental one.
It should be mentioned that the data basis for the higher actinides both in the UK and in Karlsruhe is ENDFB/2 or slightly modified data, which are also included in ENDFB/4.

6. Investigation of the sensitivity of actinide build up and decay due to nuclear data changes

In the preceding chapter 4 it has been shown that different reactor types and different operation schemes have a large influence on the amount of higher actinides and on the associated radiation hazard (up to a factor of 10). In chapter 5 it was demonstrated that the present uncertainty in the prediction of isotopic contents may amount to about 40 % for important radiation emitters; fission rate ratios can be predicted to a simular accuracy, the capture in Am241 seems to be uncertain to an even larger degree. This background has to be kept in mind when the effects to cross section changes are investigated. In order to make a first assessment, it appears reasonable not to use sophisticated theoretical tools for obtaining the effects of the many changes in cross sections and decay properties for transactinides. In addition, one is interested not only in the uncertainties of isotopic contents of the actinides during reactor operation, but also in the uncertainties of the radiation hazards during shipping and reprocessing of fuel and the fuel waste. In order to have a first indication of cross section effects, we used simple fundamental mode calculations for describing the fuel life including fuel waste storage.

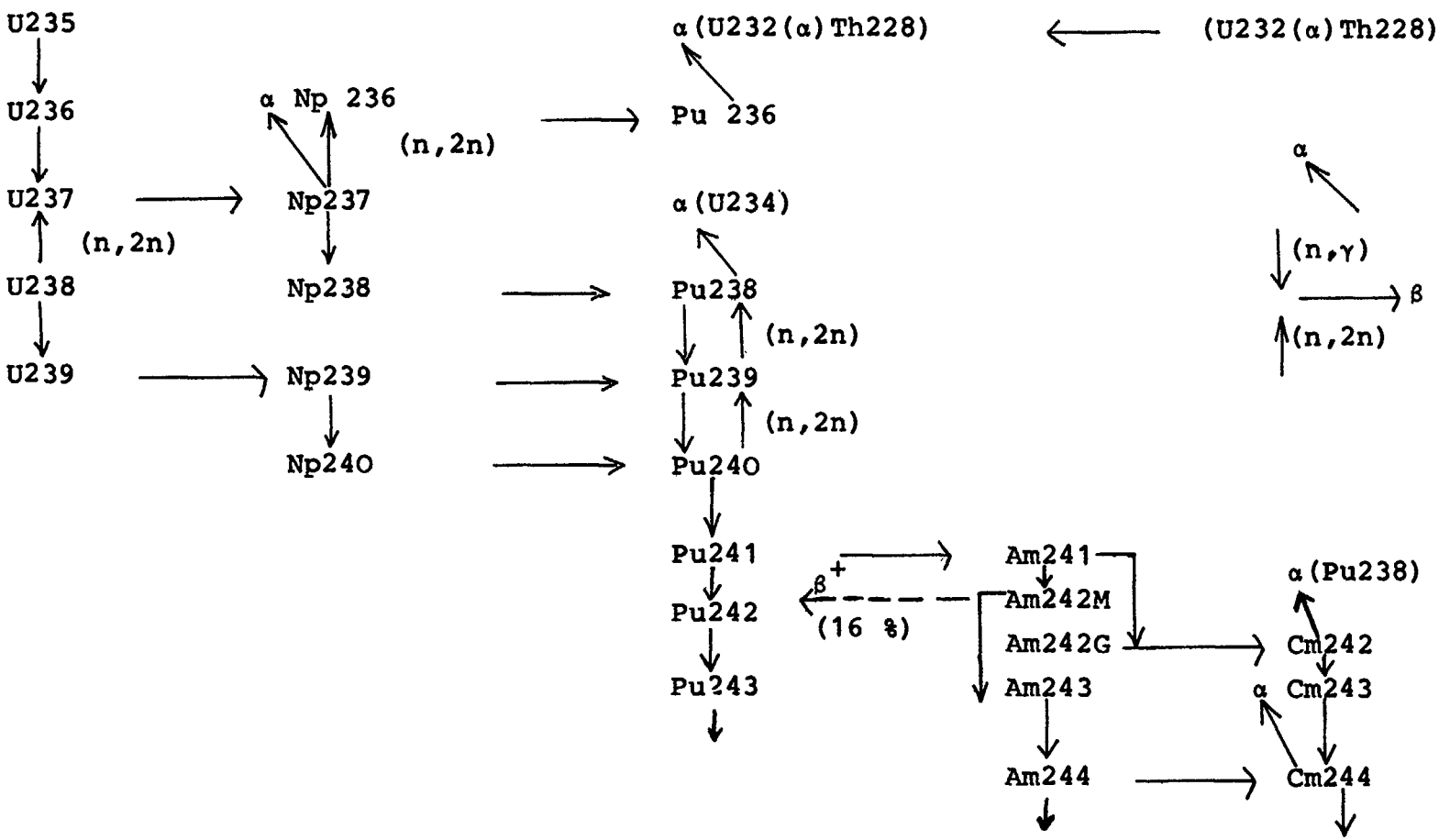
6.1 The importance of transactinide isotopes in the reactor fuel cycle

This section summarizes the reasons for the interest in transactinide isotopes. In Tab. 8 the nuclear data for these nuclides together with a comment on associated effects (last column), are given. The data are taken from ref. /8/ for a LWR, LMFBR and HTGR system.

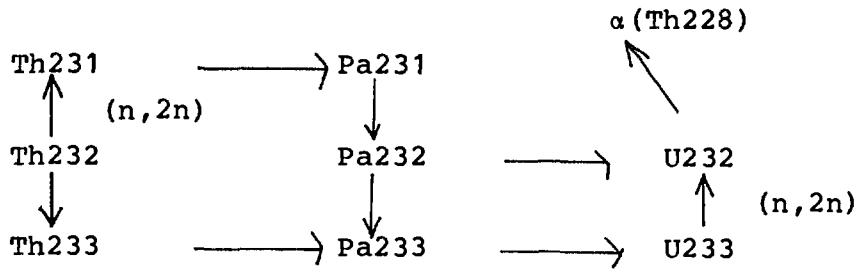
Tab. 8:

ISOTOPE	$\tau_{1/2}$	$\overline{\sigma_\gamma}$ (barns)		$\overline{\sigma_f}$ (barns)		$\overline{RI}_\gamma$	$\overline{RI}_f$	Effect of interest
		LWR	FBR	LWR	FBR	LWR	LWR	
Th232	$\approx 10^9$y	7.4	0.44		0.0014	83.		reactivity
Pa231	3.2 10^4y	200.	0.80		0.38	480.		reactivity, rad. precursor -
U233	1.8 10^5y	49.	0.39	525.	3.15		746.	reactivity
U236	24 10^6y	6.	0.66	0.	0.11	210.	0.	reactivity
Np237	2.1 10^6y	170.	0.76	.002	0.36	756.	0.	Pu38 build up $\alpha + \gamma$ source
Np239	2.3d	60.	0.8	0.	0.36	415.	0.	β source, reactivity
Pu236	2.85y	0.	0.22	170.	1.40	0.	0.	α-source, rad. precursor
Pu238	89.y	500.	0.22	17.	1.4	150.	25.	α + n source
Pu240	6760.y	366.	0.41	-	0.35	2000	-	reactivity, α source
Pu241	14.6y	550.	0.432	1480.	2.5	140.	537.	reactivity, β source
Pu242	3.8 10^5y	18.5	0.34	.003	0.28	1280	.6	reactivity, α source
Am241	433.y	925.	0.99	3.1	0.46	1500.	0.	reactivity $\alpha + \gamma$ source
Am242G	16hr.	0.	0.43	2900.	1.83	0.	0.	$\beta+\gamma$ source
Am242M	152y	2000.	0.40	6000.	1.83	0.	0.	α source
Am243	7650.y	105.	0.55	0.45	0.24	1500.	1.5	α source
Cm242	163D	30.	0.38	5.	0.42	0.	0.	α + n source
Cm243	32.y	200.	0.40	600.	0.32	500.	1850.	α source
Cm244	18.1y	10.	0.37	1.2	0.41	650.	12.	α + n source

Tab.9: U and Pu Chains, Main Isotopes



In Tab. 9 the route for build up of the main isotopes in the U and Pu chains are given. These isotopes are of concern in LWR and LMFBR systems. We will not give here the complete routes for the thorium cycle in HTGRs, only the important ones in Tab. 10.



Tab. 10: Main Isotopes of the Th-Chain.

According to the listed nuclear data in Tab. 8 and the isotope chains in Tab. 9 and Tab. 10, we can find the following importance of transactinides in reactor systems:

a. Reactor operation:
The main interest here is the contribution of the transactinides to the reactivity of the system. Of importance are: U236, Pu240, Pu241 and to a minor extent Np239 and Am241 at end of cycle or in LWR systems with actinide recycling.

b. Refuelling operations and fuel management on load:
Again the introduced reactivity is of major concern. In addition to the content of fissile and fertile nuclides as U235, U238, Pu239 and U236, Pu240, Pu241, Am241 (minor) and the contributions from spontaneous fission neutrons from Cm242 and Cm244 have to be considered.

c. Reactor shutdown:
Two aspects are of importance: first the shutdown reactivity has to be within the requested bounds and this has to be safely indicated by monitors. Again the spontaneous fission source from Cm242 and Cm244 has to be taken into account. Secondly, the heat, mainly produced by β-and γ emission, up to some hours after shutdown has to be predicted to an accuracy of less than 20 %. The main contributors besides the fission products are the transactinides U239, Np239, Pu241, Am241 (γ), Am242G and of Th233, Pa233 for a system operating in the thorium cycle.

d. Transport,reprocessing and refabrication of fuel:

After a cooling period of some hundred days up to a few years, discharged fuel has to be shipped for reprocessing. Cooling and shielding of the transportation casks have to be designed. α and β emitters with corresponding half lives as U239, Np239, Pu238, Pu241, Th233, Pa233 and spontaneous fission neutrons from Pu238 and Cm244 as well as neutrons from (α,n)do contribute to the radiation level. In a thorium system U232 and the hard γ-emitter daughter Th228 are of importance.

e. Fuel Waste:

Non recovered fuel (waste) has to be stored safely. Besides the Pu- and fission product radiation the long lived transactinide α-emitters are of great importance: Np237, Pu238, Am241, Am242, Am243, Cm243, Cm244 and Pa231, U232.

6.2 Procedure of Investigations

In this section we outline briefly the procedure for obtaining the response of interesting physical quantities by cross section changes. As expressed earlier in this chapter, it is aimed in keeping the study as simple as possible, but nevertheless meaningful with respect to a first indication of relative deviations as consequence of nuclear data variations. Therefore fundamental mode calculations with ORIGEN were performed for typical 1000 MWe LWRs, HTGRs and a 2000 MWe LMBFR, excluding fuel management schemes. From these calculations we cannot deduce true cost-benefit conclusions with respect to a definite accuracy of transactinium nuclear data for a given system. But a thourough cost-benefit analysis can hardly be established presently, probably with the exception for the main uranium and plutonium isotopes, which determine the fuel burn up; these investigations have been performed already within the frame of thermal and fast reactor projects.

The cross sections for the main uranium and plutonium isotopes were estimated to be uncertain to about $\pm$ 20 % (this is truly pessimistic for U235, U238 and Pu239). From the discussion in the preceding chapter on the theoretical assessment of post-irradiation and integral zero power experiments we attributed a $\pm$ 50 % uncertainty to the basic cross sections of the other actinides; (n,2n) cross section were changed by 100 %. The half

Tab.11: Relativechanges of nuclear data, used in the present sensitivity study.

		LMFBR	PWR	HTGR
ISOTOP	σ	$\delta\sigma$ [%]		
Th232	(n,γ)			20
	$(RI)_\gamma$			20
	$(n,2n)$			100
Pa231	(n,γ)			50
U233	$(RI)_f$			- 20
U235	$(RI)_\gamma$		20	
U236	$(RI)_\gamma$		50	20
U238	(n,γ)	20	20	
	$(RI)_\gamma$		20	
	$(n,2m)$	50		
N237	(n,γ)	50	50	
	$(n,2n)$	100	100	
Np239	(n,γ)	50	50	
Pu239	(n,γ)	20		
	(n,f)	- 20	- 20	
	$(n,2n)$	100		
Pu240	(n,γ)	20		20
	$(RI)_\gamma$		50	
Pu241	(n,γ)	- 20		
	(n,f)	- 20		
Am241	(n,γ)	- 50	+ 50	- 50
	(n,f)	- 50		
Cm242	(n,γ)	50	50	
Cm243	(n,γ)	50	50	

lives of nuclides were varied according to the listed uncertainties in ref /18/; these uncertainties are below about 10 %. The changes are summarized in Tab. 11.

These variations were introduced into the ORIGEN data library. In addition, we replaced some of the ORIGEN data by more recently evaluated data of ref /2/. The application was done for a 2000 MWe LMBFR system, which is forseen to be of interest in the future. For this system we also did separate investigations for core and blanket fuel performance with the appropriate material and neutron flux densities.

In the following tables, there is also listed the response of the systems investigated, to a cumulative change of nuclear data. These data changes are taken from Tab. 11, with the corresponding + or - signs.

The changes due to these variations for each isotope were investigated for following quantities:

- nuclide concentrations during operation of the reactor and after discharge of fuel, including cooling times before reprocessing.

- heat production of discharged fuel for various cooling times.

- neutron source intensity due to spontaneous fission and (α,n) processes at and after discharge of fuel.

- the radioactive hazard potential of discharged fuel and of fuel waste.

6.3 Results of the performed sensitivity studies

As a first example, for a 1000 MWe-PWR the relative comulative change of transactinide concentration at discharge of the fuel is given in Tab. 12.

Relative Cumulative Change of Concentrations (%) at Discharge										
ELEMENTS				ISOTOPES						
Pu	Am	Cm	Np	U6	Np7	Pu8	Pu41	Am41	Cu42	Cu44
28	31	26	37	1	39	49	37	35	42	24

Tab.12: Relative Cumulative Change of Concentration for a 1000 MWe PWR.

The discharge point was chosen, because the deviations in some nuclide concentrations during operation reach a maximum value in our studies. As can be seen, the reasonable data changes cumulate up to about 50 % for the high actinide concentrations. In reality, the data changes might compensate thus that the true response becomes smaller. Compared to the uncertainties, which may arise from the methodical description in LWRs and from different operation schemes, the obtained uncertainties due to data changes are not too overwhelming. As a general rule, the reactivity effects are clearly dominated during operation from the most important isotopes as U235, U238, Pu239, Pu240 and Pu241. The data for these isotopes have to be known to better than 10 %, partly even 5 % as is well known from the data request lists. The change in Cm concentration, especially with respect to the production of spontaneous fission sources, is not to be neglected, but nevertheless, this change has not to be known to an accuracy better than about 20 %. Therefore we conclude that one should aim at an accuracy of about 30 % for the basic nuclear data in question for special actinides.

We have studied also the effect of special cross section changes as Np237 capture and in addition, Np239 capture and Np237 (n,2n) with respect to the reference case. The resulting effects are small. No special requirements for these cross sections can be deduced from our study.

For the investigation of integral quantities of discharged fuel and waste, the effect of cumulative data changes amount up to about 40 %, see Tab. 13. The quoted hazard index is defined as that quantity of water, which is needed to dilute the concentration

of the radionuclides to the maximum permissible concentration with respect to radiation dose. In the present situation it is not clear whether this uncertainty can be tolerated; in any case, if an accuracy of about 30 % for the nuclear data is achieved, very probably no major obstacles might arise in the near future.

The real problems today are connected with a convincing technological assessment of fuel reprocessing with regard to an optimal resolving and separation scheme, together with a proper definition of the routes for the long-lived α-emitters.

Nuclide	Nucl.data Change δσ%	DISCHARGED FUEL									WASTE		
		HAZARD-INDEX			THERM.-POWER			SPONT.FISS.			HAZARD-INDEX		
		0D	150D	1Y	0D	150D	1Y	0D	150D	1Y	10Y	10^2Y	10^3Y
U235	$\delta(RI)_c$=20	2.	4.	5.	2.	3.	3.	2.	2.	3.	3.	1.	1.
U236	$\delta(RI)_c$=50	4.	10.	13.	0.	5.	8.	0.	0.	0.	0.	0.	0.
U238	δCAP.=20	0.	0.	0.	0.	0.	0.	-1.	-1.	-1.	-2.	0.	0.
	$\delta(RI)_c$=20	1.	-3.	-5.	2.	0.	-4.	-10.	-12.	-13.	-11.	4.	4.
N237	δCAP.=50	0.	0.	0.	0.	0.	0.	0.	0.	0.	0.	0.	0.
	δ(n,2n)=100	0.	0.	0.	0.	0.	0.	0.	0.	0.	0.	0.	0.
Np239	δCAP.=50	0.	0.	0.	0.	0.	0.	0.	0.	0.	0.	0.	0.
Pu240	$\delta(RI)_c$=50	-1.	8.	7.	0.	11.	9.	15.	14.	15.	15.	11.	10.
Pu241	δFISS.=-20	1.	6.	5.	0.	7.	6.	8.	8.	8.	8.	8.	8.
Cumulative Effect	δSIGMA	12.	38.	37.	11.	40.	39.	29.	26.	25.	26.	35.	31.
Reference Values		$2.9 \cdot 10^{11}$ [m³]			10^5 [WATTS]			$1.7 \cdot 10^9 \left[\frac{\text{NEUTR}}{\text{SEC}}\right]$			$3.2 \cdot 10^9$ [m³]		

Tab.13: Relative Changes of Integral Quantities for Discharged Fuel and Waste of a 1000 MWe PWR.

The question of data uncertainties has to be taken up again if long term strategies for handling all the fuel of the growing reactor population have been decided. This has not been investigated here.

For a U233-HTGR system, corresponding results are given in Tab. 14 and Tab. 15. We do obtain an appreciable uncertainty in the U232 and U233 contents, U232 being the precursor for the strong radiation from Th228. Here the data of Th232, with a great importance also (n,2n), and the Pa data are essential. In addition, Pa233 radiation at discharge and transport of HTGR fuel is large. The build up of Pa232 strongly depends on the (n,γ) cross sections of Th232, as can be seen from Tab. 10, the (n,2n) route via Pa231 and Pa232 is of minor importance in this case. If the U233 cycle is used in HTGRs, similar arguments as for the LWR system hold: those basic data which are connected with the primary isotopes, very clearly have to be known to an accuracy of about 5 - 10 %, relaxing to 20 to 50 % with respect to (n,2n) processes. For the other actinide isotopes again a 30 % accuracy requirement for the basic data seems reasonable.

In Tab. 16 we have listed the effects of data changes for a 2000 MWe LMFBR system. This system is started with Pu fuel obtained from LWRs. Especially we considered the influence of data changes, according to Tab. 11, from "neighbouring" nuclides to Am and Cm. The results in concentration changes are up to about 50 %, somewhat lower figures are obtained for thermal power production and for the spontaneous fission source. To the reactivity the main U and Pu isotopes contribute essentially, therefore the data of these isotopes have to be known according to present request lists. As in the LWR system, the spontaneous fission from Cm has to be known to some detail; the strong dependence on the capture and fission data of Am and Cm presently indicates that again a 30 % uncertainty of the corresponding data would not create great problems for reactor operation and shut down margins. The major interest is related to the emission of radiation (including neutrons) for transport, reprocessing and waste management. This aspect is given in Tab. 17 for a 1000 MWe system, where core and blanket elements are reprocessed commonly. The cumulative effects yield about a 20 % difference in

Relative Cumulative Change of Concentrations (%) at Discharge					
ELEMENTS		ISOTOPES			
Pa	U	Pa231	Pa233	U232	U233
9	12	80	8	120	32

Tab.14: Relative Cumulative Change of Concentration for a 1000 MWe HTGR.

Isotope	Nucl.Data Change δ%	DISCHARGED FUEL									WASTE		
		HAZARD-INDEX			THERM.POWER			SPONT.FISS.			HAZARD-INDEX		
		OD	150D	1Y	OD	150D	1Y	OD	150D	1Y	30Y	10^2Y	10^3Y
Th232	δCAP.=20	1.	0.	3.	0.	0.	3.	9.	9.	10.	5.	3.	1.
	$\delta(RI)_c$=-20	2.	0.	8.	-2.	0.	8.	24.	24.	25.	13.	8.	3.
	δ(n,2n)=100	0.	0.	2.	0.	1.	4.	0.	0.	0.	6.	13.	46.
U233	δ(RI)f=-20	3.	5.	8.	3.	5.	9.	25.	26.	27.	15.	9.	8.
U236	$\delta(RI)_c$=+20	2.	6.	15.	1.	6.	15.	3.	3.	3.	9.	11.	2.
Pu240	δCAP=20	0.	0.	0.	0.	0.	0.	1.	1.	1.	0.	0.	0.
Am241	δCAP=-50	0.	0.	0.	0.	0.	0.	-5.	-3.	-2.	0.	0.	1.
Cumulative Effect	δSIGMA	7.	12.	17.	7.	11.	19.	-4.	-2.	0.	15.	24.	52.
Reference Values		$3.8 \cdot 10^{11}$ [m³]			$1.3 \cdot 10^5$ [WATTS]			$8.2 \cdot 10^7 \left[\frac{\text{NEUTR.}}{\text{SEC}}\right]$			$6.7 \cdot 10^9$ [m³]		

Tab.15: Relative Changes of Integral Quantities for Discharged Fuel and Waste of a 1000 MWe HTGR.

Isotope	Nucl. data change δσ|%|	Relative Change (%) after Discharge								
		Concentration	Therm. Power			Spont. Fiss			Haz. Index	
		OD	OD	150D	1Y	OD	150D	1Y	OD	150D	1Y
Pu241 Am241	δ CAP = -20 δ FISS= -20 δ CAP = -50 δ FISS= -50	Pu238 -37. Pu241 7. Am241 8. Am242M -47. Cm242 -47. Cm244 5.	0.	-33.	-23.	-44.	-30.	-20.	1.	5.	0.
Am241 Cm242 Cm243	δ CAP = 50 δ FISS= -50 δ CAP = -50 δ CAP = 50	Pu238 40. Am241 - 2. Am242M 50. Cm242 50. Cm244 0.	2.	36.	25.	40.	33.	23.	1.	10.	5.
Am243 Cm242 Cm243 Cm244	δ CAP = 50 δ CAP = 50 δ CAP = 50 δ CAP = -50	Am243 - 1. Cm242 0. Cm244 41.	0.	0.	0.	6.	9.	15.	0.	0.	0.
Reference Values			$2.5 \cdot 10^5$ [WATTS]			$1.5 \cdot 10^9 \left[\frac{\text{NEUTR.}}{\text{sec.}}\right]$			$7 \cdot 10^{11}$ [m^3]		

Table 16: Results of Data Changes for a 2000 MWe LMFBR.

the corresponding integral quantities. From these figures again we conclude that a 30 % accuracy for the transactinium data might be sufficient at the moment.

As a last point, we discuss the effects, obtained by changing the data of Np237, Pu238, Am241 and Cm242 according to the evaluation of ref. /2/. The differences obtained in nuclide concentration, thermal power in spent fuel elements and the hazard index of fuel and waste are mostly much less than 20 %, this figure occuring only for the effects of Cm. Thus we have an indication for possible compensation effects, because these differences are less than those obtained earlier in this paper.

Isotope	Nucl. data change [%]	Discharged Fuel									Waste		
		Hazard Index			Therm.Power			Spont Fiss			Hazard Index		
		OD	150D	1Y	OD	150D	1Y	OD	150D	1Y	30Y	10^2Y	10^3Y
U238	δ CAP =20	11.	1.	2.	11.	0.	1.	-5.	-5.	-4.	0.	0.	0.
Pu239	δ CAP =20	1.	1.	2.	1.	2.	2.	1.	1.	1.	1.	2.	3.
	δ FISS=-20	10.	5.	5.	10.	8.	7.	12.	12.	12.	3.	2.	3
Pu240	δ CAP =20	0.	4.	4.	0.	3.	1.	0.	0.	0.	6.	7.	1.
	δλ =-8	0.	2.	2.	0.	2.	2.	0.	0.	0.	0.	0.	0.
Pu241	δ CAP =-20	0.	0.	0.	0.	0.	0.	0.	0.	0.	0.	0.	0.
	δ FISS=-20	0.	3.	3.	0.	2.	1.	0.	0.	0.	5.	5.	4.
	δλ = 7	0.	-3.	-3.	0.	-3.	-3.	0.	0.	0.	0.	0.	0.
Am241	δ CAP=-50	0.	-7.	-3.	0.	-24.	-14.	0.	0.	0.	0.	0.	2.
	δ FISS=-50	0.	0.	0.	0.	0.	0.	-34.	-27.	-17.	0.	0.	0.
	δλ =- 7	0.	0.	0.	0.	0.	0.	0.	0.	0.	0.	0.	0.
Cm242	δ CAP =-50	0.	0.	0.	0.	0.	0.	0.	0.	0.	0.	0.	0.
Cumulative Effect	δ SIGMA	22.	11.	15.	22.	-12.	0.	-23.	-15.	-5.	18.	20.	20.
	δλ	0.	0.	0.	0.	0.	0.	- 3.	- 1.	-1.	0.	0.	-6.
Reference Values		$5.5 \cdot 10^{11}$ [m^3]			$2 \cdot 10^5$ [WATTS]			$1.4 \cdot 10^8$ [neutr./sec.]			$7 \cdot 10^8$ [m^3]		

Table 17: Relative Changes of Integral Quantities for Discharge Fuel and Waste of a 1000 MWe LMFBR with Mixed Core/Blanket Reprocessing.

7. Conclusion

As has been discussed in the preceding chapters, especially in 6.1 and 6.2, an accuracy of the basic data for trans-actinium isotopes is being estimated to about 30 % for capture and fission, relaxing on (n,2n) processes up to 50 %. In this request the data for the main isotopes of Th, U and Pu are excluded; they have to be known to an accuracy already known by existing request lists. It would be appreciated , if further integral experiments, as discussed in chapter 5, could be performed. In fuel cycle strategies for a growing reactor population these accuracy limits might have to be narrowed, if the

technological assessment has been developed further. Because the present uncertainties of nuclear data are larger than the figures requested here at least for the epithermal and fast energy range, one should try to achieve the goal within the next 5 years. At that time a more precise knowledge on further developing fuel cycle schemes and strategies very probably will be available so that the present request has to be reinvestigated including sound cost-benefit analyses.

List of References:

/ 1/ Pigford T.H., Enviromental Aspects of Nuclear Energy Production Anu.Rev.Nucl.Sci.24(1974)

/ 2/ Hinkelmann, B. Report KFK 1186 EANDC 128-U (1970)

/ 3/ Lederer C.M., Hollander J.M. and Perlmans, Table of Isotopes, 6th ed. John Wiley and sons, N.W. (1967)

/ 4/ Review of Nuclear Data for Transactinides, Session B, This Conference.

/ 5/ Nodvik, R.J., Report WCAP-3385-56 (1970)

/ 6/ Johansson, E., Report TPM-RF-74-1065 (1974), AB Atomenergi, Studsvik

/ 7/ Mattes, M., Emendörfer D., Keinert, J., Brestrich, I., Lutz, D., Report IKE C-80 (1974),in German.

/ 8/ Bell, M.J., Report ORNL-4628 (1973)

/ 9/ Fleischmann, R.M., Mevrill, E.T., Heeb, C.M., Quaterly Report BNWL 1876

/10/ Pigford, T.H., see ref. /1/

/11/ Ezure, H., Private Communication (Contributor)

/12/ Koch, K., et al.: Report EUR 4690 d (1971)

/13/ Arima, A., et al.: Report EUR 4638e (1971)

/14/ Bresesti, A.M., et al.: Report EUR 4909 e(1972)

/15/ Nodvik, R.J., see ref. /5/

/16/ Sweet, D., Private communication (1975)

/17/ Scholtyssek, Report KFK 1275/3 and Private communication (1975)

/18/ Vaminbraukx, R., Report CBNM/RN/1/74 (1974)

Review Paper No A4

THE REQUIREMENTS FOR TRANSACTINIUM NUCLEAR DATA FOR THE DESIGN AND OPERATION OF NUCLEAR POWER PLANTS

R.M. Nunn (CEGB, BNL)

Abstract

This review is concerned with the importance of transactinium nuclear data to reactor designers and operators, solely in the context of actinides as emitters of radiation. Their importance in the physics design of reactor cores and in criticality aspects of fuel storage and handling is covered in review A2. Here we are concerned with their importance in shielding, access, maintenance, heat removal, instrumentation and fuel handling and storage.

1. Introduction

Commercial nuclear power plants in operation employ a once through uranium cycle. For low burnup reactors (e.g. Magnox) the actinides of importance are the familiar isotopes of plutonium (Pu239, Pu240 , Pu241), the decay product Am241, and the fertile capture products U239 and Np239. For higher burnup reactors (e.g. LWR, AGR) a wider range of actinides -including U237, Np237, Pu236, Pu238, Cm242 and Cm244 - are of significance.

Trial quanitities of mixed oxide fuel (PuO_2/UO_2) are already in use in anticipation of extensive recycling of fuel during the 1980's, and two mixed oxide fast reactor prototypes (Phenix and PFR) are in operation in the presumption that this reactor system will be of commercial significance before 1990. Detailed design work for the first full scale plant is at an advanced stage in both U.K. and France. For mixed oxide fuels a wider range of actinides are produced in significant quantity but only actinides noted above are likely to be of importance except in very extreme hypothetical accident situations.

Thorium fuels are not yet of commercial significance, but two major reactor prototypes (Fort St. Vrain and THTR) have been committed, and there is considerable interest in utilizing thorium fuels in adaptations of existing reactor types (e.g. LWR breeder and CANDU). It is possible that Th/U235 fuels will be of commercial significance in the 1980's and that full recycle Th/U233 fuels will be in service by 1990. For this fuel the range of actinides of interest is small (U232 and its decay products, Th233 and Pa233).

2. Importance of actinides

2.i. Fresh fuel storage and handling

For uranium fuels the radiological problems are minimal and fuel is handled and inspected without significant restriction or operator exposure. For mixed oxide fuel doserates will be small

but may be significant-dependant upon the isotopic content of the plutonium (i.e. prior history). The doserate at 1 metre from an LWR or LMFBR fuel assembly would be ~1mR/hr (neutron plus γ), and although this is unlikely to present major problems, it would require revision of handling and inspection procedures and, possibly, additional fuel store shielding. The important actinides are Pu238, Pu240, Pu241 and Pu242, and the required accuracy in prediction of source strength is ± 50 % for the design of the fresh fuel route.

Repeated recycling of U and U/Pu fuels may result in the build up of significant quantities of Pu236 and U232 (plus daughter products, including the high energy γ-emitter Tl 208) in fresh fuel. The quantity will be very dependant in the prior history of the fuel, but in some circumstances could give rise to a significant doserate from fresh fuel (>1 mR/hr at 1 metre), and there is a speculative requirement to predict the source strength from these isotopes to ± 50 %.

Recycle Th/U233 fuels will present greater problems. The doserate at 1m from a fuel assembly may exceed 1R/hr and shielded handling, inspection and storage facilities would be a necessity. The dominant actinide is Tl 208 (decay product of U232) and the major uncertainty is not in the TND, but in determining the fuel cycle logistics for which the plant is to be designed (i.e. prior history of fuel including shelf life after reprocessing). An uncertainty in the prediction of the U232 content after reprocessing of 30 % would be of significance in the design of the fresh fuel route.

2.ii. Reactor Problems

2.ii.i. Radiological

Under normal conditions actinides make no significant contribution to problems of access, maintenance or operational releases of radioactive material - the problems are caused by activated corrosion products, component activation and fission products. Even in the event of a major release of fuel material into the reactor circuit it is unlikely that the actinides would cause problems directly - but the fission products arising from continued fissioning of the exposed fuel could present problems.

2.ii.ii. Decay heat removal

Decay heat rejection systems are designed to protect against extreme fault situations, and have ample margins to cover normal reactor trip situations; decay heat production over the first few hours (10^4 secs) post shut down determines the system requirements. The accuracy requirement for total heat production during this period is ± 10 %. The contribution of actinides to total heating depends upon the fuel, but could amount to ~ 20 % of the total. Fig. 1 (Ref.1) shows the contribution of actinides to the total heating (excluding neutron fission heating) for three typical fuels. The only actinides of significance at 10^4 secs are U239 and Np239 (U and U/Pu fuels) and Th233 and Pa233 (Th fuels). These are the capture products of the major fertile isotopes (U238 and Th232) and in view of their importance to the physics design of reactor cores, the inventory of these isotopes will be known with a greater precision (± 5 % or better) than will be required for decay heat removal design (± 50 %).

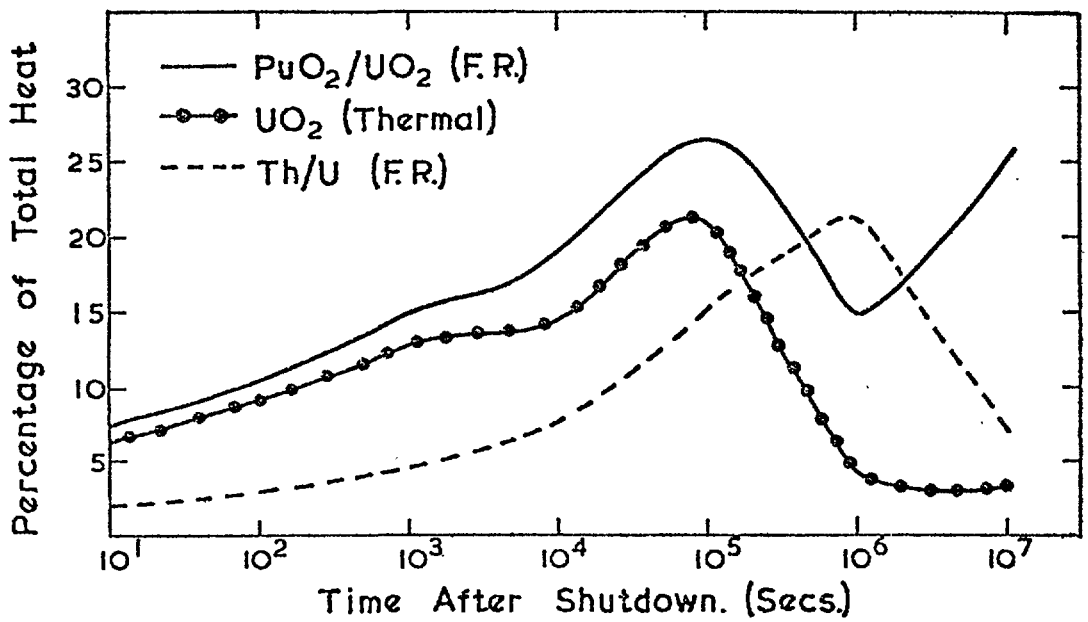


Fig. 1. Contribution of actinides to total heat generation. The peak at $\sim 10^5$ secs is due to the decay of Np239 or Pa233. The contribution from neutron fission is significant during the first 50 secs but has been ignored.

2.ii.iii. Shutdown flux instrumentation

Requirements for flux indication at shutdown can present problems in the design of the reactor and its instrumentation, including requirements for in-reactor artificial neutron sources. For some designs of fast reactor access for instrumentation is particularly difficult, and system design to ensure adequate flux indication requires an accuracy in the predicition of spontaneous neutron production of $\pm$ 50 %. The actinides of importance are Pu240 and Pu242 (clean core) and Cm242, Cm244 (irradiated core).

2.ii.iv. Shutdown reactivity measurements

For reactors refuelled off-load there is a potential requirement to monitor reactivity during fuel and absorber movement, to prevent the inadvertent attainment of criticality. The most usual technique is via sub-critical neutron counting, relating reactivity to flux levels via an assumed knowledge of the effective neutron source strength. In fast reactors, this may well prove to be the most practicable form of routine monitoring, but in application will require that corrections are applied for changes in effective source strength. Fig. 2 (Ref. 2) illustrates the change in source strength of fast reactor fuel during its dwell in the reactor. For certain fuel management situations it is possible that the total reactor source strength could change by an order of magnitude during the course of refuelling. The overall system requirements are that the spontaneous neutron emission of an irradiated subassembly be predictable to an accuracy of $\pm$ 50 %.

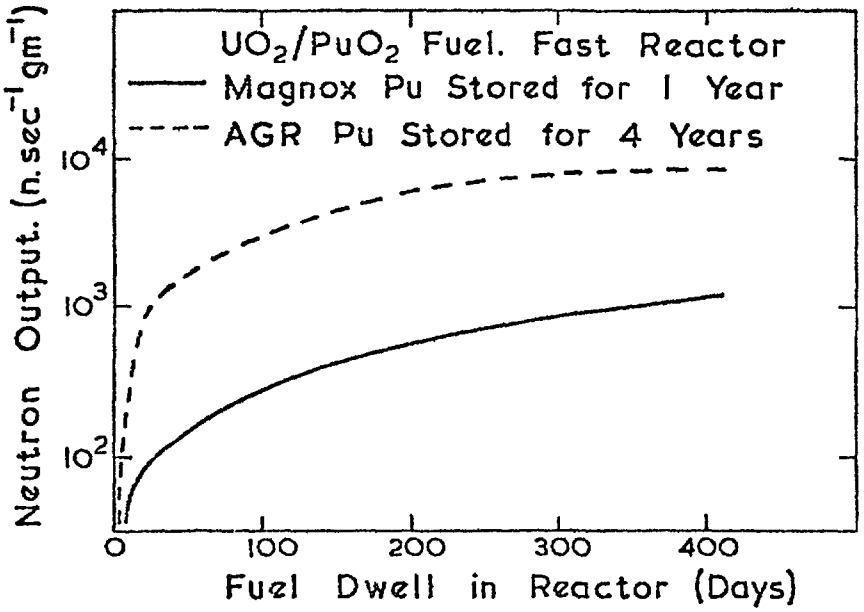


Fig. 2. Change of neutron output for fast reactor fuel. This change results from the build up of Cm242, and, to a lesser extent from Cm244.

2.iii. Irradiated fuel

2.iii.i. The fuel discharge route

An important design aspect is the provision of adequate cooling during fuel transfer from the reactor to spent fuel storage ponds or stores. Design requirements vary from reactor to reactor but are, typically, $\pm$ 10 % in total heat generation over the first few days relaxing to $\sim \pm$ 30 % for longer decay times. For on-load fuelled reactors (e.g. AGR), the $\pm$ 10 % requirement covers the period of zero cooling time up to about 1 week, whilst for off-load fuelled reactors the critical period is from a few hours (SGHWR) up to a few days. Because of the high power density of fast reactor fuels, the $\pm$ 10 % requirement may apply from a decay time of $\sim$1 day up to the time the fuel is transported to the reprocessing facility ($\sim$100 days).

Fig. 3 (Ref. 1) illustrates the relative importance of actinides as a heat source. At decay times in excess of 1 day, α heating (Cm242 and Cm244) makes a significant contribution and after $\sim$7 days this becomes the dominant source of actinide heat production.

Concrete and water are the usual radiation shielding materials for irradiated fuel discharge routes, and shield design is therefore mainly determined by consideration of high energy γ's emanating from fission products. On occasion, space and weight dictate the use of other materials (e.g. steel and lead), and neutron emission may then be of consequence. However, in particular instances known to the author (e.g. fuel discharge machinery for on-load fuelled reactors) it is delayed fission neutrons rather than spontaneous fission neutrons which dominate shielding requirements.

Damage to paints and cables in I.F.D. routes is almost entirely due to high energy γ radiation from fission products and the additional contribution from actinides is insignificant.

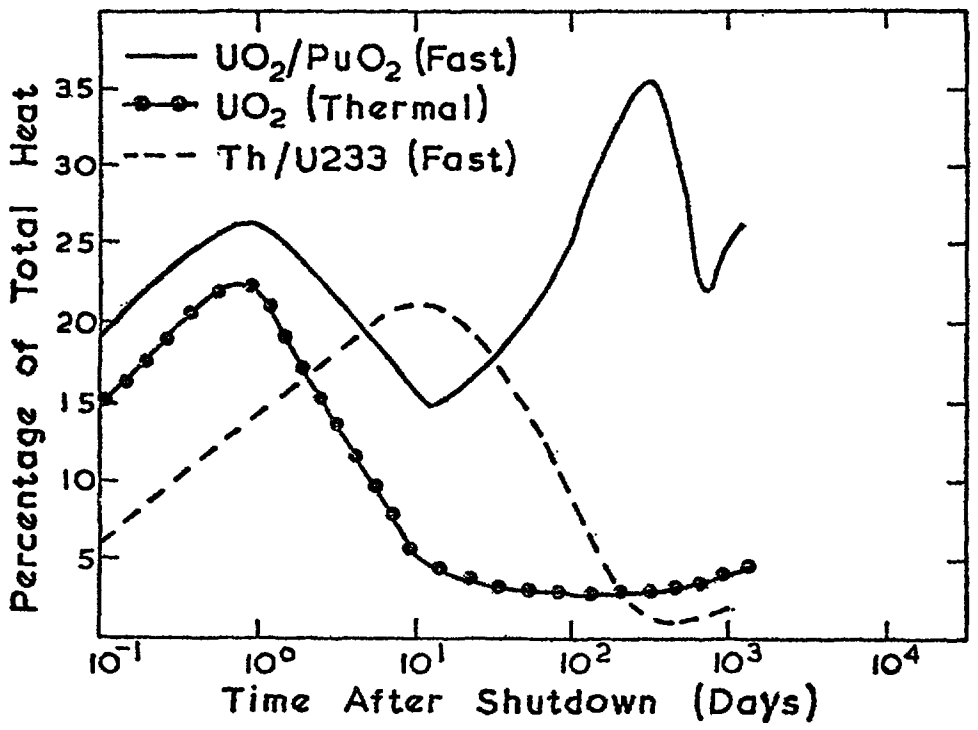


Fig. 3. Contribution of actinides to total shutdown heating. After ~7 days the dominant isotopes are Cm242 and Cm244 (U/Pu fuels).

2.iii.ii. Irradiated fuel storage

For pond storage of thermal reactor fuels decay heat load is not a crucial design issue, and there is usually no requirement for forced cooling. Improved T.N.D. is unlikely to be needed either for this or other aspects of pond management. On site storage arrangements for irradiated fast reactor fuel requires careful design attention to decay heat removal and at this stage it should be assumed that accuracies in total heat output in the range 10–20 % for one or two hundred days cooling may be required. At 200 days up to 40 % of the total heat generation may arise from actinides (Cm242 and Cm244 principally) implying a requirement to determine the inventory of these isotopes to ± 30 %.

3. Summary of requirements. Table 1

Parameter	Fuel/reactor	Transactinide Accuracy	Notes.
γ and neutron output.	Unirradiated) Mixed oxide fuels) Thermal and fast)	± 50 %	For design of fresh fuel route. Pu238, Pu240, Pu241, Pu242. Pu236 and U232.
	Th/U233 fuels	± 30 %	U232 and daughters.
Shutdown Heat output	Whole reactor Mixed oxide (Thermal and fast) Uranium (Thermal) Thorium	± 50 %	For decay heat removal system design-first few hours after shutdown only. U239, Np239, Th233, Pa233.
Shutdown Neutron output.	Irradiated fuel. Mixed oxide, (Fast reactor).	± 50 %	For design of shutdown flux instrumentation and for shutdown reactivity determination. Cm242 and Cm244.
Heat output	Discharged fuel. Uranium (Thermal)	± 50 %	For design of fuel route. For first few days, relaxing for most reactors after ∿1 week. U239, Np239, Th233, Pa233, Cm242, Cm244.
	Mixed oxide (fast)	± 30 %	

4. Requirements for T.N.D.

Table 2 summarises the accuracy requirements (1σ) for the most important transactinium data. Where a multiplicity of routes to the formation of an important actinide exist, some judgement has been exercised in selecting the dominant routes. For example, for the formation of U232, two routes starting at Th232 and U233 are of roughly equal importance, whilst for the formation of Pu238, only the route from Pu239 is likely to be significant.

Table 2. Accuracy requirements. (1σ, thermal and fast reactor integral data).

Area	Data	Th 232	Pa 231	U 233	U 236	U 238	Np 237	Pu 239	Pu 242	Am 241	Am 243	Notes
γ and n output.	σ(n,γ)		20		50				100	50*	100	Decay schemes for U232, Cm242 & Cm244 required.
	σ(n,2n)	20		30		50	50	100				
Heat output.	σ(n,γ) σ(n,2n)	50				30			100	30*	100	Decay schemes for Th233, and U239 required.

* For formation of Am242.

Notes:

1. Assuming inventory of Pu239, Pu240, Pu241 and Pu242 is known to $\pm$ 20 % from core design calculations.
2. Assuming that U232 production is 40 % from U233, 60 % from Th232.
3. Data requirements for U236 and Np237 are speculative. See 2.i. above.
4. Assuming ratio of neutron output Cm242:Cm244 is 4:1.
5. Assuming ratio of heat output Cm242:Cm244 is 10:1.

Acknowledgement:

This review is published by permission of the Central Electricity Generating Board. The contribution of Dr. A.R. Baker from UKAEA, Risley is acknowledged.

References:

(1) Tobias. Annals of Nuclear Energy. Vol. 2, pp. 3-10. 1975. Also private communication from the author.

(2) Clarke, MacDonald, Fitzpatrick and Goddard. Annals of Nuclear Energy, Vol. 2, pp. 451-466. Also private communication from Clarke and MacDonald.

(3) Contribution from A.R. Baker, UKAEA, Risley. This is not specifically referenced in the text, but forms part of the description of fast reactor data requirements.

Review Paper No A5

IMPORTANCE OF TRANS-ACTINIUM NUCLEAR DATA FOR FUEL HANDLING

R F Burstall
Central Technical Services
United Kingdom Atomic Energy Authority
Risley, Nr Warrington
Cheshire

ABSTRACT

The paper considers the importance of the actinide group for fuel handling, particularly in comparison with the fission product group. Decay heating and dose rate are considered for thermal and fast reactors. The effects of variants in fuel usage and reactor operation are examined, and it is shown that the storage time of plutonium feed fuel and the origin of this fuel are important features.

The implications of the actinide group are examined for a possible FBR transport flask, and it is shown that uncertainties in the actinide group contribution are not likely to greatly affect the design. The implications are also examined for fuel fabrication and reprocessing.

Suggestions to re-cycle members of the actinide group in reactors are shown to significantly increase the fuel handling problems.

Recommendations are given for desirable improvements in nuclear data.

1. INTRODUCTION

Before irradiation in a reactor, fuel is fabricated into fuel elements and is transported to the reactor. At the end of the irradiation of a fuel element, the element is unloaded from the reactor and stored on-site for a period to allow reduction in both heating and radioactivity. The element is then loaded into a transport flask, and is taken to a plant for reprocessing and refabrication. These are the processes which are considered under the heading of fuel handling.

At the end of irradiation, there are 3 main groups of sources of heating and activity:

1. The fission products group; the importance of this group for fuel handling was considered by Merz and Laser (Ref 1).

2. The transactinium group.

3. The structural material.

The emissions which produce the heating and activity are:

1. Gamma-rays, which are almost entirely produced by the decay of fission products and, to a lesser extent by the isotopes of the structural material.

2. Beta-rays, which are almost entirely produced by the decay of the fission products.

3. Alpha-rays, which are entirely produced by the decay of the actinides.

4. Neutrons, which are almost entirely produced by the spontaneous fission of the actinides, and by (alpha, n) reactions.

The importance of the actinide group, and the accuracy requirements for the associated data, depends, to a large extent, on the fuel handling methods employed. For current UK methods as described by Boyle et al (Ref 2), the fission product group, the actinide group, and the structural materials group are all closely associated up to the reprocessing stage. At this point the structural materials are removed in the decanning process, the 'useful' members of the actinide group, generally uranium and plutonium, are removed as far as possible for further use, and the remaining actinides and the fission product group are stored together as 'high-level' waste. In this case the importance of the actinide group is mainly determined by the relative magnitudes of the fission product group effects and of the actinide group effects. Thus if the actinide group effect is much smaller than that of the fission product group, good accuracy for the actinide group effect is not necessary. It is shown in Sections 2 and 3 that the importance of the actinide group can change significantly depending on the composition and the storage time of the feed fuel, and, to a lesser extent, on the operation characteristics of the reactor.

Possible future changes in the methods of fuel handling can affect the importance of the actinide group. A severe disadvantage with the present methods of waste management is that the activity produced by the actinide waste extends over a much longer time than does that produced by the fission product group. For example, Haug (Ref 3) has shown that for a uranium-fuelled LWR, fission product activity of wastes predominates up to 300 years after the end of irradiation, but at later times the reverse is true. In order to reduce the storage time of the wastes, there is therefore an incentive to separate the actinide waste from the fission product waste. It has been suggested, eg by Claiborne (Ref 4), that the actinide waste could be recycled in a reactor to reduce the long-term activity. The actinide waste could be recycled with the plutonium or fabricated into special elements. There is also an incentive in separation in that interest has been expressed (see, for example, Ref 5) in the possibility of using curium - 244 as a heat source. If these methods were to be adopted, then the requirements for the accuracy of the actinide group would become more stringent, partly because larger quantities of the members of the group would be present, and partly because the fission product group would no longer be dominant.

The time for which the spent fuel is stored at the reactor site varies for different reactors. For the uranium-fuelled thermal reactors, the intermediate storage time is taken to be about one half year. For the plutonium-fuelled fast reactor, however, there is an economic incentive to reduce the time, since the spent fuel is of considerable value; times as low as 30 days have been quoted (Ref 3). The period of fuel handling is unlikely to extend past 3 years after the end of irradiation, and the review of this paper therefore is only taken over this time.

In Section 2 of this paper, the importance of the actinide group for both decay heating and activity is compared for various reactor systems. In Section 3, the effect of variants is considered, particularly for the case of a plutonium-uranium fuelled FBR. In Section 4, the actinide group is considered in relation to the problems of the spent fuel transport and reprocessing and of fuel fabrication. In Section 5, the preceding sections are reviewed, and recommendations are given for additional work.

2. ACTINIDE IMPORTANCE FOR DIFFERENT REACTOR SYSTEMS

2.1 Decay heating

Decay heating has been calculated for the various reactor systems, and a representative selection of the results is given in Table I. For the uranium-plutonium fuel systems, the actinide power immediately following the end of irradiation is almost entirely due to the beta decays of uranium - 239 and neptunium - 239. As these isotopes have relatively short half-lives, their contribution is negligible after about 10 days. The contribution of the actinide group to the decay heating does not exceed 10% in this early period.

The HTR system, fuelled with thorium as breeder material, shows a different type of actinide contribution. The contribution to decay heat at 30 days is considerably higher than for the other systems, due to the protoactinium - 233 content, which decays with a half-life of 27 days. Since this half-life is relatively low, the actinide contribution to decay heat drops after 30 days, and, after about six months decay, the proportional contribution is no larger than for the other systems.

The results of Table I are not completely comparable, since they have been produced on the basis of different assumptions and with different computer codes. The assumptions and differences are briefly described below:-

1. LWR of 1000MWE (Ref 3)

The enriched uranium design was specified to fuel burn-up of 34000 MWD/Te at a specific power of 29.5 MW/Te. The plutonium fuelled design was specified to be fuelled with the plutonium discharged from a uranium-fuelled LWR; the burn-up was 34000 MWD/Te and the plutonium content was 19%. For both designs the fuel was assumed to be re-processed at 150 days after the end of irradiation. At reprocessing, 1% of the uranium and plutonium is assumed to be lost to the high level waste. The ORIGEN code (Ref 6) was used for the calculations.

2. HTGR of 1160 MWE (fuelled with thorium and uranium - 233 when available). (Ref 7)

The mean fuel burn-up was 94000 MWD/Te. The fuel was assumed to be reprocessed one year after the end of the irradiation, and 0.5% of both uranium and thorium were assumed to be lost to the high-level waste. The calculations were performed by the ORIGEN code (Ref 6), augmented by the RADEC code (Ref 8).

3. FBR of 1000 MWE (fuelled with plutonium from an LWR of Pu239/240/241/242 content of .60/.24/.12/.04)(Ref 9)

Fuel burn-up of 80,000 MWD/Te is assumed for the core region, with a feed plutonium enrichment of .156; the plutonium is assumed to be used without storage time. The calculations are based on core and blanket material mixed in the same proportions as they are discharged from the reactor. Reprocessing is assumed to begin 30 days after discharge, and 0.5% of both uranium and plutonium is assumed to be lost to the high-level waste. The ORIGEN code (Ref 6) was used for the calculations.

4. FBR of 1000 MWE (fuelled with plutonium from an AGR of Pu239/240/241/242 content of .495/.335/.110/.060) (Ref 10 and more recent unpublished work)

The fuel burn-up was 10% equivalent to about 90,000 MWD/Te for the core region with a feed plutonium enrichment of .266. The plutonium is

assumed to be used without storage time. The breeder contributions are not included in the calculations. No reprocessing time is specified, so the americium - 241 content will continue to build up. The FISPIN code (Ref 11) was used for the calculations.

TABLE I

PERCENTAGE OF DECAY HEAT PRODUCED BY ACTINIDES

Time after end of irradiation	30 days	90 days	150 days	1 year	3 years
LWR uranium enrichment (Ref 3)	3.0	4.1	4.5	3.1	3.3
plutonium re-cycle (Ref 3)	7.8	10.7	12.0	12.4	
HTGR (Ref 7)	22.5	11.1		3.5	
FBR plutonium feed from LWR (Ref 9) core and breeder mixed	3.8			4.8	3.7
plutonium feed from AGR (Ref 10) core fuel only	4.5		7.9	9.3	

While the differences between the models for calculation and the different methods of calculation should be borne in mind, the Table I shows that, with the exception of the HTGR, the contribution of the actinides to the decay heating in the first year after discharge from reactor are unlikely to greatly exceed 10% of the total.

For the uranium-plutonium fuel systems, at 30 days after the end of irradiation, the curium - 242 is the predominant nuclide, producing about 80% of the heat from the actinide group. The remaining 20% is contributed by curium - 244, plutonium - 238, and small amounts from other nuclides. At one year after the end of irradiation, the curium - 242 is still predominant for the fast reactor systems. For the LWR systems at this time the curium - 244 contributes a comparable proportion to the curium - 242, and for the plutonium-enriched LWR, the curium - 244 contributes 65% of the total actinide heating.

Clarke et al (Ref 12) have examined a variety of reactors using the codes FISP (Ref 13) and HYLAS (Ref 14), and it may be seen from their work that the actinide heating effects for AGR and SGHWR systems are of the same order as those of the uranium-enriched LWR of Table I. For the Magnox reactors with natural uranium as feed, Clarke et al show that the relatively low fuel burn-up means that the actinide contribution to decay heating in the first year after irradiation is only about 1% of the total heat. It is also shown in Ref 10 that use of the uranium - 233/thorium oxide fuel cycle in an FBR results in actinide heating lower by large factors compared with the plutonium-uranium fuelled FBR, at times greater than 100 days after the end of irradiation.

For the uranium-plutonium fuelled systems the low contribution of the actinide group to the total decay heating in the first year after discharge from the reactor makes it therefore unlikely that errors in actinide data are sufficient to appreciably affect the total heating for the period of fuel handling, though at later times the actinide heating effect becomes more significant relatively.

As far as the HTGR is concerned, the heating in the first two months after discharge is significant, and the cross-sections are less well known for the thorium cycle compared with the plutonium cycle. However this higher actinide contribution only occurs during the time in which spent fuel is in site storage.

2.2 Neutron emission

Table II gives the neutrons produced in spent fuel for the reactors already listed in Table I. The total neutrons are included, those from spontaneous fission, and those produced in (alpha, n) reactions in oxygen and carbon. The latter contribute about 10-20% of the total. The data for the first listed fast reactor system are calculated from the activities quoted in Ref 9.

For the LWR, use of plutonium enrichment (with feed fuel direct from LWR reprocessed fuel) leads to an increase by a factor of about six in neutron emission over the uranium enrichment case. For both cases, the curium - 244 is the predominant nuclide for neutron production. The build-up of curium - 244 is more important relatively for the neutron production than for the heat production, since this nuclide produces about 20 times as many spontaneous fission neutrons per decay as from curium - 242. For the uranium-fuelled LWR, the curium - 244 spontaneous fission contribution rises from 57% of total neutron production at 30 days after the end of irradiation to 81% at 1 year. For the plutonium-fuelled LWR, the corresponding rise is from 79% to 91%. The remaining percentages are almost entirely made up from contributions from curium - 242 spontaneous fission, and from the (alpha, n) reactions.

TABLE II

NEUTRONS PRODUCED BY ACTINIDES (/TeHM/sec)

Time after end of irradiation	30 days	90 days	150 days	1 year	3 years
LWR uranium enrichment (Ref 3)	7.0,8	6.4,8	5.8,8	4.3,8	3.6,8
plutonium enrichment (Ref 3)	4.0,9	3.9,9	3.7,9	3.2,9	2.9,9
FBR plutonium feed from LWR (Ref 9)No(alpha,n) (core and breeder mixed)	6.0,8			2.6,8	1.6,8
Plutonium feed from AGR (Ref 10) (core fuel only)	5.8,9		4.7,9	3.8,9	

Comparing the plutonium-enriched LWR data and the FBR data listed in Table II, it would appear that the range of neutron production per tonne of fuel charged to the reactor is similar for the two reactor systems. The plutonium enrichment of the FBR systems is much higher, but the probability of production of actinides is lower in a fast reactor spectrum, and these effects tend to cancel out.

It will be seen from Table II that the two sets of results for the two fast reactor systems differ by a factor of about ten. This difference is due to the following factors:-

1. The US reactor system is based on a mixture of core and breeder fuel, while the UK system relates to core fuel at peak rating and burn-up. The former assumption is probably more useful for intermediate storage and reprocessing plant calculations. The second assumption is more useful for transport flask calculations, since in this case the design is fixed by the peak behaviour at the fuel centre. The effect of mixing is to reduce the burn-up from the core fuel value of 80,000 MWD/Te to a mean value of 33,000 MWD/Te. Bell (Ref 15) has given data for actinide build-up in the core and axial and radial breeder regions separately. It is shown that the radial breeder, after a dwell time of about 2.5 years, produces less curium - 242 than produced in the core by a factor of 10^{-4}, and even less proportion of curium - 244. The axial breeder contribution is still lower. The activity and heat generation of actinides in the breeder region fuel is thus negligible in comparison with the core data.

   It is estimated that the effect of mixing the reject fuel is to reduce the neutron emission by a factor of about three. In the data of Ref 9 the effect of mixing is increased by use of a uranium-enriched radial breeder, which reduces the dwell time of fuel in this region.

2. The plutonium enrichment of the core fuel in the US case was 0.156, while that of the UK case was 0.266. The rating of the US case was 148 MW/Te, while that of the UK case was 250 MW/Te, which is a peak value. The effect of enrichment and rating changes is considered in section 3. The net effect of the differences in enrichment and rating is to make a neutron emission reduction in the US case of about a factor of two compared with the UK case.

3. As mentioned in section 2.1, the percentage of plutonium - 242 present in the feed to the US case was 4% (from an LWR), while that in the UK case was 6% (from an AGR). For the FBR curium - 244 contributes 57% of the total neutron emission at 30 days after the end of irradiation, and 85% at 1 year. The build-up of curium - 244 is effectively proportional to the plutonium - 242 content, since the half-lives involved are relatively short. The effect of the increased content of plutonium - 242 in spent AGR fuel is therefore to increase the neutron emission from curium - 244 by some 50%, and the total neutron emission by about 30% at 30 days after the end of irradiation.

4. The US values do not include neutrons from (alpha, n) reactions, inclusion of which would increase the dose rate by about 10%.

5. Differences in the data sets of computer codes (see section 2.1) account for any remaining differences.

The comparison of the two quoted sets of results for the FBR systems show that different assumptions on fuel handling and operation can lead to significant changes in the neutron emissions to be expected from spent fuel.

Neutron emissions for the SGHWR spent fuel are quoted in Ref 12. The values are about 60% of the uranium-enriched LWR values of Table II. The Ref 12 also reports that neutron emissions from thorium-uranium - 233 fuel cycles are lower by factors of about six than from the plutonium-uranium fuel cycles, for both FBR and HTR.

2.3 Gamma radiation

The actinide group produce gamma radiation from three processes, first, from decay of members of the group, second, from spontaneous fission of members of the group, and third, from decay of fission products resulting from this spontaneous fission.

Haug (Ref 3) gives data on photon emission from the actinide group, the fission product group, and the cladding and structural materials. This data relates to a uranium-fuelled LWR. Thirty days after the end of irradiation, the total photon energy release by the three processes listed above from the actinide group is about 0.1% of the total. After one year, the actinide group contributes a still smaller fraction of the total gamma ray heating. While other fuels or reactor systems may produce spent fuel with higher activity of the actinide group, it seems unlikely that gamma ray heating of this group could be significant while the group is associated with the fission product group (see Section 2.1).

The dose rates of gamma rays from the actinide group are also much lower than from the fission product group, though Haug indicates that the former produces more photons at higher energies. A further source of gamma rays results from capture of the neutrons emitted from the actinide group. The importance of these gamma rays depends on the location of the neutron capture, and it is shown in Section 4.1 that these gamma rays can be significant.

3. VARIANTS OF IMPORTANCE TO THE ACTINIDE GROUP

3.1 Effect of plutonium storage

For fast reactor systems, plutonium used for enrichment will, at least for the initial fast reactors, have been stored for a number of years, during which the plutonium - 241 content decays to americium - 241 with a half-life of about 14 years. The subsequent irradiation in a reactor will lead to greater production of curium - 242 than for plutonium with no storage time. The effect of storage time on decay heating for an FBR is given in Table III, and on neutron production in Table IV. Data is given for an FBR fuelled with plutonium discharged from an AGR. Data is from Ref 10 and from more recent unpublished work.

Tables III and IV show that storage time is a significant feature of actinide importance. In the decay heating over the first year after irradiation, storage time increases the actinide contribution appreciably. If the plutonium is stored for 10 years, then the actinides can contribute up to 50% of the decay heating. For this storage time, the neutron emission is increased by a factor of about 5 over the case with no storage time.

3.2 Effect of plutonium composition

The plutonium used as feed to FBR systems will vary in composition, depending on the source of the plutonium. In the UK, plutonium in the early FBR's will be provided by Magnox reactors and by AGR's. The spent fuel plutonium composition of the Magnox plutonium is .76/.195/.0395/.0055 for Pu239/240/241/242, while that of the AGR's is given in Section 2.1. The effect of changes in plutonium composition are given in Tables III and IV. Data is also given for Magnox plutonium which has been re-cycled many times in an FBR,

TABLE III

EFFECT OF STORAGE TIME, PLUTONIUM COMPOSITION, BURN-UP, RATING AND ENRICHMENT ON FBR ACTINIDE DECAY HEAT PROPORTION

Reactor producing plutonium	Storage time of plutonium before use (years)	Burn-up of fuel (%)	Power rating of fuel (MW/Te)	Enrichment Pu/Pu+U	Decay heat proportion (%) at time after irradiation of		
					30 days	150 days	1 year
AGR	0	10	250	.266	4.5	7.9	9.3
AGR	4	10	250	.266	21.7	32.1	31.8
AGR	10	10	250	.266	35.5	48.3	47.4
AGR	4	10	200	.266	23.1	32.9	32.0
AGR	4	8	250	.266	22.0	33.6	34.1
AGR	4	10	250	.216	19.1	28.9	28.8
Magnox	4	10	250	.227	8.0	13.0	13.1
Magnox plutonium re-cycled many times through FBR	4	10	250	.300	12.6	19.9	20.1

TABLE IV

EFFECT OF STORAGE TIME, PLUTONIUM COMPOSITION, BURN-UP, RATING AND ENRICHMENT ON FBR ACTINIDE NEUTRON PRODUCTION (/TeHM/sec)

Reactor producing plutonium	Storage time of plutonium before use (years)	Burn-up of fuel (%)	Power rating of fuel (MW/Te)	Enrichment Pu/Pu+U	Neutron production (/TeHM/sec) at time after irradiation of		
					30 days	150 days	1 year
AGR	0	10	250	.266	5.8,9	4.7,9	3.8,9
AGR	4	10	250	.266	1.9,10	1.25,10	7.0,9
AGR	10	10	250	.266	3.4,10	2.2,10	1.1,10
AGR	4	10	200	.266	1.7,10	1.2,10	6.7,9
AGR	4	8	250	.266	1.7,10	1.1,10	5.8,9
AGR	4	10	250	.216	1.7,10	1.1,10	6.6,9
Magnox	4	10	250	.227	5.2,9	3.2,9	1.5,9
Magnox plutonium re-cycled many times in FBR	4	10	250	.300	9.2,9	6.0,9	3.1,9

and has a feed plutonium composition of .53/.40/.05/.02. The data does include the changes in plutonium enrichment required to achieve the 10% fuel burn-up target. A storage time of four years is assumed for all the cases. The results show that for the 'clean' plutonium from the Magnox reactors, the actinide contribution to both decay heating and neutron emission is appreciably reduced. The FBR re-cycled plutonium also shows a lower actinide contribution relative to the AGR plutonium.

3.3 Effect of fuel burn-up, rating, and enrichment

Table III gives the effect on decay heat of reducing the target burn-up from 10% to 8%, of reducing fuel rating from 250 MW/Te to 200 MW/Te, and of reducing fuel enrichment from .266 to .216. Table IV gives the effects on neutron emission. The effects may be seen to be relatively minor, compared to the effects of changes in storage time, or composition.

3.4 Effect of actinide re-cycle

Claiborne (Ref 4) has considered the effects of actinide re-cycle for a PWR system, and Hirst (Ref 16) has performed similar calculations for a small FBR. Both show a ten-fold increase of neutron emission of the actinide group after repeated re-cycling of americium and curium, and the fuel handling effects are correspondingly increased. Other papers to this meeting consider this topic in more detail (Ref 17, 18).

3.5 Effect of nuclear data errors

For the plutonium-uranium systems, the heating and activity of the actinide group are dominated during the period of fuel handling by the two isotopes curium - 242 and curium - 244.

The major uncertainty in the production of curium - 242 is in the capture cross-section of americium - 241. The uncertainties in this cross-section and others are considered in other papers presented to this meeting. The effect on heating and activity of errors in the cross-section data have been considered by Richardson (Ref 10) and in more recent work. For the FBR specified in row 2 of Table III, ie with feed plutonium from an AGR with four years storage time, results are given in Table V for times after irradiation of 30 days, 150 days and one year.

TABLE V

PERCENTAGE CHANGES IN DECAY HEATING AND NEUTRON EMISSION DUE TO CROSS-SECTION UNCERTAINTIES

Time after end of irradiation	30 days	150 days	1 year
10% increase in Am-241 capture			
Increase in total decay heat	+1.3	+2.2	+2.0
Increase in neutron emission	+5	+5	+3
30% increase in Pu^{242} capture			
Increase in total decay heat	-	+0.3	+0.6
Increase in neutron emission	+6	+9	+14
30% increase in Am^{243} capture			
Increase in total decay heat	-	+0.2	+0.4
Increase in neutron emission	+5	+8	+13

The major uncertainties in the production of curium - 244 are thought to be in the capture cross-sections of plutonium - 242 and of americium - 243. Table V gives the effect of uncertainties in these cross-sections.

It may be seen from Table V that the given uncertainties in neutron capture cross-sections do not produce large effects on decay heating and neutron emissions. The effects are small compared with those produced by changes in plutonium storage time or composition.

For the thorium-uranium systems, Clarke et al (Ref 12) have shown that the heating and neutron emissions involved are less significant (at the important times) than for the plutonium-uranium systems, and the effect of cross-section uncertainties will also be less significant. The thorium-uranium systems do give rise to gamma ray emissions, however, for which additional data is required (see Section 4.2).

4. SHIELDING

Shielding for gamma radiation is best achieved by material of high atomic number. In the case of neutrons such material is useful at high energies (more than about 800 keV for steel), where inelastic scattering is an important effect, but at lower energies material of low atomic number is required to produce rapid loss of neutron energy to a level at which neutron capture is very probable. Thus if there is an appreciable neutron source in the spent fuel, shielding design requires materials of both high and low atomic number. A further complication is that neutron capture generally results in the emission of gamma radiation, which must be included in assessment of dose rates.

4.1 Transport Flasks

In the UK, a design has been prepared for the transport flasks for spent FBR fuel. This design may or may not be ultimately used. Each FBR sub-assembly is to be contained in a sodium-filled can, and several cans may be carried in each flask. The cans fit into cylindrical channels in the steel flask, which has an outer diameter of 1676 mm. The steel is surrounded by a 76 mm thick water jacket, which is contained in 25 mm thick steel extending for 305 mm above and below the fuelled length of the sub-assembly. The flask is fitted with circumferential fins on its outer radial surface. The requirements to transport the flask by rail restrict the size and weight of the flask.

Calculations have been performed by Avery (Ref 19) on the dose rates at the surface of the flask, and at a distance of 2000 mm, for fuel 30 days after the end of irradiation. The spent fuel is assumed to be that specified in the row 2 of Table III, ie plutonium from an AGR with a storage time of 4 years. The calculations indicate that of the total dose rate at the mid-plane of the flask surface, 25% is due to the fission product gamma ray dose rate, 58% is due to the neutron dose rate, and the remaining 17% is due to the gamma ray dose rate from neutron capture. At 2000 mm from the flask, the fission product contribution has dropped to 14%. The peak neutron dose rate at the flask surface is calculated to be at the top and bottom ends of the water jacket, where the neutron dose rate is about double that at the mid-plane.

The total dose rates for a flask carrying 5 spent FBR sub-assemblies are well within the targets specified by the IAEA (Ref 20); these are 200 mrem/h at the surface and 10 mrem/h at 2000 mm. The calculated dose rates are lower than these targets by factors of about 15 at the surface mid-plane and by about 7 at 2000 mm mid-plane. It is possible that the targets would have to be made more stringent for any transport flask which would require decontamination to be carried out after loading. The above calculations make no allowance for neutron multiplication in the fuel of the flask. For a flask holding 5 sub-

assemblies, the multiplication factor could be 1.5. In this case, the dose rate due to the neutrons (both direct and due to gamma rays from neutron capture) will considerably exceed that due to the fission product decays.

Estimates have also been made of the heating of the flask. Since there is no forced cooling in the design, the heat which can be accommodated is restricted to 40 kW. For the spent fuel specified in row 2 of Table III and for a time of 30 days after the end of irradiation, a flask containing 5 sub-assemblies would produce too much heat by a factor of about 2. If the fuel were to be stored for 150 days after irradiation before loading the flask, then the heat would have reduced to an acceptable level. Alternatively, the flask could be loaded by a smaller number of sub-assemblies, at some economic cost.

It may be seen from Table IV that the effect of increasing the period between the end of irradiation and fuel transport is to reduce the neutron emission by about 40%. The economics of varying this period of time have been assessed. The major quantifiable costs to be balanced are the value of the out-of-pile inventory and the expense of reprocessing highly active fuels. Neither of these costs are well known, but it appears that an optimum delay is about 180 days, in contrast to US estimates for a cooled flask.

Features which could lead to increases in the dose rate and heating are:-

1. Increase of plutonium storage time. Table IV shows that an increase from 4 years to 10 years would give an increase of dose rate (from neutron emission) of nearly 100%.

2. Re-cycle of the trans-plutonium isotopes. It is shown in Section 3.4 that continued re-cycle of the americium and curium isotopes could give increases in neutron emission by a factor of about 10. An increase of neutron emission by a factor of 10 would result in dose rates slightly outside the IAEA limits.

On the other hand, features which could reduce the dose rate are:

1. The results of Table IV are based on sub-assemblies from the core region. Inclusion of sub-assemblies from the radial breeder region would reduce the dose rate, but this could not be achieved for all transport flasks.

2. The results of Table IV are based on a peak fuel rating for the reactor. Sub-assemblies of lower rating will be discharged from the reactor and will reduce the dose rate somewhat, as may be seen from Table IV. Also, the axial variation of rating along the length of a sub-assembly means that the dose rate calculation over-estimates the true situation.

3. Table IV shows that use of plutonium produced by Magnox reactors rather than AGR's reduces neutron emission by a factor 3. Early fast reactors in the UK will presumably be fuelled with plutonium from Magnox reactors, and will give useful information on the accuracies of data and calculations before the later plutonium is used. The use of plutonium originating in a Magnox reactor and re-cycled in an FBR will also give reduced neutron emission, as will plutonium from the breeder regions of fast reactors.

To assess the effect of changes in predicted neutron emission on the transport flask design, Butler et al (Ref 21) show that for neutron emissions from a point source of curium - 244, an increase in emission by a factor of 5 would require an increase in water thickness from 76 mm to about 190 mm to achieve the same dose rate. An increase in neutron emission by a factor of 10 would require an increase in water thickness to about 240 mm.

Summarising, the decay heating of the UK FBR transport flask design is a more important restriction than is the dose rate. The dose rate is well within the limits set by the IAEA. The dose rate can be dominated by the actinide group, depending on the assumptions on plutonium storage time and composition. The decay heating, however, is dominated by the fission product group for most assumptions, as may be seen from Table III. It would therefore appear that the actinide group is only of limited importance in the transport flask design.

The exception to the above arises if the method of re-cycling actinides is used. In this case, the dose rate is just outside the IAEA limits, and this dose rate is largely due to the actinide group neutron emissions. The decay heating is also made up mainly from the actinide group in this case.

4.2 Fabrication

The feeds to the existing Windscale fabrication plant for plutonium-uranium fuel are uranyl nitrate and plutonyl nitrate, which are blended and co-precipitated to give the oxide powder feed to the pellet line. Dust contamination can provide a radiation hazard to operators, so a 'wet' process is being developed, the vibro gel-precipitation process. The major source of activity of this fuel is from plutonium - 241 decay, though the penetrating radiation, the neutron emission, is chiefly due to decays of plutonium - 242 and plutonium - 240. The neutron dose rate is lower by a factor of about fifty or more compared with the dose rate for irradiated fuel. The heating is lower by a factor of about 700 or more. It is planned that light shielding will be required for handling of fuel in store and reactor loading, and adequate margins may readily be incorporated to allow for uncertainties. Storage of plutonium and consequent build-up of americium - 241 does not create extra problems, though decays of this isotope form the largest contribution to the overall heating for storage times of four years or more.

The actinide group is of limited importance in fuel fabrication for conventional processes, but this is not true if the method of actinide re-cycling is employed. In the case of re-cycling curium and americium with the plutonium, the activity of the fabricated fuel is only slightly lower than that of spent fuel. The heating of the fabricated fuel is also significant. Hence the fabricated fuel would have to be handled with similar shielding and heat removal as that used for spent fuel.

As pointed out by Clarke et al (Ref 12) there is a problem associated with fabrication of thorium-uranium 233 fuel elements. Build-up of the isotope uranium - 232 occurs in the thorium burn-up chain either from an (n, 2n) reaction in uranium - 233, or from beta decay of protoactinium - 232. Uranium - 232 decays with a half-life of 72 years to thorium - 228, which decays with a 36% probability of emitting a 2.65 MeV gamma-ray. The activity of the uranium will depend on the storage time before use, but the dose rate is sufficient to require significant shielding compared with the plutonium-uranium fuel. There is a need for improved data on the (n, 2n) reactions in uranium - 233 and thorium - 232.

4.3 Reprocessing

Aqueous reprocessing is the technique generally used at present for spent fuel from thermal reactor systems. Calleri et al (Ref 22) suggest that similar techniques will be adopted for fast reactor spent fuel, due to the experience available. For thermal reactors, intermediate storage and decanning takes place under water. For fast reactors, the presence of sodium requires storage under sodium for a cooling period. This is followed by cleaning to remove sodium from the sub-assembly. The sub-assembly fuel may then be converted to an aqueous phase, and the waste products remain in such a phase right up to and including the time at which they are stored in tanks. In contrast to the trans-

port flask of Section 4.1, therefore, there is no lack of material of good moderation in intimate contact with the waste products. The neutron emissions are rapidly moderated, and there is adequate parasitic absorption of thermal neutrons. In addition, the various components are surrounded by thick concrete shields, to degrade the fission product gamma activity, and these shields are effective in reducing neutron activity. Thus in the storage and reprocessing areas, the actinide group is not expected to produce important effects.

Allowance is made for the heating in the reprocessing plant, and the relatively small contribution of the actinide group for most cases is not thought likely to be important.

The actinide group has some influence on criticality considerations. Criticality is normally prevented by limiting the fissionable material, by imposing dimensional limitations, and by adding neutron absorbers. The margins of uncertainty assumed in these factors are sufficiently large that the actinide effect on criticality is not of importance.

As with other aspects of fuel handling, the problems of reprocessing are considerably increased if the method of actinide re-cycling is employed.

5. CONCLUSIONS AND RECOMMENDATIONS

1. For uranium-fuelled reactors, and for plutonium-fuelled reactors in which the plutonium has not been stored for appreciable times after reprocessing, the contribution of the actinide group to the decay heating is less than 15%, over the period of fuel handling.

2. For the FBR natural circulation-cooled transport flask considered in this paper, the actinide group contributes up to 50% of the dose rate at the flask surface (mid-plane). For this flask, however, heating is a more important restriction on the design than dose rate. The actinide group dose rate is due to neutron emissions, plus the (n, gamma) reactions which follow. In the reprocessing plant, the actinide group will be in an aqueous phase, and the importance of the neutron emissions will be greatly reduced.

3. The plutonium-fuelled LWR and FBR show similar heating and neutron emission effects. Though the FBR has a higher throughput of plutonium, the harder spectrum of this reactor increases the possibility of fission rather than capture in members of the actinide group.

4. Storage time of plutonium before subsequent use in a reactor is a significant factor in the importance of the actinide group, with a 10 year storage time of AGR plutonium resulting in the actinide group contributing nearly 50% of the decay heating. The actinide contribution to the dose rate for this assumption is about 85% for the transport flask as above.

The important effects of storage time may lead to changes in the reprocessing strategy. Storage of plutonium may be possible as poisoned nitrate liquor with removal of poison, impurities and americium in a solvent extraction unit regarded as a head end of the fabrication line.

5. The composition of the plutonium feed is a factor in the importance of the actinide group, with use of plutonium from a Magnox reactor in an FBR resulting in the actinide group contributing only about one-third as much of the decay heating as for plutonium from an AGR. Plutonium from the breeder regions of an FBR will also result in low actinide contributions. For these plutonium feeds, storage of plutonium is much less important than above.

6. Increases of fuel burn-up, rating, and enrichment all lead to increases of actinide group heating and dose rate, but for the scale of changes to be generally expected, the changes are minor compared with changes in feed plutonium storage time and composition.

7. The actinide contributions to both decay heating and to dose rate are dominated by the two isotopes curium - 242 and curium - 244. Likely errors in the nuclear data leading to the production and destruction of these isotopes cause small uncertainties in fuel handling effects, compared with changes due to plutonium storage time and composition. It seems that the nuclear data is adequate, at least until the latter effects are established. Neutron sources should be accurate to $\pm$ 20%.

8. In the IAEA review of Fission Product Nuclear Data (Ref 23), it was stated that the total heat released by fission products should be known to $\pm$ 5% or better from about three months after the end of irradiation. The heating of the actinide group is generally less than 15% of the total decay heating, and for this group the above uncertainty limits may be relaxed by a factor of about 5. The accuracy requirement increases with storage time, and for plutonium discharged from an AGR or similar thermal reactor, and then stored for 10 years and then used in an FBR, the accuracy of the actinide group heating is needed to similar accuracies as for the fission product group.

9. Assumptions on fuel handling can make significant differences to the actinide group importance. Thus if for an FBR, the core spent fuel is assumed to be associated with the breeder spent fuel, then the actinide group importance is reduced by a factor of about three.

10. The possibility of re-cycling members of the actinide group in a reactor to reduce long-term storage problems could lead to dose rate increases of spent fuel by factors of up to ten, and large increases in decay heating also occur. Improvements in fuel handling uncertainties would be needed, as considered in other papers (Ref 17, 18) to be presented to this meeting. From the fuel handling view-point, it would probably be preferable to re-cycle the actinides separately from the uranium or plutonium fuel.

11. For the thorium-uranium fuel systems, the problems generally are less during the period of fuel handling compared with the plutonium-uranium systems. There are two exceptions, however. First, up to about 100 days after the end of fuel irradiation the decay heating of the former is higher, and, second, and more importantly, a member of the burn-up chain emits gamma rays of sufficient intensity to cause problems in the uranium - 233 fuel fabrication. To obtain suitable accuracy of estimates of the dose rate, improvements in the (n, 2n) cross-sections of uranium - 233 and thorium - 232 are needed.

12. Computer codes which calculate the actinide group effects should be capable of printing data in a convenient form for the user; this data should include weight, curies, curie - MeV, neutron emission and energies, and heat output in watts for each nuclide or isomer.

13. There is some evidence that the data sets used in different countries produce significant differences in estimates of fuel handling effects. The differences between data sets should be examined.

ACKNOWLEDGEMENTS

I would like to express appreciation of the work of the following contributors to this paper:

Dr A R Baker	UKAEA, Risley
Dr W R Burton	UKAEA, Risley
Dr R H Clarke	CEGB, Berkeley
Mr A L Mills	UKAEA, Dounreay

and also of the helpful advice of other colleagues at UKAEA, Risley.

REFERENCES

1. Merz, E and Laser, M, — Importance of Fission Product Nuclear Data for Fuel Handling. Proceedings of Bologna panel on Fission Product Nuclear Data IAEA-169/P7 (1974)

2. Boyle, J G et al — Operating Experience with the United Kingdom Fuel Reprocessing Plants at Windscale and Dounreay. Fourth United Nations International Conference on the Peaceful Uses of Atomic Energy A/CONF49/492 (1971)

3. Haug, H O — Calculation and Compilations of Composition, Radioactivity, Thermal Power, Gamma and Neutron Release Rates of Fission Products and Actinides of Spent Power Reactor Fuels and Their Processing Wastes KFK/ 1945 (1974)

4. Claiborne, H C — Neutron - induced Transmutations of High-level Radioactive Waste ORNL-TM-3964 (1972)

5. Carney, H C — Curium - 244 Recovery from Spent Reactor Fuel Transactions of American Nuc Soc Vol 18, P13, (1974)

6. Bell, M J — ORIGEN - The ORNL Isotope Generation and Depletion Code ORNL - 4628 (1973)

7. Angelo, J A and Post R G — Nuclear Fuel Cycle and the Isotopic Compositions Nuc Tech 24, 323 (1974)

8. Angelo, J A — Heat Transfer from Radioactive Wastes in Deep Rock, Doctoral Dissertation, University of Arizona

9. — Siting of Fuel Reprocessing Plants and Waste Management Facilities ORNL-4451 (1970)

10. Richardson, B L — UKAEA Internal Document (1974)

11. Richardson, B L — UKAEA Internal Document (1975)

12. Clarke, R H, MacDonald H F Fitzpatrick J and Goddard A J H — Waste Disposal Aspects of the long Term Cooling Characteristics of Irradiated Nuclear Fuels
Ann. of Nuc. Energy Vol 2 pp 451

13. Clarke R H — FISP a Comprehensive Computer Programme for Generating Fission Product Inventories
Health Phys 23, 565 (1972)

14. Clarke, R H — HYLAS Code
CEGB Internal Document

15. Bell, M J — Heavy Element Composition of Spent Power Reactor Fuels
ORNL-TM-2897 (1970)

16. Hirst, I L — UKAEA Internal Document (1975)

17. Koch, L — European Programmes in Waste Management (Incineration) of Actinides
These Panel Proceedings - A6

18. Raman, S — US Programmes in the Waste Management (Incineration) of Actinides
These Panel Proceedings - A7

19. Avery, A F — UKAEA Internal Document (1974)

20. — Regulations for the Safe Transport of Radioactive Materials (Revised 1973)
IAEA Safety Standards Safety Series No 6
Vienna 1973

21. Butler, T A, Ketchen, E, and Distefane, J R — Curium Data Sheets
ORNL-4910 (1973)

22. Calleri, G et al — Reprocessing of Fast Breeder Reactor Fuel using Aqueous Reprocessing Technology
EUR-5010e (1973)

23. Lammer, M (Editor) — Summary of IAEA Panel on Fission Product Nuclear Data Bologna 1973
INDC (NDS) - 65/L+N (1975)

Review Paper No. A6

European Programmes in Waste Management (Incineration) of Actinides

L. Koch

European Institute for Transuranium Elements, Karlsruhe

Contributors: D. Breton, CEA, Fontenay aux Roses
R.H. Clarke, CEGB, Berkeley, U.K.
H. Krause, Gesellschaft fuer Kernforschung, Karlsruhe
J.O. Liljenzin, Chalmers Tekniska Hoekskola, Goeteborg
M.G. Sowerby, UKAEA Harwell

Abstract

The composition of actinides in power reactor fuel is presented, and the partioning of actinide wastes during fuel fabrication and reprocessing is considered. Concepts for actinide waste management, with an emphasis on nuclear incineration, and the associated requirements for transactinium isotope nuclear data are presented.

1. Introduction

Actinide bearing wastes are mainly discarded from the nuclear fuel cycle during fuel fabrication and fuel reprocessing. Waste management is confronted with three problem areas:

- actinide bearing waste constitutes a health hazard caused mainly by possible ingestion of α-emitters and by possible neutron exposure,
- a final waste treatment technique has to cope with difficulties caused by the decay heat and the radiation damage in part of the waste,
- a final waste disposal technique has to assure a complete isolation of the actinides from the biosphere for a period that is sufficient for actinide decay.

The problems are well recognized and solutions have been proposed which are now being studied by national and international organizations.

2. Composition and Partitioning of Actinide Waste

2.1. Waste from Fuel Fabrication (10)

Wastes from fuel element fabrication plants consist mainly of gloves, paper, plastics, slags, ion exchange resins, tools, glass, glove boxes, etc. Aside from plutonium, uranium, or thorium, they contain practically no other actinides or radioactive fission or corrosion products. Plutonium recovery is carried out only from wastes containing this element in relatively

high concentration. As the cost of plutonium recovery is relatively high and the price of plutonium is relatively low, there is no strong incentive for recovery from an economic point of view.

Plutonium recovery can be achieved by:

- incineration of burnable waste and extraction of plutonium from the ashes,
- redissolution of slags or cleaning of contaminated material and chemical separation of plutonium from the resulting solutions.

Due to the composition of the waste, the purification of the recovered plutonium does not pose insuperable technical problems. Table 1 gives a survey of the types and amounts of plutonium containing wastes arising at fuel element fabrication plants. From this table it can be seen that significant amounts of plutonium are lost to the radioactive stream in this type of installation.

Type of waste	Volume of waste per ton of fuel (m^3/t)	Pu-content of waste (g Pu/m^3)	Pu-losses to waste per ton of fuel (g Pu/t)
non-burnable waste	0,5	30	15
com-bustible waste	5	50	250
liquid waste	1	75	75
Total	6,5		340

Table 1: Amount of Pu-containing wastes generated at fuel-element production plants for LWR-Pu (3 %) -recycle-fuel.

2.2. Fuel Reprocessing Waste (7, 10, 12)

The actinide content of spent fuel has been determined experimentally (11) and predicted by computer codes (6,7). Results of different reactor types are summarized in Tables 2 and 3. The production rate (Kg/GWe year) depends on various parameters such as reactor type, initial fuel composition, flux level, irradiation history and exposure.

During reprocessing of spent nuclear fuels, plutonium and uranium (or thorium) are separated from fission products and the other actinides. For this purpose, a number of chemical processes (extraction, ion-exchange, precipitation) are used.

Isotope	Mass (kg)/GWe-year	
	BWR (*)	Fast Reactor (**)
Np-237	16.	2.4
Am-241	1.5	16.
Am-242m	0.031	0.54
Am-243	3.1	12.
Cm-242	0.18	0.87
Cm-243	0.0037	0.15
Cm-244	1.1	3.0
Cm-245	0.063	0.16
Cm-246	0.0072	0.001

Table 2: Calculated yield of higher actinides produced per year (6).

(*) BWR — Burn up to 33,000MWD/tonne.

(**) Fast Reactor — Fuel AGR PuO/UO mixture. Enrichment 22.8 %. Burn up 10 % Stored 1 year before irradiation and cooled 200 days after irradiation.

In the course of these processes, plutonium and uranium become more and more purified, in other words, contain less and less fission products and transplutonium elements. As in any of the processing steps, a small portion of uranium and plutonium is lost. All wastes from reprocessing plants contain some actinides.

The high activity liquid waste from the first extraction cycle contains about 0,1 - 0,5 % of the uranium and plutonium originally present in the spent fuel, and more than 99 % of all the fission products and other actinides (Am, Cm), except neptunium, in the BNFL plant (6).

Wastes from the intermediate process steps contain much smaller amounts of actinides and fission products. Wastes from the final purification and product separation in the uranium and plutonium tail-end contain these elements in a fairly pure form, and almost no fission products or other actinides are present. As a consequence, wastes from the tail-end can be treated more or less like wastes from plutonium-fuel fabrication, and plutonium recovery does mot present major technical problems. On the other hand, the recovery of actinides from the other waste streams poses considerable problems and is, up till now, not yet solved.

Reactor Name, Type	Enrichment (W/o)	Exposure (GWD/t)	Pu-236 (mg/t)	Np-237 (g/t)	Pu-238 (g/t)	Pu-241 (g/t)	Am-241 (g/t)	Am-242 (g/t)	Am-243 (g/t)	Cm-242 (g/t)	Cm-244 (g/t)
GARIGLIANO	U5 = 1.6	14	0.08	114	27	550	18	0.27	14	7.2	2.5
BWR	U5 = 2.1	10	0.12	219	13	385	27	0.51	5	3.3	0.6
	U5 = 2.4	17	n m	n m	36	559	37	2.52	17	6.8	2.7
	Pu = 3.2	16	n m	n m	50	2220	216	4.04	49	31	9.1
TRINO	U5 = 2.7	16	n m	n m	34	685	6.6	0.02	0.58	4.6	2.2
VERCELLESE	U5 = 3.1	17	n m	n m	34	675	1.7	0.004	0.14	6.8	1.9
PWR	U5 = 3.9	12	n m	n m	18	346	13	0.03	0.4	1.2	0.5

Table 3: Actinide content in spent BWR and PWR fuel at reactor shut down (12) (nm = not measured).

In the case of high active waste, the complete separation of actinides is made difficult by the fact that -among otherthings- reagents are partially decomposed by action of the high radiation doses and that a part of the plutonium forms non-extractable species during reprocessing. The difficulties in the separation of actinides from intermediate level wastes result mainly from the large amounts of complex chemicals contained in these waste streams. In addition, the cost of such a process seems to be prohibitive. Thus, for the time being, nowhere are actinides separated from high and intermediate level wastes. Types and amounts of α-wastes arising in reprocessing are listed in Table 4.

Type of waste	Waste volume per ton of fuel (m^3/t)	Pu-content of waste ($g\ Pu/m^3$)	total α-activity of waste (Ci/m^3)
High- level waste concentrate	1	30	10.000
Solid high-level waste	0,5	50	30
Liquid intermediate level waste	2000	0,01	0,01
Liquid low-level waste	1500	-	-
Solid intermediate and low-level waste	0,5	10	2
Liquid -waste from Pu-tail-end	0,1	50	10
Solid α-waste from Pu-tail-end	0,1	50	10

Table 4: Plutonium-content and total α-activity in wastes generated in a LWR-fuel reprocessing plant.

2.3. Data requirements

The uncertainties of long-term actinide content in wastes depends mainly on future reactor strategies, fuel exposure, and to what extent and where plutonium will be recycled. Therefore, the data requirement, concerning this topic in general, should be covered by the review papers A2, A3 and A4. Specifically more accurate data are needed for the production routes to curium and eventually californium, which are important for $^{238}U/Pu$ cycles. Cross sections for capture and fission as a function of energy and values of the ^{241}Am (n,γ) branching ratio as a function of energy are needed. It is known, for example that for typical fast reactor runs, the ^{241}Am and ^{243}Am capture cross section values in the ORIGEN library are factors of 2 or 3 less than the U.K. recommended values in use (7) and are considered to be incorrect (6).

3. Disposal of Actinide Bearing Waste

Several concepts for the final disposal of nuclear waste have been studied; presently, the disposal on earth in suitable geological formations, or the engineered storage have received acceptance. Advanced waste management concepts, such as disposal in space, deep sea or ice sheets, still require additional research, as does nuclear incineration. Disposal in geologioal formations can be a suitable method, if complete isolation from the biocycle can be assured for the necessary period that is sufficient for actinide decay.

3.1. Waste Storage

All waste management concepts, with the exception of nuclear incineration depend on disposal techniques which have to isolate actinide waste from the biosphere for a million years. To achieve this difficult task the effects associated with radioactive decay, in addition to corrosion, erosion, etc. have to be studied. Long term disposal requires the waste to be solidified; and various matrices have been proposed (e.g. glass etc.) for this purpose. In order to evaluate the radiation damage as well as the decay heat transfer in these materials, the decay schemes and energies have to be known. Neutron emission in particular causes problems during the handling of the waste. Apart from dose rates of spontaneous fission neutrons, those generated by (α,n) reactions must be known. As this reaction is matrix dependent, (α,n) cross sections for several elements are required. The time dependent decay heat production in spent fuels and reprocessing waste of various initial fuel compositions (Magnox, AGR, SGWR, LWR, HTR and Fast Reactors) has been calculated (7).

3.2. Nuclear Incineration

The use of plutonium in a fast reactor allows for complete access to the energy production by U-238. Although they could be made into reactor fuels, the recycling of the other major actinides, such as neptunium, americium and curium, however, cannot be justified on economic grounds. The separation of these elements from the spent fuel is very costly. However, the long term hazard of these actinides might justify this complete recovery, even if the benefit would not be apparent for 600 years (6). As the amount of neptunium, americium and curium is considerably less than that of plutonium, the associated risk is also smaller. Any argument making plutonium recycling acceptable to the public would therefore also be valid for recycling neptunium, americium and curium.

A list of European nuclear incineration studies, which are known to the author, are listed in Table 5.

Fast reactors incinerate actinides more efficiently into fission products (7,12) then do thermal nuclear power plants. The amount of actinides transmuted into other actinides - mainly by Cm-242 and Cm-244 decay - is low (see Figure 1). In order to assess quantitatively the amount of actinide built-up along the transmutation path in fast reactos, cross sections for radioactive neutron capture and neutron induced fission are needed. The energy range pertinent to the cross sections should cover the neutron energy spectrum of pre-

Country and/or organization	Objective of Research Programme	Reference
France C.E.A.	Neutronic calculations in fast reactors. Actinide chemistry	(1)
Germany F.R. G.f.K.	System analysis of the incineration process	(2;3)
Sweden Chalmers Technical University	Flowsheet studies of chemical separations and reactor calculations	(4)
United Kingdom UKAEA/CEGB	Calculation of incineration in fast reactors and evaluation of radiological hazards	(5,6,7)
EURATOM		
Indirect action	Assessment studies of nuclear incineration	(8)
Direct action	Studies on chemical separation, cross section measurements, systems analysis	(9)

Table 5: List of European research activities in nuclear incineration.

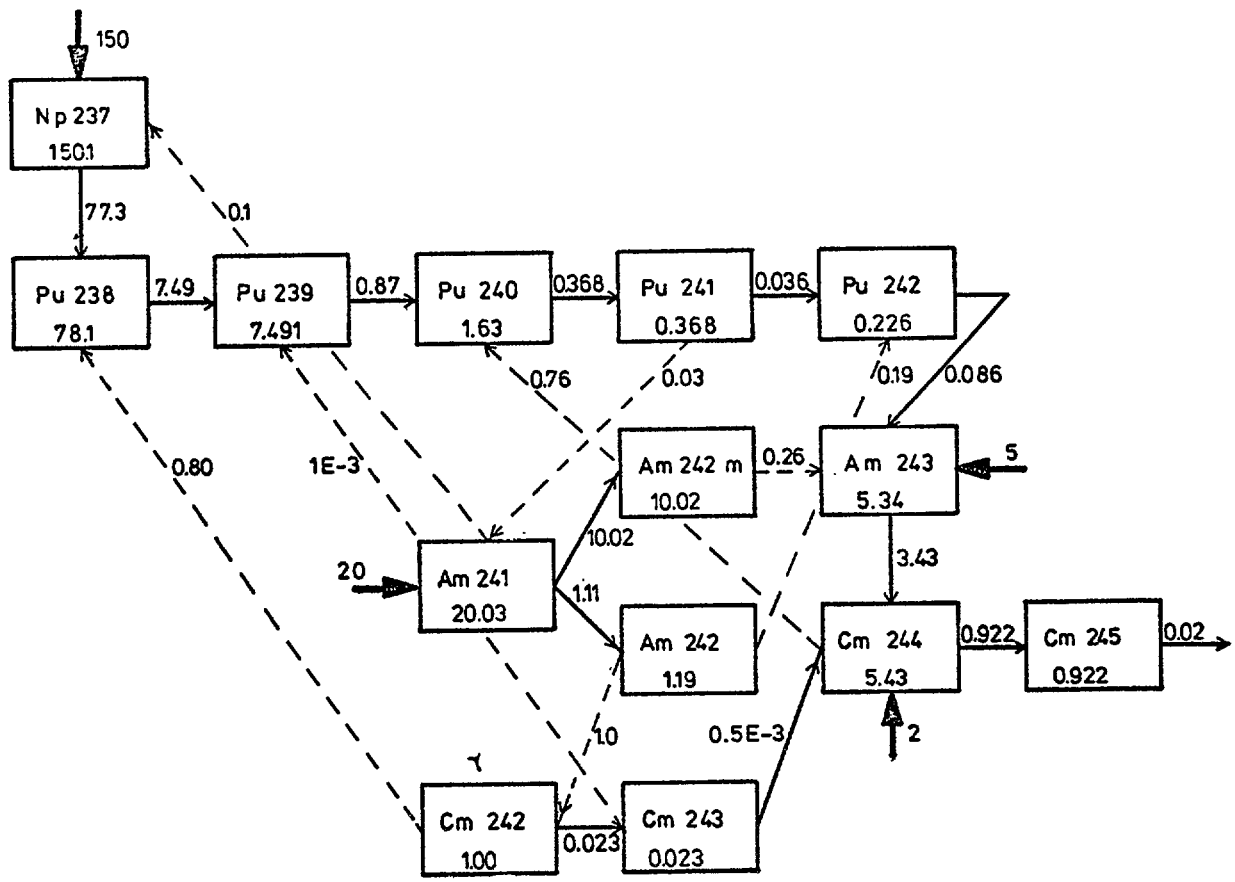


Fig 1: Build up path of actinides

sent fast reactor prototypes. For the major actinides, a precision of 20 % would be required at this conceptual stage in order to compare the potential of oxide and carbide fueled fast reactors for nuclear incineration purposes. The branching of the built-up path at Am-241, Am-242, Am-243 should be known with the same precision. In the event that nuclear incineration becomes operational, higher nuclear data accuracies will be required. The decisive advantage of fast reactors compared to thermal ones is a better neutron economy for the incineration process. For this reason, precise data (± 10 %) of the ν-values are required.

For nuclides produced in minor quantities during the incineration process a lower precision of 50-100 % is needed. As most of the nuclides will follow the plutonium chain, the data requirement is fortunately reduced (see Figure 1) (6,12).

The fate of typical actinide waste discharged during LWR fuel reprocessing, if exposed to a fast reactor flux of 7.10^{15}n/sec cm^2, can be seen from Figure 1. The fraction of the actinide nuclides not incinerated into fission products, but transmuted by radiative neutron capture or by decay into another actinide is indicated. The built-up of actinides beyond Cm-245 is negligible. The largest portion of the actinide waste not incinerated is transmuted into plutonium isotopes, for which the precision of nuclear data is already better than that needed for nuclear incineration.

4. Nuclear Data Requirements

Table 6 summarizes nuclear data required for the different aspects of nuclear material management. The precisions of nuclear data for nuclear incineration correspond to the present requirements of conceptional studies. A sensitivity study under way at Harwell and CCR Ispra will define more precisely the data needs for these calculations.

The required accuracies attributed to each nuclide and topic may even be too high. The uncertainty in the prediction of future actinide built-up depends more on the possible reactor type and its mode of operation than on unprecise nuclear data. The health physics aspect of nuclear waste management does not call for precise nuclear data.

Priorities concerning data requirements are difficult to allocate to each of the various topics due to changing importance of the various research programmes. It seems, however, that in the first place the danger of neutron exposure of workers during waste handling would justify an improvement of $\sigma(\alpha,n)$ data.

NUCLIDES		Pa	Th	U						Np	Pu						Am					Cm					REMARK
		231	232	232	233	234	235	236	238	237	236	238	239	240	241	242	241	242 m	242	243	244	242	243	244	245	246	
NUCLEAR DATA REQUIREMENTS																											
1. Actinide prediction in spent fuels	$\sigma(n,\gamma)$	100	20		20	100	20	50	20	30	100	30	20	20	20	30	20	30		30		30	30	30	100		Thermal and fast Reactor n-energy spectra
	$\sigma(n,f)$		20		20		20		20	30		30	20	20	20	20	20	20		30		30	30	30	100		
	$\sigma(n,2n)$		50		50				50				50														
2. Decay heat of and radiation damage by actinides	decay scheme & energy	+	+	+	++	+	+	+	+	++		++	++	++	++		+	+		+			+	+			and daughter products
3. Neutron dose rates in different matrices	spont, fission			100		100		100			100	100		100		100	100		100	100		100		100		100	Handling of waste
	$\sigma(\alpha,n)$	100		100	100				100	100		50	50	50			50	100		100		100		100			
4. Nuclear incineration	$\sigma(n,\gamma)$	50		50						30		30	30	30		30	30	30	100	30		30	30	30	50	100	Fast reactor n-energy spectrum
	$\sigma(n,f)$	20		20						20		20	20	20		20	20	20	20	20		20	20	20	20	50	
	decay scheme																				+						
	ν-value	10		10						10		10	10	10	10	10	10	10	++	10		10	10	10	20		

Table 6: Summary of data requirements in waste management of actinides (accuracies given in percent, referring to integral cross sections of power reactors, + less important, ++ very importan)

References:

(1) Contribution to the review paper by D. Breton, France

(2) H.O. Haug, KFK-1945 (1974)

(3) P.E. McGrath, KFK-1992 (1974)

(4) Contribution to the review paper by J.O. Liljenzin, Chalmers Tekniska Hoekskola, Goeteborg

(5) N.J. Keen paper presented at I.A.E.A. Technical Committee on high-level and α-bearing wastes, Vienna (1975)

(6) Contribution to the review paper by M.G. Sowerby, U.K.A.E.A., Harwell

(7) Contribution to the review paper by R.H. Clarke, C.E.G.B., Berkeley, U.K.

(8) Europaeische Gemeinschaften, Programm fuer die Bewirtschaftung und Lagerung radioaktiver Abfaelle, R/1447/75 (ATO 73) 1975

(9) EURATOM: Pluriannual programme

(10) Contribution to the review paper by H. Krause, G.f.K., Karlsruhe

(11) H.H. Grain, WCAP -3385-37 (1973)

(12) L. Koch, Accumulation of TPu elements in the wastes and possibilities for their nuclear incineration, 4th Internation TPu-Element Symposium, Baden-Baden, 1975

Review Paper A 7

Some Activities in the United States Concerning the Physics Aspects of Actinide Waste Recycling*

S. Raman
Oak Ridge National Laboratory, Oak Ridge, Tennessee 37830

Abstract

This review paper briefly discusses the reactor types being considered in the United States for the purpose of actinide waste recycling. The reactor types include thermal reactors operating on the 3.3% ^{235}U-^{238}U and the ^{233}U-^{232}Th fuel cycles, liquid metal fast breeder reactors, reactors fueled entirely by actinide wastes, gaseous fuel reactors and fusion reactors. This paper also discusses cross section measurements in progress or planned toward providing basic data for testing the recycle concept.

INTRODUCTION

One of the most urgent needs of the nuclear power industry today is a viable plan which is technically sound and has public acceptance, for the long-term management of radioactive waste. Many suggestions are being considered for dealing with these wastes in terms of disposal or storage such as retrievable storage in near surface vaults and deep underground mines, irretrievable disposal in salt domes, deep holes drilled in stable rock formations, in the sea, outer space, etc. Although it may be feasible to package and safely store wastes for periods as long as a thousand years, there is no way to ensure complete containment for the more than 250,000 years necessary for the actinides (and a few fission products) to decay to innocuous levels. One suggested method for alleviating this problem would involve removing nearly all of the long-lived actinides (thorium, protactinium, uranium, neptunium, plutonium, americium, and curium) contained in the wastes and disposing of this fraction by transmutation to fission products (which decay essentially to nonradioactive isotopes in less than 1000 years).

*Research sponsored by the U.S. Energy Research and Development Administration under contract with the Union Carbide Corporation.

Recycling requires a high-flux neutron source, which might be the same reactor producing the wastes or a specially designed burner reactor or even a fusion reactor. Transmutation calculations require neutron cross sections which are more complete and of higher accuracy than are now available. This review paper will focus attention on only two aspects of the actinide waste problem: (a) reactor types under consideration for recycling and (b) cross section measurements in progress or planned in support of the recycling concept. The specific cross section requirements have been outlined in a separate review paper [1]. It should be mentioned at the outset that recycle calculations carried out till now are preliminary in nature. They are in the form of feasibility studies carried out quickly with whatever cross section data were readily at hand. The computer codes used for these calculations contain no provisions for entering data uncertainties. Detailed sensitivity studies have not been carried out to the best of our knowledge.

RECYCLING IN THERMAL REACTORS

The first reported detailed study of actinide recycling was carried out by Claiborne [2] who considered recycling in light-water reactors (LWR) operating on the 3.3% ^{235}U-^{238}U fuel cycle. The reductions in the hazards due to actinides are brought about in two ways. U and Pu recoveries of up to 99.5% have been attained in the conventional reprocessing of LWR fuel; in some cases, 90 to 95% of neptunium has also been recovered, but Am and Cm have been routinely discharged in the high-level waste. Claiborne [2] showed that it is possible to reduce the hazard (at >1000 years) associated with high-level wastes to values comparable to those from naturally occurring uranium deposits provided that 99.99% of the Pu, 99.9% of the U, Am, Cm and ^{129}I, and 95% of the Np are recovered from LWR fuels. Except perhaps for U and Pu, the removals required are beyond present technology.

If essentially all of the actinides normally discharged in the waste are recovered and recycled in a reactor, the inventory becomes stabilized at a value several times the quantity produced in the first cycle. In other words, the inventory approaches a maximum value, an asymptote, rather than increasing linearly with time (see Figure 1). Claiborne [2] showed that through the combination of chemical separations and recycling, the hazards in the fission product wastes can be reduced by a factor of 50 to 200. Claiborne's calculations have been repeated and extended by the group at Battelle Pacific Northwest Laboratories [3] with similar results. The technical assessment of transmutation is only one of several topics discussed in the Battelle study [3] which is a comprehensive review of potential alternative methods for long-term management of high-level radioactive waste. This study identified continued recycling in thermal power reactors as the

most promising method of transmutation of actinides within the constraints of existing technology provided that the chemical separation problems can be solved.

The low flux level of 3×10^{13} n/cm^2 sec used by Claiborne in his calculations results in long irradiation times of approximately 50 to 60 years. Even then, sustained recycling of actinides reduces the overall inventory of the actinides by only a factor of 3. The calculations carried out by Raman, Nestor and Dabbs [4] showed that further reductions in actinide inventories, especially those due to transuranium isotopes, should be possible through recycling in a ^{233}U-^{232}Th reactor. In such a reactor, the capture of neutrons by the ^{232}Th fertile material leads to the replenishment of the original ^{233}U fuel. Successive neutron captures by ^{233}U result in higher U isotopes until ^{237}U is reached, where a higher Z isotope, ^{237}Np, is produced. It is further obvious that plutonium and transplutonium isotopes are generated to a far lesser extent in a ^{233}U-^{232}Th reactor when compared to a ^{235}U-^{238}U reactor. For nuclides above ^{238}Pu, the typical reduction factor is $>10^6$ because in the ^{233}U case, five additional neutron captures are required to reach the same mass. Therefore, due to the absence of substantial new production of these elements, the ^{233}U-^{232}Th reactor can be effectively employed to reduce the Np, Pu, Am and Cm inventories.

RECYCLING IN FAST REACTORS

The ultimate aim in recycling actinides is to induce fission because transmutation of one actinide to another probably has little potential for decreasing toxicity. It was recognized early in the recycling studies that the use of fast reactors should result in faster burnup than in thermal reactors. The fission to capture ratio is generally higher for fast reactor neutron spectra. The combination of flux and cross section results in higher fission rates and lower capture rates as shown in Table I. Preliminary calculations for the burnup of actinides in a Liquid Metal Fast Breeder Reactor (LMFBR) have been carried out by Breen [5] and by Beaman [6]. The equilibration of actinides recycled in an LMFBR is qualitatively similar to that shown in Figure 1 except that equilibrium is approached in a shorter time and the equilibrium values are lower suggesting a more efficient approach for the removal of the actinides. Kubo and Rose [7] have pointed out that recycling of the actinides in an LMFBR has several additional advantages over recycling in a thermal reactor. Extreme separations of chemical groups may not be required because a less pure actinide product should be recyclable without degrading the neutron economy in an LMFBR. The incorporation in the fuel element of the actinides to be recycled should not be too difficult since the fabrication of LMFBR fuel

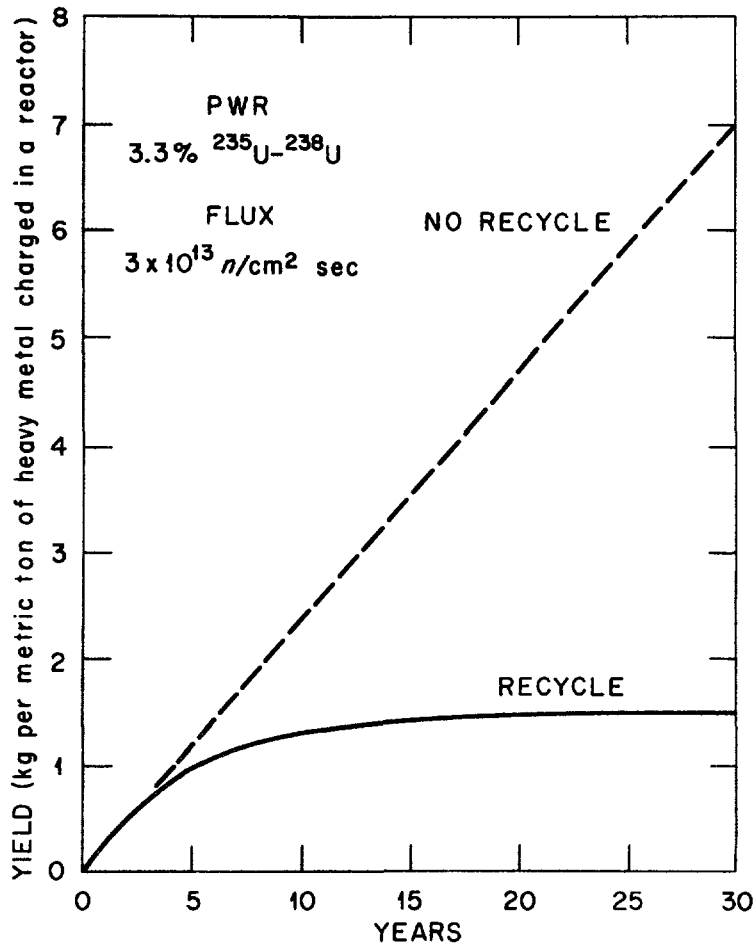


Figure 1. Influence of recycling in the yield of selected actinides (^{237}Np + ^{241}Am + ^{242}Am + ^{243}Am + ^{242}Cm + ^{243}Cm + ^{244}Cm + ^{245}Cm). Figure based on calculations given in Reference 3.

TABLE I

ACTINIDE REACTION RATES IN FAST AND THERMAL REACTORS[†]
(Reactions/sec/Atom)

		Fast Spectrum*		Thermal Spectrum**	
Isotope	Half-Life, Years	Fission Reaction Rate	Capture Reaction Rate	Fission Reaction Rate	Capture Reaction Rate
Np^{237}	2.14×10^{6}	2.2×10^{-9}	1.03×10^{-8}	6.18×10^{-12}	4.9×10^{-8}
Am^{241}	433	2.7×10^{-9}	2.35×10^{-8}	6.18×10^{-10}	1.38×10^{-7}
Am^{242m}	152	4.7×10^{-8}	9.69×10^{-9}	1.49×10^{-7}	1.33×10^{-7}
Am^{243}	7370	1.39×10^{-9}	4.5×10^{-9}	1.55×10^{-11}	3.18×10^{-8}
Cm^{244}	17.9	3.47×10^{-9}	2.77×10^{-9}	4.02×10^{-10}	5.9×10^{-9}

[†]From Reference 6
*Average Total Flux = 6.93×10^{15} in Core Zone 1
**Average Total Flux = 3.09×10^{14}

elements would already include provisions for handling the highly toxic Pu isotopes.

RECYCLING IN OTHER TYPES OF REACTORS

A group of fifteen senior and graduate students working with Binney and Spinrad [8] of Oregon State University (Nuclear Engineering Department) have carried out interesting studies on the use of fast reactors for the destruction of actinide wastes. Initially, they considered liquid metal fast breeder and gas cooled fast breeder reactors. They found that the actinide destruction rates in such reactors was better but not by what they deemed to be a significant factor over rates to be expected in thermal reactors. These reactors are not really "fast" but intermediate and fail to take full advantage of the excellent neutron economy of the fast chain reaction. The follow-up study focused attention on hard-spectrum (>100 keV) fast reactors of three types: sodium-cooled, metal fueled; sodium-cooled, carbide fueled; and helium-cooled, carbide fueled. The fuel, and this is the interesting part, consisted of all the higher actinides (Np, Am, Cm, and 0.5% of Pu from twenty 1000 MWe LWRs). It was found that a reactor with a core of the size of the Idaho Experimental Breeder Reactor II (EBR-II) or smaller would go critical on essentially pure actinide fuels with a margin of reactivity sufficient to permit some considerable dilution. A key question was the extent to which dilution with inert materials such as Zr and ZrC could be tolerated and still maintain very high specific powers. To answer this question and to explore fully the possibilities of fast reactors operating on actinide fuels, extensive data (σ_f, ν, η, etc.) for E_n > 100 keV would be needed. In the absence of such data for ^{241}Am, ^{243}Am and ^{244}Cm, Binney and Spinrad and their coworkers were forced to employ the known cross sections for ^{240}Pu for all three of these materials.

Paternoster, Ohanian, Schneider and Thom [9] have considered the use of a conceptual gaseous core reactor for transmutation of fission product (^{129}I and ^{99}Tc) and actinide wastes (Am and Cm). The main feature of such a reactor is the use of fissile fuel in a gaseous or plasma state thereby breaking the barrier of temperature imposed by solid-fuel elements. The fuel is UF_6 enriched to 6 percent ^{235}U. A reactor with a continuous fuel cycle (whether of the gaseous type or the molten salt type) requires a much lower inventory of fissile and fertile material. With continuous fuel addition and removal of xenon and other volatile fission product poisons, higher fuel burnup rates can be achieved. For these reasons, the UF_6 gas core reactor generates less actinide waste than the conventional LWR. The rates of transmutation were found to depend strongly on the ratio of resonance-to-thermal neutron fluxes. Since the neutron energy spectrum of a gas core

reactor is not known with certainty, Paternoster et al considered three representative values 0.058, 0.29 and 0.58 for the resonance/thermal ratio. A five-year transmutation of Am and Cm wastes resulted in an order-of-magnitude decrease in the overall hazard potential of actinide wastes generated in 60 reactor-years of LWR operation. The calculations assumed an average thermal neutron flux of 6.4×10^{14} n/cm^2 sec.

RECYCLING IN FUSION REACTORS

In 1973, Wolkenhauer, Leonard and Gore [10] made a survey study of actinide transmutation in the blanket of a conceptual thermonuclear reactor (CTR). Calculations were made for neutrons from both a D-D and a D-T plasma. The authors assumed two wall loadings, 1.0 MW/m^2 and 10.0 MW/m^2, in order to determine the neutron flux. With Be in the blanket by which the 14-MeV neutrons from the D-T reaction could be multiplied by a factor of 2.5, thermal neutron fluxes of 3×10^{15} and 3×10^{16} were obtained with the two wall loadings. These flux levels are 100 to 1000 times higher than in an LWR. Therefore, it was possible to consider not only the rapid transmutation of actinides but also the transmutation of problem fission products such as ^{85}Kr, ^{90}Sr, ^{137}Cs and ^{129}I. In addition to the fission and capture cross-sections, the CTR studies established the need for (n,2n) and (n,3n) cross section data for many nuclides, especially for ^{237}Np, ^{239}Pu and ^{243}Am. Draper and Parish [11] have undertaken a more detailed evaluation of a CTR device for transmutation of high-level wastes. With an array of computer programs, they plan to study plasma conditions, blanket designs, heat removal systems and waste configurations that will optimize waste disposal.

Quite recently, Rose, Olhoeft, Kellman, et al [12] have suggested that fission product and actinide waste burning can be accomplished with high energy neutrons from a device similar to the proposed Tokamak Fusion Test Reactor at Princeton which is a device of intermediate size wherein the conditions for D-T burning are achieved by the injection of high energy neutral beams into a relatively cool plasma. Further neutron multiplication is envisaged through the use of a subcritical assembly of fissile fuel in the blanket walls -- a fusion-driven fission reactor. The blankets considered will include fast fission liquid cooled blanket, beryllium-graphite moderated helium cooled thermal blanket, molten salt blanket and aqueous homogeneous blanket.

CROSS-SECTION NEEDS AND MEASUREMENTS

The quantitative prediction of the various nuclei produced, transmuted and fissioned in reactors is necessary for systematic actinide management.

These quantitative predictions are made with special computer programs which require as input values all relevant cross sections of all the nuclides in the region of interest. At present, most codes utilize effective values at thermal, resonance and fast neutron energies. These are obtained from suitable weighted averages over the reactor spectrum in question, and require detailed data. Benjamin [13] has surveyed neutron cross-section measurements of the actinides with emphasis on those desired for an LWR for ^{252}Cf and ^{238}Pu production. In many isotopic cases, an adjusted set of cross sections has been used because of the urgent need to perform the calculations in spite of gaps in our knowledge (see Table II) of these cross sections. Such an "adjusted set" of cross sections has limited applicability. Therefore, it will be necessary to make neutron cross-section measurements to obtain better predictions. There are certain obvious deficiencies in the data which can be attacked immediately. There is a total lack of differential fission cross section data below 15-eV neutron energy from underground nuclear explosions on several fissile nuclides of Cm and Cf. The deficiencies in capture cross sections are numerous especially over the fast-reactor spectrum region. In the determination of fission and capture widths in the resolved resonance region, a knowledge of total cross sections over the same neutron energy region can supplement fission and capture measurements. For the fast reactor calculations, some determinations of (n,2n), (n,3n) and (n,charged particle) cross sections will likely be needed. Pulsed neutron sources and the techniques for the measurement of many of the needed cross sections are in existence. Underground nuclear explosions [14] have already provided a wealth of fission cross-section measurements on the actinides. We discuss briefly some measurement programs on the actinides currently proposed or underway in the United States, spurred mainly by the urgency of the actinide waste problem.

INTEGRAL MEASUREMENTS

Recycling studies in the LMFBR are seriously handicapped by the non-availability of differential cross section data. In the interim, the Argonne group composed of Fields, Fried, Friedman and Unik [15] propose that gross burnout measurements made at selected locations in the Idaho Experimental Breeder Reactor-II (EBR-II) will provide integral data which can be employed for feasibility studies. In fact, forty samples of separated nuclides ranging in mass from ^{232}Th to ^{241}Pu have been already irradiated for four years in EBR-II receiving an integrated dose of 10^{23} fast neutrons. These samples have been cooling for over four years. The Argonne group intends to carry out mass spectrometric and radiometric analyses of the purified actinide and selected fission product fractions. It is planned that addi-

TABLE II[†]

Actinide Cross Sections in Barns

(Uncertainties given as standard deviation)

	$\sigma_{n\gamma}^{2200}$	σ_{nf}^{2200}	Resonance Integrals $I_{n\gamma}$	Resonance Integrals I_{nf}
92-U-236	5.3 ± 0.2	a	358 ± 20	a
92-U-237	478 ± 160	a	b	a
93-Np-237	172 ± 3	a	819[c]	a
93-Np-238	b	2200 ± 200	b	1500 ± 500
94-Pu-238	559 ± 20	17.3 ± 0.4	164 ± 15	25 ± 5
95-Am-241	832 ± 21	3.14 ± 0.10	1538 ± 118	21 ± 2
95-Am-242	b	2100 ± 200	b	< 300
95-Am-243	77 ± 2	a	1813 ± 70	b
96-Cm-242	b	5	b	150 ± 40
96-Cm-243	b	960 ± 50	b	1860 ± 400
96-Cm-244	14 ± 4	1.1 ± 0.5	606 ± 23	18 ± 1
96-Cm-245	342 ± 20	2018 ± 37	102 ± 7	772 ± 30
96-Cm-246	1.3 ± 0.3	0.17 ± 0.10	121 ± 7	10 ± 0.4
96-Cm-247	60 ± 30	82 ± 5	800 ± 400	778 ± 50
96-Cm-248	4 ± 2	0.34 ± 0.07	300 ± 75	13.2 ± 0.8

[†]From Reference 13
a - unknown, but probably quite small
b - unknown, but probably quite appreciable
c - probably too high

The cross sections for Bk and Cf are either unknown or very poorly known.

tional samples of ^{242}Pu, ^{241}Am and ^{243}Am will be inserted in EBR-II for irradiation. A computer model will be developed to calculate the yields of various actinides and fission products which will be compared with measured yields.

The Los Alamos Radiochemistry Group [16] has also made several integral cross section measurements in critical assemblies utilizing activation and fission chamber techniques. The samples studied include ^{235}U, ^{236}U, ^{237}U, ^{238}U, ^{237}Np, ^{238}Pu, ^{239}Pu, ^{240}Pu, ^{241}Pu, ^{242}Pu and ^{241}Am. The data have not been fully analyzed but with sufficient impetus and interest generated

by the actinide recycle concept, the cross section data should become available.

MEASUREMENTS WITH ELECTRON LINEAR ACCELERATORS

The electron linear accelerators provide a versatile pulsed neutron source for detailed differential cross section measurements. Sample acquisition, purification and preparation constitute a major part of any cross section measurement program. The minimum quantity of separated isotope needed for cross section measurements is about 30 μg, 10 mg and 100 mg for fission, total and capture cross section measurements, respectively. The availability of samples is discussed at length in another review paper[1].

The ratios of the fission cross sections of ^{234}U and ^{236}U relative to the ^{235}U fission cross section have recently been measured [17] at the Lawrence Livermore Laboratory (LLL) Linear Accelerator in the neutron energy range 0.1 to 30 MeV. Typical energy resolution was 5% at 20 MeV and 1.5% at 1 MeV. Counting uncertainties were less than 4%; overall systematic uncertainties were estimated to be less than 5%. These data were presented recently [18]. Future plans include the measurement of the fission cross section ratios of ^{238}Pu, ^{240}Pu, ^{241}Pu, ^{242}Pu and ^{244}Pu relative to ^{235}U over this same energy range.

Preliminary results for the fission cross section of ^{245}Cm have also been obtained at the LLL linac in the neutron energy range 0.006 eV to 20 eV [17]. Similar measurements covering the neutron energy range from 0.01 eV to 14 MeV are planned with ^{242m}Am, ^{243}Cm, ^{245}Cm, ^{247}Cm and ^{249}Cf. The LLL facility also possesses the capability of measuring $\bar{\nu}$, the number of prompt fission neutrons, as a function of incident neutron energy from 0.01 eV to 14 MeV. Future plans for these measurements will be based on need and interest for various actinides.

A detailed program for the measurement of actinide cross sections has been recently formulated at the Oak Ridge Electron Linear Accelerator (ORELA). The initial set of nuclides to be investigated is given in Table III. Preliminary results for the fission cross section of ^{245}Cm and total cross section of ^{249}Bk are discussed in another review paper [1]. Capture studies on ^{240}Pu, ^{241}Pu, and ^{241}Am are currently being carried out at ORELA [19]. Some short-lived isotopes have enough gamma ray and spontaneous fission activity to preclude normal differential capture cross section measurements. Some of these measurements may possibly be done with underground nuclear explosions [14]. Total cross section measurements can give good estimates of the capture cross section at low energies in the well-resolved region if appreciable fission is not present. Such measurements also are needed to give accurate resonance fission widths. Total cross section measure-

TABLE III

Proposed Five-Year Program at the Oak Ridge National Laboratory for Carrying out Needed Cross-Section Measurements in the Actinide Region

Isotope	Type	Proposed Energy Range	Measurement Techniques Required	Sample Availability
^{237}Np	σ_c	Thermal - 0.5 MeV	Current	Available
^{242m}Am	σ_c	Thermal - 0.5 MeV	Current	Very difficult[a]
^{242m}Am	σ_f	Thermal - 1.5 MeV	Current	Available
^{243}Am	σ_c	Thermal - 0.5 MeV	Current	Available
^{243}Am	σ_f	0.3 eV - 1.5 MeV	Some development required	Available[b]
^{243}Cm	σ_f	0.3 eV - 1.5 MeV	Some development required	Available[b]
^{245}Cm	σ_c	Thermal - 0.5 MeV	Current	Very difficult[c]
^{245}Cm	σ_f	0.3 eV - 1.5 MeV	In progress	12 μg available
^{247}Cm	σ_c	Thermal - 0.5 MeV	Current	Extremely difficult[d]
^{247}Cm	σ_f	0.3 eV - 1.5 MeV	Current	Very difficult[e]
^{251}Cf	σ_f	0.3 eV - 1.5 MeV	Current	Very difficult[f]

a. Irradiation of feed material and calutron separation necessary.
b. Feed material available. TRL isotopic separation required.
c. Calutron separation necessary. Available facilities require additional shielding and containment.
d. Preparation and irradiation of feed material very difficult. Calutron separation necessary. Available facilities require additional shielding and containment.
e. Feed material available. Difficult TRL isotopic separation required.
f. Small quantity of feed material available. Very difficult TRL separation required.

(TRL - Transuranium Laboratory (ORNL))

ments with samples as small as 10 mg have been carried out at ORELA. It should be noted that total cross section measurements are not practical with underground nuclear explosions.

(n,f) CROSS SECTIONS FOR EXOTIC ACTINIDES

It is highly desirable to have reliable (n,f) cross sections for as many actinides as possible. If for various reasons direct neutron measurements become impractical, Wilhelmy, Britt, Gavron, Konecny and Weber [20] have proposed an indirect method of extracting equivalent (n,f) cross sections from experimentally measured charged-particle induced fission probabilities (P_f) via the expression

$$\sigma_{nf}(E_n) = P_f(E_n+B_n) \times \sigma_{CN}(E_n)$$

where σ_{CN} stands for the total compound-nuclear neutron cross section calculated with the optical model and E_n and B_n represent the incident neutron energy and the neutron binding energy, respectively. The assumption made is that the fission probabilities are independent of the reaction mechanism which the authors have verified to within ≈10% by producing the same compound nucleus with different reactions on appropriate targets. The charged-particle reactions employed were the (^{3}He,df) and (^{3}He,tf) reactions with future plans for ^{6}Li and ^{7}Li induced reactions. Wilhelmy et al have carried out DWBA calculations which show that the angular momentum transfer in the (^{3}He,d) reaction is quite similar to that expected for incident neutron energies in the few MeV range. However, the charged-particle simulation technique is not expected to be reliable for neutron energies below 2 MeV. The compound nuclei produced with the (^{3}He,d) and (^{3}He,t) reactions and the maximum equivalent neutron bombarding energy studied are given in Table IV. These measurements would be equivalent to carrying out (n,f) measurements on targets listed in the "Neutron Target" column. Future plans for these types of measurements being carried out at the Los Alamos Scientific Laboratory would include ≈50 isotopes at excitation energies ranging up to ≈24 MeV.

The preceding four sections show that a substantial start has been made in developing programs for neutron cross section measurements in the actinide region. The individual measurements would be guided by sensitivity studies which have also begun. Many important decisions regarding waste utilization, management and storage of waste depend on the knowledge to be acquired through these measurement programs.

ACKNOWLEDGMENTS

The author wishes to acknowledge helpful discussions with many colleagues at the Oak Ridge National Laboratory, particularly J. O. Blomeke, H. C. Claiborne, J. W. T. Dabbs, G. F. Flanagan, J. A. Harvey, R. E. Leuze, R. L. Macklin, J. P. Nichols, R. W. Peelle, and L. W. Weston. He wishes to also thank W. C. Gough of the Electric Power Research Institute (Palo

TABLE IV

Charged-Particle Induced Fission Probability Measurements Carried Out at Los Alamos Scientific Laboratory to Simulate (n,f) Measurements[†]

Neutron Target	$t_{1/2}$	Max Energy Neutron (MeV)	Compound Nucleus
^{229}Pa	1.4 d	5.2	^{230}Pa
^{230}Pa	17.4 d	3.7	^{231}Pa
^{231}Pa	3.25×10^4 y	6.3	^{232}Pa
^{232}Pa	1.32 d	4.4	^{233}Pa
^{230}U	20.8 d	7.0	^{231}U
^{231}U	4.2 d	5.2	^{232}U
^{232}Np	14.7 m	3.0	^{233}Np
^{233}Np	35 m	3.9	^{234}Np
^{234}Np	4.4 d	6.2	^{235}Np
^{235}Np	396 d	6.6	^{236}Np
^{236}Np	1.3×10^6 y	4.3	^{237}Np
^{237}Np	2.1×10^6 y	7.7	^{238}Np
^{238}Np	2.12 d	5.2	^{239}Np
^{236}Pu	2.85 y	7.2	^{237}Pu
^{237}Pu	45.6 d	5.6	^{238}Pu
^{238}Am	1.63 h	5.1	^{239}Am
^{239}Am	11.9 h	5.5	^{240}Am
^{240}Am	51 h	3.9	^{241}Am
^{241}Am	433 y	6.8	^{242}Am
^{242}Am	152 y	4.6	^{243}Am
^{240}Cm	26.8 d	6.2	^{241}Cm
^{241}Cm	36 d	4.6	^{242}Cm
^{242}Cm	163 d	7.1	^{243}Cm
^{243}Cm	28 y	5.3	^{244}Cm
^{247}Bk	1.4×10^3 y	5.9	^{248}Bk
^{248}Bk	18 h	4.2	^{249}Bk

[†]Reference 20

Alto, California) and E. T. Jurney of the Los Alamos Scientific Laboratory for making relevant material available to him. This review was made possible through the splendid cooperation provided by the people listed below who sent to the author reprints, proposals, progress reports, explanatory material, etc., on the actinide waste problem.

Contributor	Institution
D. W. Barr	Los Alamos Scientific Laboratory, Los Alamos, New Mexico
S. L. Beaman	General Electric Company, Sunnyvale, California
R. J. Breen	Westinghouse Electric Corporation, Madison, Pennsylvania
J. C. Browne	Lawrence Livermore Laboratory, Livermore, California
E. L. Draper, Jr.	University of Texas, Austin, Texas
B. R. Leonard, Jr.	Battelle Pacific Northwest Laboratories, Richland, Washington
R. P. Rose	Westinghouse Electric Corporation, Pittsburgh, Pennsylvania
B. I. Spinrad	Oregon State University, Corvallis, Oregon
K. Thom	National Aeronautics and Space Administration, Washington, D.C.
J. P. Unik	Argonne National Laboratory, Argonne, Illinois
W. C. Wolkenhauer	Washington Public Power Supply System, Richland, Washington

REFERENCES

[1] Raman, S., Review Paper A1.

[2] Claiborne, H. C., Neutron-Induced Transmutation of High-Level Radioactive Wastes, ORNL Report TM-3964 (1972); Effect of Actinide Removal on the Long-Term Hazard of High-Level Waste, ORNL Report TM-4724(1975).

[3] Schneider, K. J. and Platt, A. M., High-Level Radioactive Waste Management Alternatives, BNWL Report 1900 (1974).

[4] Raman, S., Nestor,Jr., C. W., and Dabbs, J. W. T., The ^{233}U-^{232}Th Reactor as a Burner for Actinide Wastes, to be published in the Proceedings of the Conference on Nuclear Cross Sections and Technology, Washington, D.C., March 1975.

[5] Breen, R., Elimination of Actinides with LMFBR Recycle, Westinghouse Advanced Reactors Division, private communication.

[6] Beaman, S. L., Actinide Recycle Evaluations, General Electric Energy Systems and Technology Division, private communication.

[7] Kubo, A. S. and Rose, D. J., On Disposal of Nuclear Waste, Science 182 4118 (1973) 1205.

[8] Binney, S. E. and Spinrad, B. I., Oregon State University, private communication.

[9] Paternoster, R., Ohanian, M. J., Schneider, R. T. (University of Florida) and Thom, K. (National Aeronautic and Space Administration), Nuclear Waste Disposal Utilizing a Gaseous Core Reactor, private communication.

[10] Wolkenhauer, W. C., Leonard, Jr., B. R., and Gore, B. F., Transmutation of High-Level Radioactive Waste with a Controlled Thermonuclear Reactor, BNWL Report 1772 (1973).

[11] Draper, Jr., E. L. and Parish, T. A., University of Texas at Austin, private communication.

[12] Rose, R. P., Olhoeft, J. E., Kellman, S., et al, Westinghouse Fusion Power Systems Department, private communication.

[13] Benjamin, R. W., Survey of Experimentally Determined Neutron Cross Sections of the Actinides, DP Report 1324 (1973). See also Review Paper B1. This meeting.

[14] Moore, M. S., Review Paper B3. This meeting.

[15] Fields, P. R., Fried, S., Friedman, A. M., and Unik, J. P., Argonne National Laboratory, private communication.

[16] Barr, D. W., Los Alamos Scientific Laboratory, private communication.

[17] Browne, J. C., Lawrence Livermore Laboratory, private communication.

[18] Behrens, J. W., Carlson, G., and Bauer, R. W., Neutron-Induced Fission Cross Sections of ^{233}U, ^{234}U, ^{236}U and ^{238}U with Respect to ^{235}U, to be published in the Proceedings of the Conference on Nuclear Cross Sections and Technology, Washington, D.C., March 1975.

[19] Weston, L. W. and Todd, J. H., Measurements of the Neutron Capture Cross Sections of the Actinides, to be published in the Proceedings of the Conference on Nuclear Cross Sections and Technology, Washington, D.C., March 1975.

[20] Wilhelmy, J. B., Britt, H. C., Gavron, A., Konecny, E., and Weber, J., (n,f) Cross Sections for Exotic Actinides, to be published in the Proceedings of the Conference on Nuclear Cross Sections and Technology, Washington, D.C., March 1975.

Review Paper No. A8

IMPORTANCE OF TRANSACTINIUM NUCLEAR DATA (TND) FOR FUEL ANALYSIS

R. Dierckx
Euratom-CCR, Ispra, Italy

Abstract

Different methods of non-destructive fuel analysis are briefly mentioned, and Transactinium isotope nuclear data requirements to obtain the necessary precision are studied. Destructive fuel analysis is discussed, and its use and importance for several applications are indicated. The Transactinium isotope nuclear data and their importance for destructive fuel analysis are studied.

1. Introduction

Fuel analysis is a means to get information and knowledge on fuel composition and absolute fuel content of fuel materials. The fuel composition is directly used as such or can be further analysed. Non-destructive and destructive methods are used for these measurements, and both fresh and spent fuel, have to be considered. The fuel material investigated is present as final product in the form of fuel elements for reactors, in waste barrels, and as an intermediate product during the transformation phase in fabrication or reprocessing plants. In this review fuel analysis and the required TND will be studied from three points of view:

- the measurement of small waste residues,
- measurements for safeguard purposes, and
- measurements for testing of computer codes used for the fuel management of power reactors.

2. Non-Destructive Fuel Analysis

2.1. Non-destructive methods

The following non-destructive methods are applied for safeguard-purposes, scrap and waste measurements (1,2,3,4).

2.1.1. Calorimetry (3,5)

The quantity of heat generated by radioactive decay is measured. The main contributions come from α-decay, a small correction has to be applied for β-decay.

This method is useful for fresh Pu fuel of which the atomic composition is known. A correction for the ^{241}Am content and its formation during the storage time has to be applied. The method permits absolute measurements, as well as relative ones.

In order to convert mW/g to number of emitting nuclei, the TND needed are α- and β-decay schemes, α- and β-energies, number of α's emitted and half-lives.

2.1.2. Passive gamma-ray techniques

Most transactinium nuclei decay by emitting γ-rays. Table I taken from ref. (1) summarizes the most important nuclei, their half-lives, their γ-energies, and their branching ratios. The γ-rays are measured by NaI, Ge(li) or some other detectors, complemented by Compton-suppression techniques. The counting of natural activity is applicable to fresh fuel and waste control from fabrication plants. It is applied on enriched ^{235}U fuel elements, pellets, HTGR fuel, Pu fuel and Pu scraps (6,7). A drawback of this method is the γ-absorption in the samples which requires an additional measurement using external γ-sources.

Normally, measurements are done relative to standards, such that the required TND are of minor importance, and are known in each case with sufficient accuracy. The measurements of Pu fuel by passive gamma-ray counting are able to determine the isotopic composition and absolute fuel content. The errors introduced by the γ absorption in the fuel material are eliminated by counting the different γ-lines and correlating them. (Dr. Fudge, AERE Harwell, is working on this, and to apply this method, the accuracy of the γ-branching ratios needs improvement). Here no standards are used. TND accuracy is sufficient because the errors of the other parameters associated with the interpretation of the measurement are of major importance.

2.1.3. Neutron techniques

The non-destructive testing techniques using neutrons are either passive or active. In the passive methods the neutron producing reactions are spontaneous fission or (α,n) reactions. The neutrons are counted as fast neutrons, or after moderation as slow neutrons. Use is made of the correlation technique.

In the active methods a neutron source activates the the fissile material and the produced neutrons or gammas are measured. Following methods are applied:

- prompt fission gamma counting
- prompt fission neutron counting
- delayed neutron counting

- delayed fission gamma counting
- photofission neutron counting
- neutron capture γ-spectroscopy
- neutron transmission
- reactivity measurements.

However for all measurements with these techniques, the precision and accuracy depend on proper calibration with standards (1), and on the similarity of the geometry and composition of the fuel material to be measured. Only relative measurements are performed so that TND are not required. Existing TND are precise enough for the development work and the setting up of instrumentation.

2.2. TND accuracy requirements for non-destructive fuel analysis (Table II)

In general, for γ-ray and neutron counting methods, TND are of minor importance. They serve only for the development of the technique, and for an initial estimation whether the method will work, and for the setting up of the apparatus. The measurements are done relative to standards and TND are only needed for correcting the results. Up to now, all these methods are not yet sufficiently standardised and are not operated on an industrial scale. Absolute measurements are required only for low abundance of transactinium isotopes, or for little known nuclides, amounts of which, standardised by other methods are usually not available.

Each measurement and the applied corrections have to be adapted to the measured fuel compound. At the moment no information is available for the required accuracy of TND utilized for these corrections. In the future this information may become available.

For the calorimetry method, which is in principle an absolute method, TND are necessary. According to the model of C. Weitkamp and al. (8,9), TND needs and accuracies have been determined (see Table II). The accuracy of the power measurements in calorimetry lies between 0,25 % and 0,5 %. The specific power output per isotope is either measured directly or calculated from the α-energies (and β-energies) and the half-lives. As the α-energies are specified to a high degree of accuracy, the errors in the half-lives are dominant. Most important is the half-life of ^{238}Pu. As Weitkamp states "a critical evaluation of these data is highly desirable". In order to reduce the error due to the specific power output to values of the order of the measurement error (0.25 %), half-life values as well as directly measured specific power output values have to be improved. The error on the measured isotopic ^{238}Pu content, which should be known to about 0.5 % instead of actual value of 1-2 % has a great effect on the overall accuracy of the calorimetric method. The error introduced by the decay correction and build-up of ^{241}Am for the time elapsed between isotopic composition determination and calorimetric measurement is irrelevant as these time intervals are normally of the order of months.

The required TND and their associated accuracies are listed for non-destructive fuel analyses in Table II.

3. Destructive Fuel Analysis

3.1. Use of destructive fuel analysis results

Results of destructive fuel analysis are widely used for waste analysis, safeguard control, isotopic correlation studies, burn-up measurements, testing of codes for fuel management of reactors, and in reprocessing plants.

The results are either used directly or are further analysed. In destructive methods a high accuracy is achieved by absolute measurements, but they are expensive and require well-equipped laboratories and trained personnel.

3.1.1. Use for safeguard purposes (1)

Destructive analysis is used for safeguard applications where fuel is present in a free form as in reprocessing and fabrication plants. Results of non-destructive fuel analysis methods are compared with results of destructive methods in their development and testing phases. Standards and reference samples for non-destructive methods are extensively measured by destructive methods.

3.1.2. Isotopic correlations (1,10)

Destructive fuel analysis results are applied in safeguard applications to the isotopic correlation technique. The correlation technique is used to determine amounts of non-measurable isotopes in the fuel, from measurements of other isotopes using established correlations. This technique is also used to test the consistency of the results of destructive fuel analysis. Isotopic data are obtained from routine measurements at the reprocessing plant input. The following correlations are of main interest:

- Pu/U mass ratio versus ^{235}U depletion
- $^{236}U/^{238}U$ ratio versus $^{235}U/^{238}U$ ratio
- Pu/U mass ratio versus ^{239}Pu. $^{242}Pu/(^{240}Pu)^2$
- ^{240}Pu versus $^{241}Pu/^{242}Pu$.

The work of Umezawa (11), which reports correlation between Am and Cm isotopes versus ^{238}U, ^{242}Am and ^{242}Cm, is of special interest.

3.1.3. Burn-up measurements

The burn-up is determined from the measured isotopic composition of spent fuel. The main contributors to fission are the normal fissile isotopes, not under consideration here. For Pu fueled fast reactors ^{241}Pu contributes about 20 % (12), other transactinium isotopes contribute only a few percent.

3.1.4. Use in fuel management

Destructive methods are widely used in the determination of fuel composition of spent fuel from nuclear reactors as a function of burn-up. Resulting isotopic compositions are compared with results of theoretical calculations for given reactor operating conditions and time history. The goal is to test the nuclear computer codes used in the fuel management of reactors.

In a first step the measured isotopic compositions are compared with the corresponding calculated values. This procedure is observed at the TRINO Vercellesse reactor project (13), the Yankee Rowe project (14), and the Saxton plutonium project (15). Similar data for other reactor fuel exist but have restricted circulation. In the Saxton project destructive fuel analysis is applied using microsamples to compare theoretical and measured isotopic radial distributions.

In a second step the isotopic compositions of specially chosen spent fuel samples are used as input in an Inverse Burn-up-Code (16,17). The samples must be irradiated in the same nuclear environment and must cover as homogeneous a wide range of burn-up as possible. Under these conditions the Inverse Burn-up-Code delivers one-group cross section ratios, averaged over the sample volume and irradiation time. These ratios are then compared with the calculated one-group cross section ratios. For samples irradiated under these conditions, the burn-up is determined independently of theoretically calculated σ_c/σ_f ratios. These ratios are directly determined by the Inverse Burn-up-Code. The neutron spectrum in which the samples were irradiated, can then be determined from these one-group cross section ratios by unfolding codes (18). The obtained neutron spectrum and the one-group cross section ratios form an additional set of elaborated results to test fuel management codes.

A further use (although at this moment disregarded by the reactor operators) is the prediction of actinide waste production and actinide inventory calculations. The actinide waste prediction will gain in importance in the future with more nuclear power stations in operation. For this reason actinide inventory calculations and the comparison of theoretical results with experimental data become more and more important. In the Saxton project, one of the objectives is described as (19): "Obtain nuclear data of interest to plutonium recycling, especially with regard to depletion and generation of transuranic isotopes".

The one-group cross section ratio obtained from the Inverse Burn-up-Code help predict actinide waste production and serve indirectly as input for inventory calculations.

3.2. TND required in fuel analysis

Table III summarizes the TND required in fuel analysis.

Raw measured data in destructive fuel analysis are obtained by chemical methods, mass spectrometry, and α-activity measurements. The raw data are first reduced to yield isotopic compositions, and then further analysed in terms of one-group cross section ratios and neutron spectrum.

3.2.1. First reduction of raw data

3.2.1.1. Chemical methods

The chemical methods yield mass ratios. Isotopic ratios are obtained knowing the isotopic abundances and the atomic masses. Since the isotopic composition is by far the largest source of error, the accuracy of the atomic mass as agreed by IUPAC is adequate.

3.2.1.2. Mass spectrometry

This method yields directly isotopic ratios. The precision on mass-spectrometric results goes from 0,2 % to 3 %, and depends on the amount of isotope present. The precision is worse when only very small amounts are present (13,20).

3.2.1.3. α-activity measurements

In α-activity measurements the measured number of α-decays are transformed by means of half-lives into isotopic ratios. The precision of α-measurements is better than 1 %. The isotopic ratio ($^{240}Pu/^{239}Pu$) used in the data reduction is known to 0.5 %. For this reason the half-lives have to be known to better than 0.5 %, otherwise the error on the half-lives determines the measurement accuracy.

3.2.2. Decay correction

Decay corrections have to be applied to some isotopes to account for the time between reactor shut-down and measurement. Of special interest are ^{241}Pu and ^{242}Cm. In order to have an error of about 1 % for the decay correction, over a period of four years, the ^{241}Pu half-life has to be known to 2 % and the ^{242}Cm half-life to 0.1 %.

3.2.3. Burn-up determination

For uranium and plutonium fuel used at present only the actinide ^{241}Pu excluding the normal fissile isotopes has a certain importance in burn-up determination. For this isotope the fission and capture cross-section are estimated to require an accuracy of 10 % in the sensitive energy range.

3.2.4. Analysis in terms of one-group cross section ratios

This analysis requires some TND half-lives, decay schemes (^{242}Am and ^{236}Np) and certain cross sections. The cross sections are the absorption cross sections of the last isotope of the chains, which are used as one-group cross sections. It is estimated that their differential curve has to be known to 20 % in the sensitive energy range.

3.2.5. Neutron spectrum determination

Some knowledge is obtained about the differential neutron spectrum using unfolding codes and one-group cross section ratios. For this purpose differential cross sections are required with a precision of 10 % in the sensitive range.

3.2.6. Actinide inventory calculations

Several codes are used (ORIGEN (USA), ISOTEX, ACTIN (EURATOM), HYLAS ...) in inventory calculations. Sensitivity studies on TND will be started in the near future. At present time no requirements for the TND accuracy can be established.

Acknowledgements

The author is indebted to many contributors for exchange of information: Dr. G. Birkhoff, Dr. M. Bresesti, Dr. M. Cuypers, Dr. E. Schmidt, Dr. A Sola and many others.

Specially the author thanks Dr. A.J. Fudge of AERE, Harwell, for his formal written contribution, and Dr. W. Hage for valuable discussions.

References

(1) Practical Applications of R & D in the Field of Safeguards. Proceedings of a Symposium sponsored by ESARDA. Rome, 7/8 March, 1974.

(2) Non-Destructive Assay of Special Nuclear Material Contained in Scrap and Waste. USAEC regulatory guide 5.11, Oct. 1973.

(3) N.S. Beyer et al., Four Passive Assay Techniques Applied to Mixed Oxide Fuel. ANL 7906, Jan. 1972.

(4) INMM journal, vol. III, no. III, 1974. Proceedings of the 15th Annual Meeting, June 19-21, 1974.

(5) F.A. O'Hara et al., "Calorimetry for Safeguards Purposes", MLM 1798/TID 4500/UC-15, Jan. 1972.

(6) J.E. Cline, A Relatively Simple and Precise Technique for the Assay of Pu Waste. ANCR 1055, 1972.

(7) J.E. Cline, E.B. McSchmidt, A Technique for Assay of L-10 Bottles of Pu Nitrate-II. ANCR 1086, 1972.

(8) C. Weitkamp et al., "Accuracy of Calorimetric Pu Determination", KFK 1299, Sept. 1971.

(9) C. Weitkamp et al., "The role of nuclear data in nuclear materials safeguards", Nucl. data in Sc. & Tech., Vol. I, IAEA, Vienna 1973.

(10) J. Koch et al., A new tool in fuel management. Trans. Am. Nucl. Soc., v. 20, p. 339, 1975.

(11) H. Umezawa et al., Correlations of the amounts of the trans-plutonium nuclides with burn-up in the JPDR 1 spent fuel. J. Nucl. Sci. Techn. 10, no. 8, 1973.

(12) W.J. Maeck, Fission product nuclear data requirements for the determination of nuclear fuel burn-up, a Review. IAEA 169, vol. I, p. 163, Vienna 1973.

(13) A.M. Moncassoli-Tosi et al., Post Irradiation Burn-up Analysis of Trino Vercellese Reactor, Reactor burn-up Physics, p. 197, IAEA Vienna 1973.

(14) R.J. Nodvik, WCAP 6086, August 1969.

(15) H.K. Crain, WCAP 3385-37, December 1973.

(16) R.P. Matsen, The determination of ratios of effective cross-sections from measured burn-up data for Yankee Rowe. Nucl. Tech. vol. 15, p. 343, Sept. 1972.

(17) R. Dierckx, One-group cross section determination from measured fuel composition of irradiated fuel, to be published.

(18) R. Dierckx, V. Sangiust, Unfolding codes for neutron spectra evaluation, possibilities and limitations. 1° ASTM EURATOM Internat. Symp. on Reactor Dosimetry, Petten, 22-26 Sept. 1975.

(19) W.R. Smalley, Evaluation of Saxton core III, Fuel Materials Performance. WCAP 3385-57, July 1974.

(20) R.J. Nodvik, Saxton core-II, Fuel Performance Evaluation, WCAP 3385-56, part II, July 1970.

(21) R. Vaninbroukx, The half-lives of some long lived actinides, a compilation. EUR 5194.e, 1974.

(22) Nuclear Data Sheets-B, vol. 4-6, Sept. 1970.

(23) Nuclear Data Sheets-B, vol. 6-6, Dec. 1971.

List of Tables

Table I - Most important gamma rays emitted by the fissionable nuclides and their daughters

Nuclide	Half-life	Gamma ray energy (keV)	Gamma ray intensity (gamma for 100 decays)
^{235}U	7.13×10^{8} y	143.8	9.72
		163.4	4.59
		(*) 185.7	54.00
		205.3	5.00
^{234m}Pa (daughter) of ^{238}U h.l.4.49 x x 10^{9} y)	1.175 m	743.0	0.057
		766.6	0.206
		(*)1001.3	0.60
^{238}Pu	87.8 y	(*) 152.8	1.01×10^{-3}
^{239}Pu	2.44×10^{4} y	(*) 129.3	6.20×10^{-3}
		375.0	1.58×10^{-3}
		(*) 413.7	1.51×10^{-3}
^{240}Pu	6524 y	104.2	7.00×10^{-3}
		(*) 160.3	4.20×10^{-4}
^{241}Pu	15.16 y	(*) 148.6	1.90×10^{-4}
^{241}Am (^{241}Pu daughter)	433 y	(*) 59.5	35.90
		99.0	2.02×10^{-2}
		103.0	1.95×10^{-2}
		208.0	7.60×10^{-4}
		335.4	4.70×10^{-4}
		662.4	3.46×10^{-4}
		721.9	1.85×10^{-4}
^{237}U (^{241}Pu daughter	6.75 d	164.6	2
		(*) 208.0	23.2
		267.5	0.83
		332.3	1.3
^{212}Pb	10.64 h	(*) 238.6	47.00
^{208}Tl (daughters of ^{232}Th h.l. 1.41 x x 10^{10} y)	3.10 m	510.7	23.00
		(*) 583.2	86.00
		860.5	12.00
		2614.6	100.00

(*) Gamma ray most useful for measuring

Table II - TND required for non-destructive fuel analysis

TND required	mean value ± error			ref.	required accuracy	motivation
calorimetry						
α(β)-energies and branching ratios	accuracy 0.1%			22 23	sufficient	Calculation of specific power output
half lives	(yr)					
^{238}Pu	(87.8±0.8)			21	0.5%	Error due to specific power output smaller than measur. error
^{239}Pu	$(2.43\pm0.025)10^4$			21	0.5%	"
^{240}Pu	(6.55±0.07)			21	1%	"
^{242}Pu	$(3.87\pm0.05)10^5$			21	sufficient	"
^{241}Am	(432 ± 4)			21	1%	"
specific power(mW/g)	directly measured	calculated out of Eα and T 1/2	Diff. %			
^{239}Pu	1.914± 0.01	1.897	0.88	8	0.5%	"
^{240}Pu	7.1046±0.015	6.935	2.4	8	1%	"
^{241}Pu	3.62±0.18	-	-	8	sufficient	"
^{241}Am	114.5±0.17	112.3	1.9	8	1%	"
passive γ-ray techniques γ-ray energies and branching ratios half lives	-		-		sufficient	relative measurements only; data necessary for corrections and development
neutron techniques spontaneous fission data, delayed neutron data, capture cross-sections etc.	-		-		sufficient	"

Table III - TND required for destructive fuel analysis

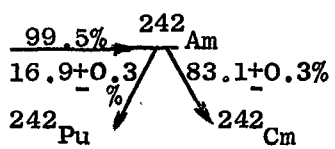
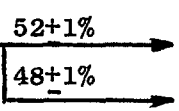
TND required	mean value ± error	minimum and max. value	ref.	required accuracy	motivation
atomic masses	-	-	-	sufficient	transformation of weight ratios in atomic ratios
half lives	(yr)				
^{239}Pu	$(2.430 \pm 0.025)10^4$	2.411-2.441	21	0.5%	transform. of α-activ. in atom. ratios
^{240}Pu	$(6.55 \pm 0.07)10^3$	6.24 -6.76	21	0.5%	"
^{238}Pu	(87.8 ± 0.8)	86.4 -87.8	21	0.5%	"
^{241}Pu	(14.5 ± 0.5)	13.8 -15.1	21	2%	correction for decay over 4 years
^{241}Am	(432 ± 4)	426 -458	21	2%	transform. of α-activ. in atom. ratios
$^{242*}Am$	(152 ± 7)	-	22	sufficient	corr. of analys. cross-sect. ratio
^{242}Cm	163.0 d	162.5-164.4	22	(0.5%)	transformation of α-activity in atomic ratios
				0.1%	correction for decay over 4 years
^{244}Cm	17.85	17.59-18.099	23	0.5%	transformation of α-activity in atomic ratios
				(2%)	correction for decay
decay schemes					
$^{242*}Am$		-	22	sufficient	cross-section ratio analysis
^{236}Np		-	22	sufficient	"

Table III - contin.

Cross-sections	needed as one-group-cross-section $\bar{\sigma} = \int_0^\infty \sigma \phi \, dE$	20% in sensitive range Threshold reactions: from thresh.+4-5 Mev others: from thermal to 2-3 Mev (thermal react. and fast reactor spectra	correction terms in cross-section ratio analysis
σ_c^{52*} σ_a^{64} σ_{2n}^{28} σ_a^{48} σ_a^{46}			
σ_a^{40}, (σ_c^{40}) σ_a^{41}, σ_c^{41} σ_a^{42}, (σ_c^{42}) $\sigma_{a,}^{51}$, $\sigma_c^{51}(n\gamma)^{242*}Am$ $\sigma_c^{51*}(n\gamma)^{242}Am$ σ_a^{52*} σ_a^{53}, (σ_c^{53}) σ_a^{26}, (σ_e^{26}) σ_a^{37}, (σ_c^{37}), σ_{2n}^{37} σ_a^{24}, (σ_c^{24}) σ_{2n}^{28}		10% in sensitive range	spectrum determination with unfolding codes out of cross-section ratios.
^{241}Pu fiss.-cross-section absorpt.cross-sect.		10% in sensit.range	burn-up determination

Review Paper No. A9

Radiation and Energy Sources, Tracer Techniques, Applications in Life Sciences, Agriculture and Industry

A.H.W. Aten Jr.*
Subfaculty of Chemistry
University of Amsterdam
Amsterdam, Netherlands

Abstract

Assessment of the uses of actinide nuclides in non-reactor application fields is presented. An evaluation of the needs for transactinium isotope nuclear data is made, and the requirements for nuclear data, particularly nuclear decay data, and their associated accuracies are tabulated.

This review will deal with applications of actinide nuclides in the following fields:

- Agriculture
- Industry
- Radiation sources
- Hydrology
- Medicine (tracers)
- Biology (tracers)
- Chemistry (tracers)
- Chemistry (activation analysis)
- Chemistry (X-ray fluorescence analysis)
- Physics (tracers)
- Production of actinide nuclides

* Correspondence: Professor Dr. A.H.W. Aten Jr.
Instituut voor Kernphysisch Onderzoek
Oosterringdijk 18, Amsterdam/Netherlands

Health physics (dosimetry)
Radiation chemistry (dosimetry)
Radiation damage (dosimetry)
Medicine (dosimetry)

Scientific research
Nuclear standards
Neutron fluence measurements

Among the quantities of the actinides the following will be considered important:

Alpha-energy
Alpha branching-ratio

Half-life

X-ray and gamma-energy
Photon to alpha ratio

Average beta-energy

Neutron to alpha ratio (^{252}Cf)
Gamma-energy per neutron (^{252}Cf)

Fission yields

ν_t
Delayed neutron number

Fission neutron energy spectrum
Cross-sections

I. Applications in agriculture will presumably always be tracer investigations, and this will also be true for the majority of the applications in industry, medicine, hydrology, biology, chemistry and physics. Results of measurements by means of tracer methods are not normally dependent on exact information about half-lives or decay schemes, because only comparisons between different fractions of the same sample of starting material are involved.
One medical application, which is not strictly a tracer experiment, is the determination of body burdens of actinides by external counting, normally in a whole-body counter. Such determinations, however, are not made on an absolute basis either, as the response obtained on the patient is compared to that observed when a known amount of the same isotope is counted

in a phantom. (As such samples are normally calibrated by alpha-counting and the maximum permissible body burden is expressed in units of alpha-activity, exact knowledge of the decay scheme of the nuclide is not required.)
An industrial application, which might also be listed under "radiation sources",is the elimination of electrostatic charges by alpha-radiation. In modern devices of this kind the alpha-active material is always covered by a thin sheet of inactive material. Under these circumstances the alpha-spectrum is so badly deformed and the fraction of the total number of alpha-particles which emerges from the surface is so uncertain, that an accurate knowledge of the decay characteristics of the nuclide will do little good.
It is by no means our intention to suggest that for the applications mentioned, no information in the field of TND is required at all. In any case two aspects must be judged on the basis of such data:

1. If the nuclide is detected by means of gamma- or X-ray counting, the number of photons emitted per decay determines the efficiency and the statistical error with which such observations are made. This information is needed to allow the experimenter to decide how much activity must be used in the investigation. It will be evident that only a very rough knowledge of nuclear data is required for this purpose.

2. If shielding is necessary in the preparation or in performing the experiment, information concerning gamma-, X-ray and neutron emission is needed to decide on the material,shape and thickness of the protective barrier. (These data are required not only for the original nuclide under consideration, but also for possible decay or fission products present in the actinide preparation.) However, problems of this kind are complicated by build-up phenomena, and, as the build-up factors are mostly not well-known, it would not serve a good purpose to insist on very accurate decay scheme data. In this connection it should be mentioned that even shielding against conversion electrons may sometimes constitute a problem, e.g. when an α-emitter like ^{242}Cm or ^{244}Am is used as a radiation source for X-ray fluorescence spectrometry. In this case some knowledge of intensity and energy of the conversion electrons may be desirable.

A third purpose for which TND are sometimes needed, is testing the purity of actinide samples by measuring alpha-spectra by means of semi-conductor detectors or in very special cases by gridded ionization chambers. For this purpose one does not need a better accuracy in the alpha-energy than what is necessary to distinguish one nuclide from another, i.e. about 15 keV.
A similar situation obtains if one wishes to determine the amount of a certain actinide by X-ray or gamma-spectrometry. If the possibility of contamination by non-actinide nuclides must be envisaged an accuracy of about 1 keV in the photon energy is required. If actinides only are present a lesser accuracy will be sufficient ($\sim$ 5 keV).
In connection with fluorescence X-ray analysis one might possibly think of a very special case when a gamma- or X-ray energy would be a critical quantity in planning such measurements. However, the energies of the more important photons emitted by actinide nuclides are known with sufficient accuracy for this purpose.
Cross-sections (mainly for neutrons, but to a lesser extent also for charged particles) are of interest for two purposes: production of actinide nuclides and activation analysis. (Although bremsstrahlen irradiation is also used for these purposes photon cross-sections present no interest in this connection due to the complicated nature of the spectrum.)
For planning the production of actinide nuclides approximate cross-section values are sufficient, the more so, because such a production normally takes place in a neutron flux, which is partly thermal, and partly of intermediate energy, with a strong contribution of fast neutrons.
A certain interest exists in the preparation of very heavy actinides (A > 244). For this purpose σ-values for neutrons from thermal energy up to 10 keV are asked with accuracies of about 10 percent. An even more important case in this category seems to be the production of ^{238}Pu, both by the $^{237}Np(n,\gamma)$ reaction and the decay of ^{242}Cm. For the use in pacemakers isotopic purity is important, which means that the σ-values for the reactions $^{237}Np(n,\gamma)^{238}Np$; $^{238}Pu(n,\gamma)^{239}Pu$ and $^{237}Np(n,2n)^{236}Np$ are of special interest, but also in this case one would hardly derive a profit from anything better than approximate values.
In activation analysis the knowledge of σ-values is useful in the first place in choosing the irradiation conditions, but it

is not used as a basis for calculating the quantity of the element or nuclide to be determined. (This must always be done by the activation of standards.) It also serves to decide whether the activity observed might have been produced from other nuclides present. This is specially important if the activity of one or more fission products is measured to determine a fissile nuclide. It is evident that only rough data are needed in this connection, but a certain amount of information concerning yields in the fission of the element under study and of possible disturbing actinides is also needed.
A special case of activation analysis by the measurement of fission products is the observation of delayed neutron emission. For the most important nuclides sufficient data are available to judge the conditions of activation and the dangers of interference, for other actinides reliable estimates can be made on the basis of delayed neutron emission systematics [1]. (It is once more assumed that the analysis is based on comparison with a standard containing a known quantity of the nuclide to be determined. If this is not possible, and if an absolute measurement must be made, a maximum error of $\sim$ 5 percent may be suggested for the number of delayed neutrons per fission.)

From the preceding remarks it appears that for the purposes listed under I some information in the field of TND is needed, but that these data need not be of a very high accuracy. In how far the requirements are fulfilled, will presumably become apparent from the review papers B6 and B7. It would seem likely that to satisfy these demands sufficient information has been published concerning most transactinium nuclides, but that in some cases more information may be needed about L-ray intensities and that almost certainly our knowledge concerning the decay-scheme of ^{237}Pu is insufficient.

II. For health physics purposes a certain amount of nuclear data is also essential, not only concerning the nuclides, the administration of which is under consideration, but in a number of instances also concerning their decay products (Table I). This is specially the case for tri- and quadrivalent actinides, of which only an almost negligible fraction passes from the gastro-intestinal tract into the body circulation. Of the alpha-energy only one percent is supposed to be effective [2]. Many

decay products of long-lived actinides are beta-emitters, which frequently also produce gamma-rays. (In some cases one even has to take into account decay products, which are not themselves trans-actinium nuclides.) These daughter activities contribute to the body dose much more efficiently than the alpha-emitters. Attempts to take their dose-rate into account are complicated by the fact that instead of maximum beta-energies the determining quantity in this case is the average beta-energy, which often is not known from experiment. It then has to be calculated by means of equations, which are not always very accurate. Another serious aspect is that the chemical form of many decay products is that of mono- or divalent cations, an appreciable part of which passes through the wall of the gastro-intestinal tract. Even so, the biological data, like biological half-lives or fractions of nuclides administered going to various organs, are so uncertain and vague, that only on very rare occasions one could hope to obtain better estimates of maximum permissible concentrations or body burdens by using better nuclear data than at present available.

III. Nuclear data of a better class are required if the quantity of an actinide is determined by alpha-counting or alpha-spectrometry. If ordinary alpha-counting is used the essential information concerns the half-life. In many instances, e.g. in fissile material accounting, an accuracy of a fraction of a percent may be aimed at, which will require an accuracy in $T_{\frac{1}{2}}$ of about 0.1 percent. If the mass of the nuclide is determined by alpha-spectrometry measurements are likely to be somewhat less precise and one would probably be satisfied with an accuracy of 0.2 percent in the intensity of the standard alpha-ray. If one measures the most intense alpha-ray, and if, as is often the case, the nuclide has two main alpha-ray components, one of about 80 percent and one of about 20 percent of the total activity, the ratio in which the two are emitted should be known with an error of one percent. (In cases like that of ^{239}Pu, where there are more than two main alpha-rays, the required accuracy may be a little but not very different.) For the purpose of activity determinations by alpha-spectrometry the energy resolution need not normally be very fine. An accuracy of $\sim$ 100 keV in the alpha-energy would in most cases be sufficient.

In certain samples one may wish to determine an actinide nuclide by means of a gamma- or X-ray measurement. (This is the normal situation for ^{241}Am, the alpha-activity of which cannot be measured with a semi-conductor spectrometer in the presence of ^{238}Pu.) In this case the measurements will normally be made against a standard consisting of the same nuclide. If, however, an absolute measurement is made,the photon-to-alpha ratio need not be known with a better precision than can be reached in the calibration of the photon detector.
The problems of mass determinations will come up again in connection with the nuclear data for standards and research.

IV. Even more difficult problems may occur in connection with the use of ^{252}Cf-sources for local irradiations during an operation or by inserts made similar to radium needles or ^{198}Au seeds. To calculate dose-rates one would in principle have to know the neutron energy spectrum, the gamma-spectrum and the ratio of gamma-to-neutron emission. (If the calibration of the ^{252}Cf sources is not done by neutron measurement but on the basis of alpha-counting, one also needs the alpha-to-neutron ratio.) The absorbed dose due to the Cf-gamma-rays is not very different from that due to the neutrons. The relative biological efficiency of the photons is, however, appreciably smaller than that of the neutrons, which means that for the gamma-rays somewhat less exact data can be accepted than for the neutrons. For the shape of the gamma-spectrum one may be satisfied with a lesser accuracy than for the total gamma energy per neutron, because most of the californium gamma-rays have energies in the range where the energy absorption coefficient as a function of energy follows a fairly flat course. Even so, it will be extremely difficult to calculate accurate dose-rates in tissues as a function of distance - largely because of geometrical factors - and it may be preferable to measure dose-rates experimentally, although this too, is very difficult. (Perhaps these alternatives can be discussed in Karlsruhe.)
A problem closely related to that of the use of ^{252}Cf for therapeutic irradiations is that of activation in vivo with ^{252}Cf-sources. (The use of ^{252}Cf for this purpose has been suggested, but it is by no means certain that this nuclide makes the most suitable neutron sources for this purpose.) Calculating the total absorbed dose (or dose equivalent) given to a patient

during activation in vivo would require the same TND information as the determination of dose-rates for therapy purposes, but the required accuracy will be much less serious in the case of activation tests, because these are always performed with fairly small absorbed doses.
It should be kept in mind that the gamma-rays emitted by a ^{252}Cf sample are not only those produced during the fission process itself, but they are in part due to accumulated fission products. The total energy of these gamma-rays is about equal to that of the fission gamma-rays. Calculation of the dose-rate due to the fission product gamma-rays requires a knowledge of the fission yields in the spontaneous fission of ^{252}Cf, together with decay-scheme data of the fission products, but the last-mentioned information does not come under TND. (It should be pointed out, that, because all fission yields are < 10 percent, a large error in the yield or the specific gamma-ray constant of a single fission product, will make a negligible error in the total energy deposition from a californium source.)

V. In view of the arguments given above it would seem that an important part of the need for better Transactinium Nuclear Data would be found in the field of scientific applications. This should not be understood to mean data needed by scientists for better understanding of nuclear physics and neutron physics, even though a great deal of scientific information is still needed for this purpose. What is envisaged here is the use of actinides as aids in scientific measurements.
In this connection the following points come to mind:

1. Use of actinides to produce a known absorbed dose for purposes of radiation chemistry and radiation damage studies.

In principle both photons and α-particles can be used to produce energy deposition for radiation chemistry purposes. If, however, energy deposition by gamma-rays or X-rays from actinide sources is used, sources containing very strong alpha activities are required. In most laboratories there will exist strong objections to the handling of such sources, unless they are contained in thick-walled capsules. In this case the radiation emitted must be measured on the source in this condition, and no nuclear data are required apart from the photon spectrum. (Even this is likely to be deformed by the presence of secondary radiation.)

Alpha-particles, on the other hand, are of the greatest value for the study of the influence of radiation on various systems. The essential point is that they deposit their energy in a much more concentrated form than electrons generated in a medium by gamma-rays.

Alpha-activity can be used as an internal or as an external radiation source.

The first method is by far the simplest. Alpha-particles distributed over a sample of not too small a size deposit their entire energy in the medium. If the average alpha-energy is known with sufficient accuracy the error in the absorbed dose is determined by the error in the measurement of the total activity. (As the energies of the main alpha-rays emitted by an actinide nuclide differ very little, the branching ratio need not be known with a very good accuracy to calculate the average α-energy from the energies of the individual α-rays.)

The situation is much more difficult if an external α-source is used, and then the absorbed dose-rate must normally be determined by experiment. However, in some cases the dose-rate can be calculated from the activity of the radiation source, but in this case one does not only need to know the energies of the α-rays, but also ranges and stopping powers in various materials.

A special dosimetry problem is the activity calibration of alpha-emitters by calorimetry. As this complicated technique will only be used for measurements of the highest accuracy, one would expect the method to be useful only if the error in the average alpha-energy does not exceed about 0.01 percent. In normal cases, where more than one gamma-ray is present, this requires the energy of the main α-ray to be known with the same accuracy, that of the next abundant α-ray with an accuracy of about 0.03 percent and the intensity ratio of the two with an accuracy of about 1 percent.)

2. Use of actinides as standards for α,γ and X-ray intensities and energies.

Presumably actinides make the best standards for α-energies. For such standards it is essential that they can be obtained as strong samples of negligible thickness. It cannot be denied that these conditions are fulfilled very well by Th B + C, ^{210}Po and ^{226}Ra. For all of these nuclides the alpha-energies are known with very good accuracies - about as high as any at

present available. On the other hand Th B + C has the disadvantage of a very short half-life and ^{210}Po and ^{226}Ra tend to give rise to very bad contaminations, although in the case of ^{226}Ra these are partly of a temporary nature. Thus it may well be that certain actinide nuclides, with half-lives sufficiently short to allow the use of very thin samples, would make a better standard for α-energies than any of the nuclides mentioned before. It is often desirable to have a combination of such standards which covers the normal energy range with intervals of the order of 0.5 MeV. A few nuclides which are suitable as alpha-energy standards, are listed in Table II. Due to the nature of the alpha-emission process, it is unavoidable that emitters of low energy α-rays have long half-lives and corresponding low specific activities, which renders them less suitable for alpha-energy standards. (The data in Table II suggest that ionium, i.e. ^{230}Th,might be a useful standard. The possibility should be considered of making small quantities of this nuclide available to all interested laboratories for calibration purposes.) For the lowest alpha-energies the only actinides available as standards are ^{238}U and ^{232}Th. Due to the low specific activities their α-energies are not known with a really good accuracy. In actinide research standards for such energies are not needed, but they may be useful for measurements on alpha-emitters of lower atomic weight.

It seems uncertain whether actinides will be used as standards for gamma- or X-ray energies. So many excellent values are available for X-ray energies (K-rays and L-rays) of a number of elements in the range of 10 - 70 keV, that the use of actinides for this purpose is certainly not essential. On the other hand there are a number of very accurate photon energy data in existence for actinide nuclides which can easily be obtained by any interested laboratory, like ^{241}Am, ^{243}Am and ^{239}Np (in equilibrium with ^{243}Am).

An even more important application of actinide nuclides, is their use as intensity standards in the X-ray energy range. It is ^{241}Am which plays the essential part. The nuclide emits photons of various energies, the most intense rays being at 14 keV, 18 keV and 60 keV. The fact, that ^{241}Am, being an alpha-emitter, can be calibrated with a very high accuracy makes it superior as a standard for this purpose, to other nuclides.

The nuclide ^{243}Am is important for a special application. It is normally in equilibrium with ^{239}Np and it can therefore be used as a standard if ^{239}Np is measured by means of its gamma-emission. (The measurement of ^{239}Np is important in neutron capture cross-section determinations for ^{238}U.)

3. Half-lives of actinides.
As mentioned already these data are essential if a quantity of material is determined by alpha-counting. The knowledge of half-lives is also essential if the amount of ^{241}Pu in a plutonium sample is determined by observing the in-growth of ^{241}Am and in similar cases. If one considers that alpha-activities can be measured with an accuracy of the order of 0.1 percent, it follows that for scientific work of the highest half-lives must be known with an accuracy appreciably better than this figure.

4. Number of prompt neutrons emitted in fission.
Determinations of $\bar{\nu}_{total}$ and $\bar{\nu}_{prompt}$ are normally made relative to one or more standard $\bar{\nu}$-values. The most important one of these is $\bar{\nu}_{total}$ for the spontaneous fission of ^{252}Cf, but $\bar{\nu}$-values for the fission of ^{235}U, ^{233}U and ^{239}Pu by thermal neutrons are also used for this purpose.

5. Number of delayed neutrons emitted in fission.
This figure is required to derive $\bar{\nu}_{prompt}$ from $\bar{\nu}_{total}$ or vice versa. Because $\bar{\nu}_{delayed}$ is small compared to $\bar{\nu}_{total}$ the relative accuracy of $\bar{\nu}_{delayed}$ need not be very good. For the thermal fission of ^{235}U, ^{233}U and ^{239}Pu a number of different determinations have been published, but for the spontaneous fission of ^{252}Cf only a single measurement seems to have been made.

6. Fission neutron energy spectra.
If often occurs that activations or irradiations are done with fission neutrons. (In most cases the energy spectrum will be somewhat deformed.) For many purposes it is important to know the energy distribution of the neutrons. The two points of main interest are the spontaneous fission of ^{252}Cf and the fission of ^{235}U by thermal neutrons. Normally the energy distribution is represented by an equation of the type:

$$N(E) \text{ prop. } E^{\frac{1}{2}}.e^{-E/E_n},$$

where only E_n is determined. It should, however, be mentioned that most measurements cover only the energy range between about

0.5 MeV and 12 MeV. There are also some observations covering lower and higher energies, although some of these would have to be re-evaluated. It is of special interest to establish between which energy limits the equation is valid.

7. Fission yields.

The determination of fission yields is frequently done in such a way that two neutron irradiations are performed: one in the system to be investigated and one in a standard system, for which one normally takes the irradiation of ^{235}U with thermal neutrons. The same series of fission products is measured for the two systems and because the ratio in which the fission products are formed from ^{235}U is known - or supposed to be known - one can calculate the relative number of fission product atoms of the various masses produced in the systems under investigation. To use this method one needs an accurate knowledge of the fission yields for ^{235}U + thermal neutrons. Specially in the region of symmetric fission the best values available do not seem to be very good, as can be seen from the differences between the figures published by different investigators [4]. Because fission yields in the region of symmetric fission are very low for ^{235}U + thermal neutrons, it might be desirable to use spontaneous fission of ^{252}Cf as a standard system instead, but in this case the available data are even less complete [5].

8. Neutron fluence measurements.

Fission of actinide nuclides offers attractive possibilities as a detector for the measurement of neutron fluences, flux densities and energy spectra. The number of fission events taking place can easily be registered directly or by counting one or more fission products. Some actinide nuclides (^{233}U, ^{235}U and ^{239}Pu) are specially useful for the measurement of thermal and epithermal neutrons, whereas others (^{232}Th, ^{238}U and ^{237}Np) can be used as threshold detectors. In combination they serve to determine the shape of the energy spectrum of the fast neutrons. For this purpose an accuracy of 5 percent is normally asked for, but for fluence measurements a better accuracy is sometimes useful.

The author wishes to express his gratitude to the contributors and to the following gentlemen: A. McKay, J.C. Post, A. Spernol, R. Dierckx and J. Konijn, who gave valuable aid in the preparation of the text.

REFERENCES

[1] MANERO, F. and KONSHIN, V.A., Atom. Energy Rev. 10 (1972) 637

[2] ICRP Committee II, Health Physics 3 (1960) 20

[3] SCHMIDT, J.J., Nuclear Data for Reactors (Conference Helsinki 1970, IAEA, Vienna, 1970) I, 3

(4) CROUCH, E.A.C., Nuclear Data in Science and Technology (Symposium, Paris, 1973, IAEA, Vienna, 1973) I, 393

(5) VON GUNTEN, H.R., Actinides Reviews 1 (1969) 275

(6) RYTZ, A., Atomic Data and Nuclear Data Tables, 12 (1973) 479

(7) Report of Working Group No.I Table 2 (These proceedings)

(8) List of transactinium nuclear data requirements for energy and related applications (These proceedings)

(9) R. Vaninbrokx EUR 5194e

(10) R. Vaninbrokx CBNM/RN/11/75

(11) H. Ramthun and W. Müller, IJARI, 26 (1975) 589

(12) Report of Working Group 1, Table 3 (These proceedings)

(13) S.A. Baranov, A.G. Selenkov and V.M. Kulakov, Rev. Paper B6 (These proceedings)

(14) C.W. Reich, Rev. Paper B7 (These proceedings)

(15) Nuclear Data Sheets, 6 (1971) 637, 638

(16) D. de Jong, unpublished evaluation

(17) Estimated on the basis of data listed by E.A.C. Crouch, Nuclear Data in Science and Technology (IAEA, Vienna, 1973) I, 393

(18) W.E. Nervik, Phys. Rev. 119 (1960) 1685

(19) V.V. Verbinski, H. Weber and R.E. Sund, Phys. Rev. C7 (1973) 1173

(20) Estimated on the basis of data in A.T.G. Ferguson, Neutron Standard Reference Data (IAEA, Vienna, 1974) 333

(21) A.B. Smith. Prompt Fission Neutron Spectra (IAEA, Vienna, 1972) 3

Table I

Actinide nuclides in equilibrium with beta-emitters

$^{243}Am(\alpha)$	RdTh = $^{228}Th(2\ y,\alpha)$	$^{227}Ac(\beta)$
$^{239}Np(2\ d,\beta)$	ThX = $^{224}Ra(4\ d,\alpha)$	RdAc = $^{227}Th(19\ d,\alpha)$
-	Tn = $^{220}Rn(1\ min,\alpha)$	AcX = $^{223}Ra(11\ d,\alpha)$
$^{237}Np(\alpha)$	ThA = $^{216}Po(0.1\ s,\alpha)$	An = $^{219}Rn(4\ s,\alpha)$
$^{233}Pa(1\ month,\beta+\gamma)$	ThB = $^{212}Pb(10\ h,\beta+\gamma)$	AcA = $^{215}Po(short,\alpha)$
-	ThC = $^{212}Bi(1\ h,\alpha+\beta+\gamma)$	AcB = $^{211}Pb(36\ min,\beta+\gamma)$
$^{238}U(\alpha)$	ThC'' = $^{208}Tl(3\ min,\beta+\gamma)$	AcC = $^{211}Bi(2\ min,\alpha)$
UXI = $^{234}Th(1\ month,\beta)$	and	AcC'' = $^{207}Tl(5\ min,\beta)$
UX2 = $^{234}Pa(1\ min,\beta)$	ThC' = $^{212}Po(short,\alpha)$	-
-		$^{236}Pu(3\ y,\alpha)$
$^{235}U(\alpha)$		$^{232}U(72\ y,\alpha)$
UY = $^{231}Th(1\ d,\beta+\gamma)$		RdTh = $^{228}Th(2\ y,\alpha)$
		etc.

Table II

Alpha-emitters for alpha-energy calibration

Nuclide	Principal alpha-energy *) in MeV	α/mg.sec **)
^{252}Cf	6118.3	1.7×10^{7}
^{244}Cm	5804.96	2.3×10^{6}
^{238}Pu	5499.21	4.6×10^{5}
^{241}Am	5485.74	1.1×10^{5}
^{240}Pu	5168.30	6.3×10^{3}
$^{230}Th(Io)$	4687.5	5.8×10^{2}
^{238}U	4196	0.0095
^{232}Th	4012	0.0031

*) Most abundant α-ray [6]

**) Number of principal α's per mg of nuclide and second

Table III (corrected)

Estimated Required Accuracies

Quantity	Purpose	Estimated accuracy	
Alpha-energy	health physics	1	MeV
	energy deposition (dosimetry)	10	keV
	purity test	15	keV
	α-energy standard	1	keV
	calorimetric activity measurement	0.5	keV
Half-life	mass determination	0.1 to 0.2	%
	mass standard (by α-counting)	0.02	% *)
	mass standard (calorimetric)	0.02	% *)
Branching ratio main α-rays	mass determination (spectroscopic)	1	%
	calorimetric activity measurement	1	%
X-ray and γ-energies	health physics	30	%
	purity tests	5 (or 1)	keV
	photon energy standard (at 60 keV)	100(or 5)	eV **)
	photon energy standard (at 17 keV)	50(or 1)	eV **)
Photon-to-α ratio	health physics	30	%
	photon intensity standard	1	%
	mass determination	1	%
Average β-energy	health physics	30	%
Neutron-to-α ratio (if ^{252}Cf calibrated by α-count)	therapy dosimetry	1	%
	activation analysis in vivo (dosimetry)	20	%
Gamma-energy per neutron (^{252}Cf)	therapy dosimetry	5	%
	activation analysis in vivo (dosimtery)	50	%
Cross-sections	activation analysis	50	%
Cross-sections	production of heavy actinide nuclides	10	%
	production of ^{238}Pu	30	%
	neutron fluence measurements (fission)	2 to 5	%
Fission yields	comparison method (^{235}U+th.n., ^{252}Cf spont.)	2	%
	neutron fluence determination	2 to 5	% ***)
	activation analysis	50	%
	therapy dosimetry (^{252}Cf)	30	%
X-ray and γ-intensities	health physics	30	%

Quantity	Purpose	Estimated accuracy
	relation between $\bar{\nu}_p$ and $\bar{\nu}_t$	0.015 ****)
Delayed neutron numbers	activation analysis	50 %
	absolute activation analysis	5 %
$\bar{\nu}_t$ (^{252}Cf)	standard for fission neutron measurements	0.015 *****)
Fission neutron energy spectrum	standard spectrum (^{235}U+th.n., ^{252}Cf spont.)	0.01 MeV in E_n
	therapy dosimetry (^{252}Cf)	0.1 MeV in E_n

*) Low-geometry alpha-counting and calorimetric assay can both be done with an accuracy of ≈ 0.03 percent. This requires an accuracy in the half-life of 0.02 percent or better.

**) The best accuracy is needed for measurements on mesonic X-rays.

***) Only for a single or a few fission products.

****) From the data in (1) it seems that the data for ν_t for the thermal neutron fission of ^{235}U, ^{233}U and ^{239}Pu are sufficiently accurate to warrant a request for this precision. The table for delayed neutron numbers in (1) suggests that this precision has not yet been reached. For ^{252}Cf only one determination seems to have been made of $\nu_{delayed}$.

*****) cf (3).

Table IV Present Accuracies in Non-neutron Nuclear Data

	E_α [6]	$T_{1/2}$ [8]	$T_{1/2}$	$I_{\alpha_1}/I_{\alpha_2}$ [7,8,13]	E_γ [14]	I_γ [14]	I_X [8]
	keV	%	%		keV	%	
^{228}Th(RdTh)	0.2		0.05 [14]	1.5	0.1	0.2	
^{230}Th(Io)	1.5		3 [14]	0.5	0.03		
^{232}Th	5		0.5 [19]				
^{234}Th(UX1)			0.1 [14]				
^{231}Pa	1		0.3 [14]	2	0.06		
^{234}Pa(UX2)			0.4 [14]		0.02		
^{232}U	0.1		3 [9]	1			
^{233}U	1		0.2 [9]	0.5			
^{234}U	1		0.3 [9]	3			
^{235}U	2	2	0.3 [9]	10	0.01		
^{236}U	3	2	1 [9]	2			
^{238}U	4		0.2 [9]	5			
^{237}Np	1		0.5 [14]	2	0.1	10	
^{239}Np			0.1 [14]		0.03	2	
^{236}Pu	1		0.3 [14]	1			
^{237}Pu			0.5 [14]		0.1		
^{238}Pu	0.2	1.5	1 [9]	0.1			
^{239}Pu	0.7	1	0.4 [10]	1	0.1		100
^{240}Pu	0.15	5	1 [9]	1			
^{241}Pu		5	0.4 [9]				
^{242}Pu	1	5	1.5 [9]	4			
^{241}Am	0.12		0.05 [11]	1	0.001 [15]	$2^{8,16}$	5
^{242}Am^m	5		5 [14]				
^{243}Am	1			0.6	0.03	0.5	
^{242}Cm	0.5	1	0.1 [12]	1.5			
^{244}Cm	0.05		1;0.06 [12]	0.8			
^{252}Cf	0.5	1	1 [9]	0.5			

Only gamma-rays of intensities useful for general applications are listed. The main gamma-line for ^{228}Th is that of ^{208}Tl(ThC[11]). For ^{243}Am the gamma-lines of ^{239}Np have not been listed.

The only actinide nuclide for which reasonable X-ray data are available is ^{241}Am.

For present accuracies for thermal $\sigma_{n,\gamma}$, thermal $\sigma_{n,f}$, $I_{n,\gamma}$ and $I_{n,f}$ the reader is referred to Ref. (7).

Table V. Present Accuracies in Delayed Neutron Numbers (1)

	Relative Accuracy (percent)		
	delayed neutrons per fission (thermal)	delayed neutrons per fission (fission neutrons)	delayed neutrons per fission (14 MeV)
^{232}Th		3.5	5
^{233}U	4.5	4	7
^{235}U	4	2.5	4.5
^{238}U		3	4
^{239}Pu	5	3	5
^{240}Pu		7	9
^{241}Pu	10		10
^{242}Pu		3	

Interpolation of delayed neutron numbers to intermediate energies should be done by linear interpolation of the logarithm between thermal energy and 14 MeV and not by a step function as suggested in ref. [1].

Table VI Present Accuracies in Fission Yields

	Percent of yield of F.P.	
	^{235}U + th.n. [17]	^{252}Cf spont [18]
^{95}Zr	1	
^{99}Mo	1	1
^{103}Rh	3	
^{105}Rh	4	
^{111}Ag	1	6
^{112}Pd	1	5
$^{115+115m}Cd$	2	6
^{132}Te	1	2
^{140}Ba	0.5	9
^{144}Ce	1	

The accuracies given for the fission of ^{252}Cf are those given by the author. These figures are based on fission yields for ^{235}U + th.n., they were obtained by the method mentioned in our text. Unfortunately the fission yields assumed for ^{235}U + th.n. are not readily acciessible. All in all a redetermination of the fission yields for ^{252}Cf would be most desirable.

Table VII Present Accuracies in Fission Data

	Neutron/α % relative	E_γ/neutron [19] % relative (above 0.14 MeV)	$\bar{\nu}_t$ [1] n per fission	E_n MeV
^{235}U+th.n.		5	0.007	0.05 [20]
^{252}Cf spont	1	4	0.015	0.05 [21]