



April 1976

KFK 2283

Institut für Neutronenphysik und Reaktortechnik

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

and

Results and Main Conclusions of the IAEA-Advisory Group Meeting on Transactinium Nuclear Data, held at Karlsruhe, November 1975

H. Küsters, M. Lalović



GESELLSCHAFT FÜR KERNFORSCHUNG M.B.H.

KARLSRUHE

# Als Manuskript vervielfältigt

Für diesen Bericht behalten wir uns alle Rechte vor

# GESELLSCHAFT FÜR KERNFORSCHUNG M.B.H. KARLSRUHE

KFK 2283

Institut für Neutronenphysik und Reaktortechnik

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

And

Results and Main Conclusions of the IAEA-Advisory Group Meeting on Transactinium Nuclear Data, held at Karlsruhe, November 1975

by

H. Küsters and M. Lalović

,

Gesellschaft für Kernforschung mbH, Karlsruhe

·

#### Summary

In this report a review is given on the actinium isotope build-up and decay in LWRs, LMFBRs and HTRs. The dependence of the corresponding physical aspects on reactor type, fuel cycle strategy, calculational methods and cross section uncertainties is discussed. Results from postirradiation analyses and from integral experiments in fast zero power assemblies are compared with theoretical predictions. Some sensitivity studies about the influence of actinium nuclear data uncertainties on the isotopic concentration, decay heat, and the radiation out-put in fuel and waste are presented. In a second part, the main results of the IAEA-Advisory Group Meeting on Transactinium Nuclear Data are summarized and discussed.

übersicht zum Aufbau und Zerfall von Transaktinidenisotopen in Reaktorbrennstoffen und Untersuchungen über den Einfluß von Wirkungsquerschnittsungenauigkeiten

sowie

Ergebnisse und Schlußfolgerungen der IAEA-Tagung über Kerndaten von Aktiniden, Karlsruhe, November 1975

#### Zusammenfassung

Dieser Bericht enthält eine Übersicht zum Aufbau und Zerfall von Actiniden in thermischen (LWR, HTR) und schnellen Reaktoren. Die Abhängigkeit der entsprechenden physikalischen Größen vom Reaktortyp, von der Art des Brennstoffmanagements, den Rechenverfahren und den Unsicherheiten der verwendeten nuklearen Daten wird analysiert. Experimentelle Ergebnisse von Nachbestrahlungsuntersuchungen und von integralen Experimenten in schnellen Nullenergieanordnungen werden mit theoretischen Vorhersagen werglichen. Einige Studien über die Abhängigkeit der Isotopenkonzentrationen, der Nachzerfallswärme und des Aktivitätsinventars im Brennstoff und im Abfall von Wirkungsquerschnittsunsicherheiten werden durchgeführt und die Ergebnisse diskutiert.

Im zweiten Teil werden die wichtigsten Ergebnisse der IAEA-Tagung über die Kerndaten von Actiniden zusammengestellt und diskutiert. IAEA-Advisory Group Meeting on Transactinium Nuclear Data Review Paper A3

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

#### by

# H. Küsters and M. Lalović

# KERNFORSCHUNGSZENTRUM KARLSRUHE Institut für Neutronenphysik und Reaktortechnik

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

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

an increasing reactor population in which use is made of recycled Pu /1/. Mainly the long lived  $\alpha$ -ermitters are of predominant concern because of the associated radiation hazard. New ideas were born to transmute the  $\alpha$ -ermitters 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 transactinide 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 -

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

3 -

- 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 transactinide area. This situation seems to be the main reason for complaints from users to data producers. 4 -

# 3. Methods used to determine fuel cycle aspects.

In order to be able to deduce requests for nuclear data of special transactinide 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<sub>1</sub> /7/. Whether a 1/Eweighting spectrum is used for collapsing to coarse energygroup mesh or a detailed many group system-spectrum, causes for 2000 MWd/t burn up about 5 % change in the effective  $\sigma_a$ between 1.1 eV and 6.5 eV; including mutual self-shielding of various resonance isotopes the differencies in  $\sigma_{a}$  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 O-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

<u>Tab.1:</u>	Production rates of Pu239, Pu240 and Pu24	1 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 chosing 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 experimental 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	<u>+</u> 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 <u>concluding</u> this section, the errors in predicting the transactinide concentrations in LWRs during burn up may turn out to be <u>20 % and even more</u> 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 interprete 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  $\alpha$ -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

- 8 -

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 <u>Tab. 3</u> /9/ (note, that the U236 concentration deviates by a factor of 7).

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	80					
Cm243						
Cm244	30 %					

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

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.

# 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 differencies 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 productions or the amount of actinides to be reprocessed yearly, for idealized reactor types with some fuel management scheme. This is so, because the differencies 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 <u>Table 4</u> by the yearly produced radioactivity in Ci. Again it can been seen that the differencies amount up to a factor of 10.

The main radioactivity stems from the  $\alpha$ -decay of Cm244, and Pu238 (5:1),  $\beta$ -decay of Pu241, Np239 and Am242 in ( $\approx$ 1200:11:1) and  $\gamma$  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

Isotope	Uranium-Fueled Water-Reactor		Uranium Water-R	-Plutonium Reactor	All Actinides Recycled	Fast Breeder Reactor		
	kg/y	Ci/y	kg/y	Ci/y	only fu kecycleu	kg/y	Ci/y	
Pu236	0.02	8.5.10 <sup>3</sup>	0.08	4•10 <sup>4</sup>		2 10 <sup>-3</sup>	9	
Pu238	4.5	7.5.104	30 · .	5•10 <sup>5</sup>		1.5	3•10 <sup>5</sup>	
Pu239	145	9 • 10 <sup>3</sup>	538	3•10 <sup>4</sup>		1318	8 • 10 4	
Pu240	59	1.3.104	494	1 • 10 <sup>5</sup>		441	10 <sup>5</sup>	
Pu241	27	3 • 10 <sup>6</sup>	291	3•10 <sup>7</sup>		120	10 <sup>7</sup>	
Pu242	9	37	219	800		75	300	
Am241	2	6•10 <sup>3</sup>	32	10 <sup>5</sup>	1.9	11	4 • 10 <sup>4</sup>	
Am242M	0.01	1 • 10 <sup>2</sup>	0.3	3•10 <sup>3</sup>	2.2	0.2	2 • 10 <sup>3</sup>	
Am243	2.5	5•10 <sup>2</sup>	43	8•10 <sup>3</sup>	1.9	5.6	1 • 10 <sup>3</sup>	
Cm242	0.09	3•10 <sup>5</sup>	0.9	3•10 <sup>6</sup>	1.85	0.3	10 <sup>6</sup>	
Cm243	0.02	10 <sup>2</sup>	0.02	9•10 <sup>2</sup>		0.02	8 • 10 <sup>2</sup>	
Cm244	0.83	7•10 <sup>4</sup>	8.8	<b>7•</b> 10 <sup>5</sup>	6.3	0.3	3•10 <sup>4</sup>	

Tab.4:	The	yearly	prod	uction	of	heavy	actini	.des [	kg/y]	and	the	associated	activity	[Ci/y]
	The	reactor	s in	questi	on	are 10	DOO MWe	powe	r plar	nts.				

for reprocessing units. The recycling of the higher actinides will very probably yield an even worse situation. In a Purecycling strategy, this is the only possible way at present, because the seperation of these strong  $\alpha$ -emitters very probably will not technically feasible for the next 2 decades.

In <u>concluding</u> this section, we find <u>differencies</u> in actinide content and radiation <u>up to about a factor of 10</u> 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 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 <u>Fig. 2</u> (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 INDC. 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 differencies 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:  $\pm$  3 %, Pu239:  $\pm$  7 %, Pu240:  $\pm$  11 %, Pu241:  $\pm$  17 %, Pu242:  $\pm$  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 %. Differencies 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 <u>Tab. 5</u> and <u>Tab. 6</u>, 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 %).

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

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

Notes: <sup>(a)</sup> Two standards deviation precision.

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 <sup>(a)</sup> Relative Uncertainty in Measurement	(b) Difference between Calculation and Measurement
Np237	Np237 dpm/g U	<u>+</u> 15	- 34
Pu236	Pu236/Pu239	<u>+</u> 24	8
Pu238	Pu238/Pu239	<u>+</u> 4	- 28
Am241	Am241/Pu239	<u>+</u> 24	- 4
Am243	Am243/Pu239	measured data not resolved	-
Cm242	Cm242/Pu239	<u>+</u> 10	- 24
Cm244	Cm244/Pu239	<u>+</u> 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.

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

In <u>conclusion</u> 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  $\alpha$  particle source in LWRs. 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.

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

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  $1 + \alpha$  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 <u>Tab. 8</u> 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.

			an and an air state of the stat		and the second se			an a
ISOTOP	<sup>τ</sup> 1/2	$\overline{\sigma_{\gamma}}$ (ba	arns)	σ <sub>f</sub> (b	arns)	RI	RI <sub>f</sub>	Effect of
	,	LWR	FBR	LWR	FBR	LIVR	LWR	interest
Th232	≃10 <sup>9</sup> y	7.4	0.44		0.0014	83.		reactivity
Pa231	3.2 10 <sup>4</sup> y	200.	0.80	-propulse garma - alba farma	0.38	480.	and and a second se	mactivity, rad. precursor -
U233	1.8 10 <sup>5</sup> y	49.	0.39	525.	3.15		746.	reactivity
U236	24 10 <sup>6</sup> y	6.	0.66	0.	0.11	210.	0.	reactivity
Np237	2.1 10 <sup>6</sup> y	170.	0.76	.002	0.36	756.	0.	Pu38 build up a +y 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.	a-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	an a	reactivity, $\alpha$ source
Pu241	14.6y	550.	0.432	1480.	2.5	140.	537.	reactivity, $\beta$ source
Pu242	3.8 10 <sup>5</sup> y	18.5	0.34	.003	0.28	1280	.6	reactivity, $\alpha$ 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.	β+y source
Am242M	152y	2000.	0.40	6000.	1.83	0.	0.	isom. trans. a source
Am243	7650.y	105.	0.55	0.45	0.24	1500.	1.5	a 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.	a source
Cm244	18.1y	10.	0.37	1.2	0.41	650.	12.	a + n source



- 20. -

In <u>Tab. 9</u> 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 <u>Tab. 10</u>.



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

According to the listed nuclear data in <u>Tab. 8</u> and the isotope chains in <u>Tab. 9</u> and <u>Tab. 10</u>, 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 <u>reactivity</u> 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  $\beta$ -and  $\gamma$  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 ( $\gamma$ ), 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.  $\alpha$  and  $\beta$  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 ( $\alpha$ ,n)do contribute to the radiation level. In a thorium system U232 and the hard  $\gamma$ -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 <u>relative</u> 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 postirradiation 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 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 <u>Tab. 11</u>, with the corresponding + or - signs.

- 23 -

()		LMFBR	PWR	HTGR
ISOTOP	σ		δσ [ɬ]	
Th232	(n, y)			20
	(RI)			20
	(n,2n)			100
Pa231	(n, y)			50
U233	(RI) <sub>f</sub>			- 20
U235	(RI)		20	
U236	(RI)		50	20
U238	(n, y)	20	20	
	(RI),		20	
	(n,2m)	50		
N237	(n, y)	50	50	
	(n,2n)	100	100	
Np239	(n,y)	50	50	
Pu239	(n, y)	20		
	(n,f)	- 20	- 20	
	(n,2n)	100		
Pu240	(n, y)	20		20
	(RI) <sub>Y</sub>		50	
Pu241	(n, y)	- 20		
	(n,f)	- 20		
Am241	(n, y)	- 50	+ 50	50
	(n,f)	- 50		
Cm242	(n, y)	50	50	
Cm243	(n, y)	50	50	

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

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

- <u>nuclide concentrations</u> during operation of the reactor and after discharge of fuel, including cooling times before reprocessing.
- <u>heat production</u> of discharged fuel for various cooling times:
- neutron source intensity due to spontaneous fission and  $(\alpha, n)$  processes at and after discharge of fuel.
- the <u>radioactive hazard potential</u> of discharged fuel and of fuel waste.

<u>6.3 Results of the performed sensitivity studies</u> 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.

	Relativ	e Cumul	ative.	Change	nange of Concentrations (%) at Discharge					
ELEMENTS						I	SOTOPES			
Pu	Am	Cm	Np	U6	Np7	Puð	Pu41	Am4 1	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 <u>Tab. 13</u>. 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.

	Nucl.data		DI	SCHA	RGED	FUEL						WASTE		
	Change	HAZA	RD-IND	EX	THE	RMPO	WER	SPON	r.FISS	•	HAZA	RD-IND	EX	
Nuclidê	00 //	OD	150D	1Y	OD -	150D	1Y	OD	150D	1Y	10Y	10 <sup>2</sup> Y	10 <sup>3</sup> Y	
U235	δ(RI) <sub>c</sub> =20	2.	4.	5.	2.	3.	3.	2.	2.	3.	3.	1.	1.	
U236	δ(RI) <sub>c</sub> =50	4.	10.	13.	0.	5.	8.	0.	0.	0.	0.	0.	0.	
	δCAP.=20	0.	0.	0.	о.	0.	0.	-1.	-1.	-1.	-2.	0.	0.	
0230	δ(RI) <sub>c</sub> =20	1.	-3.	-5.	2.	0.	-4.	-10	:-12.	-13.	-11.	4.	4.	
N237	<b>&amp;CAP.=</b> 50	0.	0.	0.	٥.	0.	0.	0.	0.	0.	ο.	0.	0.	
	δ(n,2n)=100	0.	0.	0.	0.	0.	0.	0.	0.	0.	о.	0.	0.	
Np239	&CAP.=50	0.3	0.	0.	0.	0.	0.	0.	0.	0.	0.	0.	0.	
Pu240	δ(RI) <sub>c</sub> =50	-1.	8.	7.	0.	11.	9.	15.	14.	15.	15.	11.	10.	
Pu241	ðFISS.=-20	1.	6.	5.	0.	7.	6.	8.	8.	8.	8.	8.	8.	
Cumula- tive Effect	δSIGMA	12.	38.	37.	11.	40.	39	29.	26.	25.	26.	35.	31.	
Reference Values		2.9	• 10 <sup>11</sup> []	m <sup>3</sup> ]	10 <sup>5</sup> [WATTS]			$1.7 \cdot 10^9 \frac{\text{NeUTR}}{\text{SEC}}$			3.2.10 <sup>9</sup> [m <sup>3</sup> ]			

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

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  $\alpha$ -emitters. 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 an 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  $(n, \gamma)$  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 <u>Tab. 16</u> 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 <u>Tab. 11</u>, 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

	Relative Cumu	lative Change of	Concentration	ns (%) at Dis	charge		
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.

	Nucl.Data			DI	SCHAI	RGED F	UEL				WASTE		
Isotope	Change	HAZ	ARD-IN	DEX	THE	RM.POW	ER	SPO	NT.FIS	s.	HAZARD-INDEX		
	8%	OD	150D	1Y	OD	150D	1Y	OD	<b>150</b> D	1 <u>,</u> Y	30Y	10 <sup>2</sup> Y	10 <sup>3</sup> Y
	&CAP.=20	1.	0.	3.	ο.	0.	3.	9.	9.	10.	5.	3.	1.
Th232	δ(RI) <sub>c</sub> =-20	2.	0.	8.	-2.	0.	8.	24.	24.	25.	13.	8.	3.
	δ(n,2n)=100	0.	0.	2.	٥.	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	δ(RI) <sub>c</sub> =+20	2.	6.	15.	1.	6.	15.	3.	3.	3.	9.	11.	2.
Pu240	&CAP=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.
Cumu- lative Effect	δSIGMA	7.	12.	17.	7.	11.	19.	-4.	-2.	0.	15.	24.	52.
Reference Values		3.8.10 <sup>11</sup> [m <sup>3</sup> ]			1.3.10 <sup>5</sup> [WATTS]			8.2.107 NEUTR. SEC			6.7•10 <sup>9</sup> [m <sup>3</sup> ]		

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

	Nugl data	Relative Chan	ge (%	) after	Disc	harge	ali da se di mana da di mandra manima anga		****		
Isotope	change	Concentration	The	rm. Pow	er	Spont	. Fiss		Haz.	Index	
	δσ   8	OD	OD	150D	1Y	OD	150D	1Y	OD	150D	1 <u>Y</u>
Pu241	δ CAP = -20 δ FISS= -20	Pu238 -37. Pu241 7.	0		-00	_44	-30	-20		5	0
Am241	δ CAP = -50 δ FISS= -50	$\begin{array}{ccc} \text{An}2421 & \text{S.} \\ \text{Am}242M & -47. \\ \text{Cm}242 & -47. \\ \text{Cm}244 & \text{S.} \end{array}$	0.		-23.	-44.	-30.	-20.	1.	J.	0.
Am241 Cm242 Cm243	$\delta .CAP = 50$ $\delta FISS = -50$ $\delta CAP = -50$ $\delta CAP = 50$	Pu238       40.         Am241       -       2.         Am242M       50.       50.         Cm242       50.       Cm244         O.       6.       6.	2.	36.	25.	40.	33.	23.	1.	10.	5.
Am243 Cm242 Cm243 Cm244	$\delta CAP = 50$ $\delta CAP = 50$ $\delta CAP = 50$ $\delta CAP = 50$ $\delta CAP = -50$	Am243 - 1. Cm242 O. Cm244 41.	0.	0.	0.	6.	9 <b>.</b>	15.	0.	0,	0.
Reference Values			2.5.	10 <sup>5</sup> [wat	rs]	1.5.1	0 <sup>9</sup> NEU se	<u>IR</u> . c	7•10	11 m <sup>3</sup>	]

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

.

•

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 <u>Tab. 17</u> for a 1000 MWe system, where core and blanket elements are reprocessed commonly. The comulative effects yield about a 20 % difference in 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 differencies 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 differencies are less than those obtained earlier in this paper.

# 7. Conclusion

As has been discussed in the preceding chapters, especially in 6.1 and 6.2, an accuracy of the basic data for transactinide 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 appreciable, 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

- 31 -

	pere a gi a farral — una quante a sta con annata		Discha	argec	d Fue	∋1		an a	1998 - <u>1999</u> - 1997 -	an de ser ante a de la desar de ser de s	Was	Waste		
<b>T</b>	Nucl. data change	Haz	ard Ind	lex	The	m.Pov	ver	Spor	nt Fis	55	Hazard Index			
Isotope	[ % ]	OD	150D	1Y	OD	1501	צו כ	OD	150	D <b>1</b> Y	30Y	10 <sup>2</sup>	ү 10 <sup>3</sup> ү	
U2.38	δ CAP =20	11.	1.	2.	11.	0.	1.	-5.	-5.	-4.	0.	0.	0.	
D220	δ CAP =20	1.	1.	2.	1.	2.	2.	1.	1.	1.	1.	2.	3.	
Pu239	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.	
	δ CAP =-20	0.	0.	0.	0.	0.	0.	0.	0.	0.	0.	0.	0.	
Pu241	δ FISS=-20	0.	3.	3.	0.	2.	1.	0.	0.	0.	5.	5.	4.	
	$\delta \chi = 7$	0.	-3.	-3.	0.	-3.	-3.	0.	о.	0.	0.	0.	0.	
	δ CAP=-50	0.	-7.	-3.	0.	-24.	-14.	0.	0.	0.	0.	.0.	2.	
Am241	δ FISS≖-50	0.	0.	0.	0.	0.	0.	-34.	-27.	-17.	о.	0.	0.	
	δλ =- 7	0.	0.	0.	0.	0.	0.	0.	0.	Ο.	0.	0.	0.	
Cm242	δ. <u>CAP</u> =-50	0.	0.	0.	0.	0.	0.	0.	0.	0,	0.	0.	0.	
Cumulative	δ SIGMA	22.	11.	15.	22.	-12.	0.	-23.	-15.	-5.	18.	20.	20.	
Effect	δλ ·	0.	0.	0.	0.	0.	0.	- 3.	- 1.	1.	0.	0.	-6.	
Reference Values		5.5	•10 <sup>11</sup> [r	n <sup>3</sup> ]	2 • 10	5 [w	vīrs]	1.4	•10 <sup>8</sup>	neutr. sec.	7~10	− <sup>8</sup> [m	3]	

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

.

.

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 achive the goal within the next 5 years. At this 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	1/	Pigford T.H., <b>Environ</b> mental Aspects of Nuclear Energy Production Anu.Rev.Nucl.Sci.24, 515, (1974)
1	2/	Hinkelmann, B. Report KFK 1186 EANDC 128-U (1970)
1	3/	Lederer C.M., Hollander J.M. and Perlmans I.,
		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
1	7/	Mattes, M., Emendörfer D., Keinert, J., Brestrich, I., Lutz, D.,
		Report IKE C-80 (1974),in German.
1	8/	Bell, M.J., Report ORNL-4628 (1973)
1	9/	Fleischmann, R.M., Mevrill, E.T., Heeb, C.M.,
		Quaterly Report BNWL 1876
1.	10/	Pigford, T.H., see ref. /1/
1.	11/	Ezure, H., Private Communication (Contributor)
1.	12/	Koch, K., et al.: Report EUR 4690 d (1971)
1.	13/	Arima, A., et al.: Report EUR 4638e (1971)
1	14/	Bresesti, A.M., et al.: Report EUR 4909 e(1972)
1	15/	Nodvik, R.J., see ref. /5/
1.	16/	Sweet, D., Private communication (1975)
1.	17/	Scholtyssek, Report KFK 1275/3 and Private communication (1975)
1.	18/	Vaminbraukx, R., Report CBNM/RN/1/74 (1974)

Results and Main Conclusions of the IAEA-Advisory Group Meeting on Transactinium Nuclear Data, held at Karlsruhe, November 1975

by

H. Küsters and M. Lalović

#### 1. Objectives of the Meeting

The meeting was held November 3-7, 1975 at the Nuclear Research Center Karlsruhe. About 50 scientists from research and industrial institutions of 16 countries had followed the invitation by IAEA to discuss the following objectives:

- a) Review of the present situation regarding the use of TND in reactor and non-reactor applications, specification of nuclear data requirements and their priorities.
- b) Review of the status of knowledge of TND, including a critical comparison of existing evaluations and compilations.
- c) Identification and discussion of TND measurements, compilations and evaluations required to satisfy user needs.

With a view to these objectives the meeting was organized into three main sessions:

A. Review of TND applications by the users (reactor and non-reactor applications).

B. Review of TND Status.

C. Detailed discussion of A and B and specification of recommendations.

2. Application and Requirements of TND (Session A)

The importance of TND in <u>reactor applications</u> comes from the following problems (in this contribution we exclude the dominating isotopes as U235, U238, Pu239):

# a) Reactor Operation including Recycling of Fuel

The main interest with respect to TND isotopes is their influence on the reactivity of the system via absorption and (spontaneous and induced) fission processes.

# b) Reactor Shutdown

- 1. The shutdown <u>reactivity</u> margin has to be within the requested bounds, which are influenced by spontaneous fission of heavy actinides (subcriticality).
- 2. The produced <u>heat after shutdown</u>, mainly by  $\beta$  and  $\gamma$ -emission of heavy nuclei, determines the energency core cooling layout.

# c) Transport, Reprocessing and Refabrication of Spent Fuel

The prediction of radiation and heat level depends on the neutron,  $\gamma$ -,  $\alpha$ - and  $\beta$ -output of spent fuel and has to be known for the design of appropriate shielding and cooling devices.

#### <u>d) Fuel Waste</u>

Non-recovered fuel forms a potential hazard because of its contents of long-lived  $\alpha$ -emitting isotopes of the heavy elements Np, Pu, Am and Cm (besides fission products). Therefore the in-pile production rate of these isotopes and the out-of-pile decay rate is of prime interest.

As a guideline the main isotopes contributing to the various items above are listed in <u>Table 1</u> for reactor systems of present interest. (The results are based on ORIGEN (version 1973) calculations).

BY DISCHARGE	LWR (	(1000 MWe)	LMFB	R (2000 MWe)	HTG	R (1000 MWe)
CONCENTRATION gr Ton Heavy M.	U238 U235 Pu239 U236 Pu240 Pu241 Np237 Pu242 Pu238 Am243 Cm244 Am241 Cm242	9.6.10 <sup>5</sup> 9.2.10 <sup>3</sup> 8.10 <sup>3</sup> 6.7.10 <sup>3</sup> 2.4.10 <sup>3</sup> 1.7.10 <sup>3</sup> 9.4.10 <sup>2</sup> 520. 475. 209 108. 42. 21.	U238 Pu239 Pu240 Pu241 Pu242 Am241 Am243 Np237 U236 Pu238 Cm242 Am242M Cm244	$1.5 \cdot 10^{6}$ $1.9 \cdot 10^{5}$ $7.9 \cdot 10^{4}$ $2.9 \cdot 10^{4}$ $1.4 \cdot 10^{4}$ $3. \cdot 10^{3}$ $5.7 \cdot 10^{2}$ $300.$ $250.$ $94.$ $62.$ $31.$ $19.$	Th232 U233 U234 U235 U238 Np237 Pu238 Pu238 Pu239 Pu241 Pu240 Cm244 Am241	8.6.10 <sup>5</sup> 2.10 <sup>4</sup> 1.5.10 <sup>4</sup> 1.10 <sup>4</sup> 3.10 <sup>3</sup> 2.10 <sup>3</sup> 1.10 <sup>3</sup> 2.7.10 <sup>2</sup> 97. 92. 5. 4.
THERM. POWER	Np239 Cm242 U237 Pu238	3.1.10 <sup>4</sup> 2.6.10 <sup>3</sup> 1.10 <sup>3</sup> 2.7.10 <sup>2</sup>	Np 239 Cm 242 Pu 240 Am 242 Pu 239 Am 241	8.8.10 <sup>4</sup> 7.6.10 <sup>3</sup> 543. 472. 370 347	Pa233 Pa234M Np239 Pu238	3.9.10 <sup>4</sup> 2.10 <sup>3</sup> 6.4.10 <sup>2</sup> 6.10 <sup>2</sup>
Sp. Fiss. [NEUTR/SEC]	Cm244 Cm242 Cm246 Pu240 Pu238	1.2.10 <sup>9</sup> 4.2.10 <sup>8</sup> 1.3.10 <sup>7</sup> 2.2.10 <sup>6</sup> 1.1.10 <sup>6</sup>	Cm242 Cm244 Pu240 Pu242 Pu238	1.2.10 <sup>9</sup> 2.2.10 <sup>8</sup> 7.4.10 <sup>7</sup> 2.8.10 <sup>7</sup> 2.2.10 <sup>5</sup>	Cm244 Cm242 Pu238 Cm246	6.3.10 <sup>7</sup> 1.5.10 <sup>7</sup> 2.5.10 <sup>6</sup> 5.5.10 <sup>5</sup>
(α,n) [NEUTR/SEC]	Cm242 Cm244 Pu238 Pu240	2 • 10 <sup>8</sup> 2 • 10, <sup>7</sup> 1 • 6 • 10 <sup>7</sup> 8 • 4 • 10 <sup>5</sup>	Cm242 Pu240 Am241 Pu239 Cm244	6. •10 <sup>8</sup> 2.8•10 <sup>7</sup> 2.1•10 <sup>7</sup> 1.9•10 <sup>7</sup> 3.8•10 <sup>6</sup>	Pu238 Cm242 Cm244 U232	3.6.10 <sup>7</sup> 7.7.10 <sup>6</sup> 1.10 <sup>6</sup> 3.7.10 <sup>5</sup>
HAZARD. INDEX [m <sup>3</sup> ]	Np 239 U237 Cm242 Pu238 Cm244 Am242 Pu241	2.3.10 <sup>11</sup> 1.6 10 <sup>10</sup> 3.5.10 <sup>9</sup> 1.6.10 <sup>9</sup> 1.2.10 <sup>9</sup> 1.2.10 <sup>9</sup> 9.10 <sup>8</sup>	Np239 Pu241 Cm242 U237 Pu240 Am242 Am241 Pu239	$6.5 \cdot 10^{11}$ $1.5 \cdot 10^{10}$ $1. \cdot 10^{10}$ $3.6 \cdot 10^{9}$ $3.5 \cdot 10^{9}$ $3.5 \cdot 10^{9}$ $2.6 \cdot 10^{9}$ $2.4 \cdot 10^{9}$	Pa233 Np239 Pu238 Cm242 Th234 Cm244	2.9.10 <sup>11</sup> 4.7.10 <sup>9</sup> 3.7.10 <sup>9</sup> 1.3.10 <sup>8</sup> 9.3.10 <sup>7</sup> 6.4.10 <sup>7</sup>

Tab. 1 Contribution of various Actinides in Composition and Radiation Output at Discharge of Reactor Fuel. For 1000 years after discharge of fuel, the relative contribution of the main isotopes to the waste hazard index is as follows:

<u>PWR</u>: (Am241 : Am243 : Pu240 : Pu239 : Cm245) ∻ (16 : 9 : 5 : 1 : 0,5)
 <u>LMFBR</u>: (Am241 : Pu240 : Am243 : Pu239 : Pu238) ∻ (70 : 3 : 3 : 3 : 0,1)

According to the various papers presented, the most restrictive requirements for TND in reactor applications at present are given by the effect these nuclei have on core performance. For the cross sections of the heavy nuclei such as U233, U235, U238, Pu239, Pu240, Pu241 and Pu242, data requests are already formulated in WRENDA; accuracies of about 5 % or better are requested.

According to Barré and Bouchard (paper A2), the <u>reactivity effects</u> of the various nuclides were calculated and are listed in <u>Table 2</u> (the main actinides such as U235, U238 and Pu239 are not included).

Reactivity	Thermal Reacto	or 30000 <u>MWd</u> PWR	Fast Reactor(1300 MWe)				
δk	Uranium Fuel	Pu-Recycling	Burnup				
k			0	120.000 MWd/t			
> 10 <sup>-2</sup>	U236,Pu240,Pu241	U236,Pu240,Pu241	Pu240,Pu241,Pu242	Pu240,Pu241,Pu242			
$10^{-3}$ to $10^{-2}$	U234,Np237,Pu242	U234,Np237,Pu241 Am241,Am243	Pu238, Am241	Pu238, Am241, Am242, Am243			
$10^{-4}$ to $10^{-3}$	Pu238,Am241, Am243,Am242m	Pu238,Am242m		Am242,Cm244,Np237, Np239			
$10^{-5}$ to $10^{-4}$		Cm242,Cm243,Cm244	Np237	Cm243			

Tab.2: Reactivity Effects of Actinides in Thermal and Fast Reactors.

For a thermal HTR the main influence on the reactivity, apart from Th and U233, is given by Pa231, U232, U234.

The effect on other core parameters such as <u>power</u> and <u>breeding gain</u> is again dominated by the higher Pu-isotopes; in a fast reactor the contribution to reactor power by Pu240/Pu241/Pu242 is about 6%/8%/1%, whereas in thermal reactors only Pu241 is of importance (2-3% for uranium fuelled cores, 5 to 10 % for Pu-recycled systems). The other actinides contribute in a fast system less than 1 % each, in total about 1 %. Pu240 is essential for its influence on the thermal neutron spectrum, but no major deficiencies have shown up to date. At the end of fuel life Am241 may have an effect on the breeding gain by about 0.02.

According to these investigations the cross section data most urgently needed are those for the higher Pu isotopes (including Pu238) and Am241, Am242m and Am243; in particular those for fast reactors, as the main data gaps exist in the epithermal and fast energy region. This is being stressed by experimental results from fast zero-power reactors, where e.g. the prediction of the capture-to-fission ratio for Am241 deviates by a factor of about 2 from experimental results (paper A3). In thermal systems the data are known to better accuracy.

It should be kept in mind that generally the formation of the  $\alpha$ -emitters Am and Cm does depend more strongly on the cumulated cross section uncertainties of the preceding isotopes in the corresponding chains than on the cross sections of the "reference nuclide" itself. For instance, Cm244 with 18y half-life is formed via 3 routes: via Pu241  $\frac{\beta}{2}$  Am241 $\frac{n\gamma}{4}$  Am242  $\frac{\beta}{2}$  Cm242 $\frac{n\gamma}{4}$  Cm243 and via Pu241  $\frac{n\gamma}{4}$  Pu242  $\frac{n\gamma}{4}$  Pu243  $\frac{\beta}{4}$  Am243  $\frac{n\gamma}{4}$  Am244, the latter being by far the dominant route because of the competing strong  $\alpha$ -emission of Cm242. A third route is given by Pu241  $\frac{\beta}{4}$  Am241  $\frac{n\gamma}{4}$  Am242m  $\frac{n\gamma}{4}$  Am243  $\frac{n\gamma}{4}$  Am244  $\frac{\beta}{4}$  Cm244. Thus the uncertainty in Cm244 content is given by the uncertainties in Pu242,Am242m and Am243 capture data and by the absorption rate uncertainty for Cm244.

Data for the first three isotopes are estimated to have an uncertainty of about 20 % and 50 %, respectively, and the estimate for Cm244 is about 50 %. If cross section uncertainties tend to be on one side of the true value systematic error, then the uncertainty in the Cm244 cross section partially compensates the cumulated uncertainty of the formation process.

- 5 -

The out-of-pile behaviour of actinides is mainly determined by two facts: (1) the actinide concentration at discharge from the reactor and (2) their corresponding decay properties. With regard to neutron production in addition to spontaneous fission the occurence of  $(\alpha, n)$  processes, especially with oxygen, is to be considered, but the neutron production is about an order of magnitude less than that from spontaneous fission. From this fact it follows that cross sections play a minor rôle after discharge of burnt fuel. Therefore, the main objective in discussing the sensitivity of the radiation output from spent fuel to cross section changes is in establishing the corresponding variation in nuclide concentration at discharge from the reactor.

First of all, it is of primary interest to <u>compare calculated and measured</u> <u>nuclide concentrations</u> for various power reactor systems. Some of the available material is discussed by Küsters and Lalović (paper A3). Apart from the uncertainty in calculational methods, which, in principle, can be relatively small if sophisticated tools are used, the major uncertainty in the theoretical prediction of nuclide concentrations arises from reactor operation changes and often not fully known irradiation and flux histories of the discharged elements. Therefore the calculated U238 content in spent fuel is sometimes adjusted to the experimental value. For PWR fuel, however, this approach causes changes of up to about 40 % in the concentrations of less dominating actinide isotopes, e.g. in the Cm244 content. With simplified models (e.g. fundmental-mode calculations) the differences are even larger and must be expressed sometimes by factors.

A sensitivity analysis shows (A3) that for various times after discharge the radiation hazard is uncertain by about 20 to 50 %, if plausible uncertainties in nuclear data are assumed.

Quite generally, the dominating cause for differences in nuclide concentration and radiation output is given by the composition, the fuel cycle strategy (e.g. Pu recycling or uranium fuel cycle for PWRs) and the degree of burn-up of the reactor fuel; these effects may amount to about a factor of 10 e.g. in hazard potential for a PWR with and without Pu recycling. In addition, one has to keep in mind that for about 500 years after discharge the fission products cause most of the radiologic hazard. After that the actinides dominate mainly with  $\alpha$ -emission from Am, until after several thousand years the longlived Pu isotopes give the main contribution to radioactivity from a unit of discharged reactor fuel.

With regard to the decay properties of nuclei, quite generally uncertainties in decay constants  $\lambda$  cause uncertainties of the same order of magnitude in the concentration e.g. of  $\alpha$ -emitters at a decay time equal to the half-life. This is only approximately true because due to the decay of other nuclides the reference nuclide may increase in concentration. In other words, without this feedback and without external manipulation, a 10 % change in  $\lambda$  results in a 10 % change of nuclide concentration at t =  $1/\lambda$ .

At present uncertainties in decay constants are about 10 % or smaller, thus the related effects are not very important.

<u>Transmutation of actinides</u> in reactors was being considered at the meeting by Koch (paper A6) as an alternative to waste storage. In this case there is a definite need for cross sections (fission and capture) of the long lived  $\alpha$ -emitters. From a physical point of view reactors with hard neutron spectra ( $\sigma_c/\sigma_f$  small) are to be preferred. At present nuclear industry is not particularly interested in this topic; the waste storage strategy (e.g. in salt mines) is the favoured route for waste disposal. In this connection there was a discussion of separation of the  $\alpha$ -emitters Am, Cm and others from spent fuel. The necessary separation technology, however, is not likely to become available within the next two decades.

Very restrictive requirements (0.5 to 1 %) for the half-lives of the Pu-isotopes were stated by Dierckx (paper A8) in order to obtain sufficiently accurate results for the analysis of non-destructive and destructive techniques to determine the isotopic contents of spent fuel; these measurements are made for small waste residues and for safeguard purposes. It is not quite obvious, whether <u>safeguard techniques</u>, which are usually based on realtive measurements, do require such high precision.

The situation is different in the field of <u>non-reactor applications</u> of TND. Aten (paper A9) points out that e.g. for application in health physics and for purity tests high accuracies are required for  $\alpha$ -,  $\beta$ -,  $\gamma$ - and X-ray energies, not to mention the question of standards.

- 7 -

# 3. Status of TND (Session B)

The difficulties of experimental techniques for the measurement of TND will be summarized briefly. Also the present situation regarding availability of evaluated TND on nuclear data files will be given as well as the capability of nuclear theory.

#### 3.1 Differential measurements on TND

The main objective is the determination of fission and capture cross sections. A major difficulty in cross section measurements of the actinides is in obtaining pure isotopic samples; either they are not readily available or they are available only in very small quantities. Thus data for many of the higher actinides are fragmentary or non-existent. Sample preparation costs for most of the higher actinides are rather high; impurities and the activity of the samples require very sophisticated techniques to obtain reliable cross section data.

According to James (paper B2) some improvements in experimental techniques have been acchieved recently. In the resonance region resonance spins were measured by using polarized neutron beams and polarized targets; low-energy resonances having meV fission widths in the U238 subthreshold fission cross section could be detected at RPI with a Linac-pulsed neutron source in a lead slowing-down spectrometer. The Saclay group used successfully a large gas scintillator chamber cooled to liquid-nitrogen temperature for the measurement of fission cross sections. A spherical design of an ionization chamber by Dabbs reduces the maximum possible alpha path length and thus a pile-up. At ORELA fission cross section measurements could be performed on an isotope with only 30y a half-life. Spark chambers have the great advantage of spatial resolution in countering a pile-up but suffer from spontaneous sparking background. If oxides and light elements are used for encapsulation, fission neutron detectors with pulse shape discrimination do overcome the a pile-up, but then background from  $(\alpha, n)$  processes is produced.

For highly radioactive actinide isotopes with half-lives of days of longer the nuclear explosion technique has been used for neutron cross section measurements by time of flight at Los Alamos (paper B3). The accuracy of the data is not high, one has to except a typical total uncertainty for fission and capture cross sections of about 10 - 30 % over the intermediate and fast neutron energy region. Data (mainly fission cross sections) for the following nuclides have been obtained and are available from the four neutron data centers: Th230, Pa231, U232, U234, U236, U237, Np237, Pu239, Pu240, Pu241, Pu242, Pu244, Am241, Am242m, Am243, Cm243, Cm244, Cm245, Cm246, Cm248, Cf249, Cf252, Es253.

To give an impression on the availability of differential measurements, in <u>Table 3</u> the <u>present gaps</u> in experimental data for the various nuclei and the corresponding energy regions are given (see paper A1).

# 3.2 Evaluation of TND and Capability of Nuclear Theory

Because there are many gaps in differential nuclear data for actinides, the evaluation must rely heavily on nuclear reaction theory. In the thermal range the cross sections are calculated from resonance parameters by means of resonance theory. In the resolved-resonance range the Breit-Wigner single level formula is used most frequently, but for the fission cross sections also multilevel representations. The Lane-Lynn formalism is adopted for the calculation of average cross sections in the unresolved range. Optical-model and statistical theories are applied in the fast neutron energy range. On the basis of both differential data (if available) and nuclear theory, nuclear data for actinides have been evaluated in the last years (see paper B5a).

<u>Table 4</u> gives a summary of the present status (Notation in Tab.4: E4  $\equiv$  ENDFB/IV; K  $\equiv$  KEDAK; LLL  $\equiv$  Lawrence Laboratory Livermore.

References of Tab. 4 can be found in Appendix 1.

It is interesting to note Lynn's remarks (paper B5b) on the degree of confidence which can be placed on the results of present model calculations. He estimates the uncertainty of cross sections of a statistical of integral character (e.g. capture, fission, summed inelastic scattering, etc.) for transactinium nuclei to about 25 - 30 % (U, Np, Pu, Am, Cm244, Cm245), being rather worse for the very high transplutonium elements ( $\pm$  50 %) and for cross sections of more specific character (e.g. radiative capture populating an isomeric state as in Am242m.) While these uncertainties are poorer than the degree of accuracy attainable with very careful experimental techniques, this accuracy

- 9 -

	TYPE of	<u>TAB.3:</u>	GAPS in	n σ(E) for	Energy Interva	ls (∆E in M	íeV)	
ELEMENT	Cross Section	Th227	Th228	Th231	Th232	Th234 Th233		
	t	015.	0-2•10 <sup>-6</sup> 1•10 <sup>-5</sup> -15	0-15	-	0-15		
Th	n,y	015.	0-15	0-15	$2 \cdot 10^{-8} - 1 \cdot 10^{-7}$ $8 \cdot 10^{-6} - 2$	0-15		
	n,f	2•10 <sup>-8</sup> -15	0-0.2 1-15	0-15	0-0.7	0-15		
		Pa230	Pa231	Pa232	Pa233			
	t	0-15	0.002-15	0-15	0.01-15			
Pa	n,y	0-15	E <sub>th</sub> -0.1 6-15	E <sub>th</sub> -15	$E_{th}$ -15			
	n,f	0-15	$E_{th}^{-0.1}$ 4-15	$E_{th}^{-15}$	E <sub>th</sub> -15			
		U231	U232	U234	U236	U237	U238	U239
	t	0-15	0.01-15	0.001-15	0.001-15	0-15	$E_{th} - 1 \cdot 10^{-6}$	0-15
-	n,y	. 0-15	E <sub>th</sub> -15	E <sub>th</sub> -15	1.10 <sup>-6</sup> -0.06	E <sub>tb</sub> -15	$E_{th} = 7 \cdot 10^{-6}$	E <sub>th</sub> -15
U	n,f	E <sub>th</sub> -15	$E_{th} = 7 \cdot 10^{-6}$	$E_{th} - 2 \cdot 10^{-2}$	$E_{th} - 8 \cdot 10^{-5}$	$E_{th} - 7 \cdot 10^{-5}$	-	$E_{th}^{-15}$
			0.02-0.15	0.02-0.15	6-15	2-15		
		Np236	Np237	Np238	Np239			
N-	t	0-15	0.01-15	0-15	0-15		· · · · · · · · · · · · · · · · · · ·	
NP	n,y	0-15	E <sub>th</sub> -0.15	0-15	E <sub>th</sub> -15	,		
	n,f	E <sub>th</sub> -15	$E_{th} = 1 \cdot 10^{-7}$	E <sub>th</sub> -15	E <sub>th</sub> -15			
		Pu238	Pu240	Pu241	Pu242			
-	t	0.01-15	0.007-0.1	0.001-15	0.009-15			
Pu	n,y	$E_{th}^{-1 \cdot 10^{-5}}$	0.5-15	0.1-15	. <sup>E</sup> th <sup>-15</sup>			
	n,f		$E_{th}$ -5.10 <sup>7</sup>	-	E <sub>th</sub> -7.10 <sup>-5</sup>			
		Am241	Am242	Am242m	Am243	Am244		
	t	6.10 <sup>-5</sup> -15	0-15	0-15	0.001-15	0-15		
Am	n,y	0.4-15	0-15	0-15	E <sub>th</sub> -15i	0-15		
	n,f	8-15	$E_{th} - 2 \cdot 10^{-5}$	0-15	$E_{th} = 7 \cdot 10^{-5}$	E <sub>th</sub> -15		
		Cm242	Cm243	Cm244	Cm245	Cm246		
	t	0-15	0-15	$E_{-7 \cdot 10^{-6}}$	$E_{th}^{-15}$	6•10 <sup>5</sup> -15.		
Cm	n <sub>ø</sub> γ	E <sub>th</sub> -15	0-15	$E_{th} = 2 \cdot 10^{-5}$	$E_{th}^{-15}$	E <sub>th</sub> -15.		
	n,f	0-15		$E_{th_{15}-2} \cdot 10^{-5}$	$E_{th}^{42.10^{-5}}$	$t_{4}^{-2} \cdot 10^{-5}$	-	

<b>ISOTOPES</b>	File and/or Reference	Latest Data Revision	Energy Range [eV]
Pa231, Pa233, U232	Drake 67(1)	1967	$10^{-3} - 15 \cdot 10^{6}$
P233	E4, Young 70 (unpub.)	1974	10 <sup>-5</sup> -20 • 10 <sup>6</sup>
U234	E4, Drake 67A(2)	1967	$10^{-5} - 20 \cdot 10^{6}$
U236	E4, Drake 67A(2) McCrosson 71 (unpub.)	1971	10 <sup>-5</sup> -20 • 10 <sup>6</sup>
U236	Parker 64(3)	1964	$10^{3}$ -15 · 10 <sup>6</sup>
U237	Caner 75 (to be pub.)	1975	10 <sup>4</sup> -7 • 10 <sup>5</sup>
Pu238, Pu242, Cm244	E4, Dunford 67(4)	1967	$10^{-5}$ -20 · 10 <sup>6</sup> (E4)
Pu238	Caner 74 (6)	1974	10 <sup>-,3</sup> -15•10 <sup>6</sup>
U232, U236, Pu238, Pu240	Thomet 74 (8,7)	1974	3·10 <sup>3</sup> -10 <sup>6</sup>
Pu240	E4, Pennigton 74 (unpub.) Hunter 73(9)	1974	10 <sup>-5</sup> -20•10 <sup>6</sup>
Pu240	K, Caner 73 (11, 12)	1973	$10^{-3}$ -15 · 10 <sup>6</sup>
Pu241,Pu242	K, Caner 73A (12, 13, 14)	1973	10 <sup>-5</sup> -20 • 10 <sup>6</sup>
Np237	E4, Smith 69(15)	1973	$10^{-5}$ -20 · 10 <sup>6</sup>
Pa231, U232, U234, U236, U237, Np237, Np238, Pu236, Pu238, Am241, Cm242	Hinkelmann 70(16)	1970	0.025 - 10.10 <sup>6</sup>
Pu24 1	E4, Hummel 73 (unpub.)	1974	10 <sup>-5</sup> -20 • 10 <sup>6</sup>
U238, Pu239, Pu240, Pu241	Prince 70 (5)		10 <sup>4</sup> -15·10 <sup>6</sup>
U236, Np237, Pu238, Pu240, Pu241, Pu242, Am241, Am243, Cm244	Pearlstein 66 (10)	1966	$0.025 - 2.10^2$
Am241, Am243	E4, Smith 66 (unpub.)	1966	$10^{-5}$ -20 · 10 <sup>6</sup>
Cm245, C <sub>f</sub> 252	LLL, Howerton 75 (unpub.)	1974	thermal - $20 \cdot 10^{6}$
C <sub>f</sub> 252	Prince 69 (17)	1969	$0.025 - 15 \cdot 10^6$

Tab.4: Survey of Recent Evaluations on TND.

might be reasonable for those isotopes for which differential measurements have not been performed. To obtain a better prediction of cross section data, Lynn demands improvements in models and parametrisation: For total cross sections, elastic scattering, compound nucleus formation and neutron transmission coefficients the coupled-channel version of the optical model needs to be explored; level density formulations in particular provide a great area of uncertainty; considerable ignorance exists on the detailed mechanisms of radiative capture and also of neutron induced fission.

#### 4. Requirements for TND

According to the discussion and recommendations of 3 subgroups, the required accuracies for TND were specified. The requests should be considered as estimates, which reflect personal judgement and are only rarely based on thorough sensitivity analyses. As outlined in section 2 of this summary, the main reason may be that the effects of TND are swamped by the effects of fission products (for the first few hundred hears after fuel discharge) and by the differences in e.g. material composition, burn-up and fuel cycle strategy in power reactors of present interest.

In <u>Table 5</u> we have listed the various requests for TND. (Remark: If no request was formulated, the corresponding position in the table as well as the status were left empty.)

Recommendations for further measurements on TND may be deduced from  $\underline{Tab. 5}$ , and can be found in more detail in the redommendations of the subgroup discussions, which will be included in the proceedings of the meeting. Priority in measurement and evaluation should be put on data of the higher plutonium and americium isotopes. For americium, especially neutron radiative capture data and branching ratios are unknown or very uncertain.

		Decay P	Decay Properties			Cross Sections							
	Half-Life		Accuracy	(%)	Тур	e	Accuracy (%)						
Isotope	Γ <sub>1/2</sub>	Туре	Requested	Status	Therm. React.	Fast React.	Reque The <b>r</b> m.	st ¦ Fast	St Therm.	atus   Fast			
Pa231	1•10 <sup>4</sup> Y	a Taranana de la filipidada de presidente de presidente de presidente de la filipidade de presidente de la filip			C/I	1 1 1	10/20		10/7				
Pa233	27.d				C/I		10/10		12/4				
Th232	1.10 <sup>10</sup> Y				(n,2n)	r I I	50	1	-				
U232	74Y				C/I_/F/I <sub>f</sub>	) }	30/30/30	/30	2/6/12/	/15			
U233	1.6.10 <sup>5</sup> Y				(n,2n)		10	1	-	[ ]			
U234	2.5•10 <sup>5</sup> Y	a-Intensity	1	3	C/I	l .	5/5	) 6	2/12	) ]			
U236	2.4.10 <sup>7</sup> Y	λ	1	2	C/I	1 1 .	4/4	1	6/6	1			
U237	6.7d				C/I	1 1 1	100/100	l t	33/18	1			
Np237	2.2.10 <sup>5</sup> Y	a-Intensity	1	20	C/I <sub>c</sub>	C,F,(n,2n)	100/10	30/50/50	2/8	50/10/-			
Np239	2.3d				C	C/F	100	20/50		1 /-			
Pu236	2.8Y				C/I <sub>c</sub>	C/F	100/100	50/50	-/-	-/-			
Pu238	86.4Y	$\lambda / (\gamma / \alpha) - Int$	0.5/1/0.1	1,5/25/1	C/I/F	C/F	30/50/50	20/7	4/10/3	30/10			
Pu240	6.6•10 <sup>3</sup> Y	$\lambda / (\gamma / \alpha) - Int$	0.2/1/0.2	5/ <sup>Fact</sup> /1	C/I <sub>c</sub>	-	2/1	l *	1/12	1			
Pu241	13.2Y	$\lambda/\gamma-Int.$	1/1	5/5	C/I_/F		3/10/1	   *	3/5/1				
Pu242	3.8•10 <sup>5</sup> Y	$\lambda/\alpha$ -Int.	1/4	5/10	C/I		10/5	! ⊦ *	4/4	1			
Am241	458Y	γ-Intensity	1	2	C/I	C/F	10/10	5/15	3/9	30/30?			
Am242M	152Y				C/I/F	C/F	50/50/30	50/15	-/-/4	-/50			
Am243	795Y				C/I	C/F/(n,2n)	15/10	10/30/50	5/3	-/10/-			
Cm242	163d	λ	0.1	1	C/I <sub>c</sub>	C/F	50/50	50/25	50/30	-/-			
Cm243	35Y				C/I_/F	C/F	50/50/50	30/20	-/-/7	-/-			
Cm244	18Y				C/I <sub>c</sub>	C/F	50/50	50/50	20/10	-/30?			

Tab.5: Comparison of Requested and Achieved Accuracy of TND.

Notation:

C: Capture I<sub>c</sub>: Resonance Integral for neutron capture

F: Fission I<sub>f</sub>: Resonance Integral for neutron fission

\* Requests according to WRENDA

Appendix 1

#### References for Table 4

- 1 DRAKE, M.K., NICHOLS, P.F., Ga-7462 (1967)
- 2 DRAKE, M.K., NICHOLS, P.F., GA-8135 (1967)
- 3 PARKER, K., AWREO-30/64 (1964)
- 4 DUNFORD, C.L., ALTER, H., NAA-SR-12271 (1967)
- 5 PRINCE, A., Proc. 2nd. Int. Conf. Nucl. Data for Reactors, IAEA (1970) Vol. II, 825.
- 6 CANER, M., YIFTAH, S., IA-1301 (1974); also: Nucl. Sci. Eng. (to be published).
- 7 THOMET, P., CEA-R-4631 (1974)
- 8 THOMET, P., JAERI M 5984 (1975) 71.
- 9 HUNTER, R.E., STEWART, L., HIRONS, T.J., LA-5172 (1973)
- 10 PEARLSTEIN, S., BNL 982 (1966)
- 11 CANER, M., YIFTAH, S., IA-1243 (1972)
- 12 CANER, M., YIFTAH, S., SHATZ, B., MEYER, R., Proc. Int. Symp. Fast Reactor Phys., Tokyo (1973) 683.
- 13 CANER, M., YIFTAH, S., IA-1276 (1973)
- 14 CANER, M., YIFTAH, S., IA-1275 (1973)
- 15 SMITH, J.R., GRIMESEY, R.A., IN-1182 (1969)
- 16 HINKELMANN, B., KFK 1186 (1970)
- 17 PRINCE, A., BNL 50168 (1969)

#### Appendix 2

#### Review Papers Presented at the Meeting

- Al <u>S. Raman:</u> General Survey of Applications which require Actinide Nuclear Data.
- A2 J.Y. Barré, J. Bouchard: Importance des Donnees Nucleaires des Transactinides pour la Physique des Coeurs de Reacteurs Rapides et Thermiques.

and Related Sensitivities to Cross Section Changes.

- <u>H. Küsters, M. Lalović:</u> Transactinium Isotope Build-up and Decay in Reactor Fuel
- A4 R.M. Nunn:

A3

.

The Requirements for Transactinide Nuclear Data for the Design and Operation of Nuclear Power Plants.

A5 R.F. Burstall:

Importance of Transactinide Nuclear Data for Fuel Handling.

A6 L. Koch:

Survey of TND Application; European Programs in Waste Management (Incineration).

- A7 <u>S. Raman:</u> Some Activities in the US Concerning the Physics Aspects of Actinide Waste Recycling.
- A8 <u>R. Dierckx:</u>

Importance of TND for Fuel Analysis.

A9 <u>A.H.W. Aten:</u> TND Applications in Radiation and Energy Sources, Tracer Techniques, Life Science, Agriculture and Industry.

- Bl R.W. Benjamin: Status of measured Neutron Cross Sections of Actinides for Thermal Reactors.
- B1/ L.W. Weston:

Contrib. Neutron Capture Cross Sections of the Actinides.

- B2 <u>G.D. James:</u> Status of Neutron Cross Sections of Actinides in the Resonance Region - Linear Accelerator Measurements.
- B2/ Contrib.  $\frac{J. Blons, C. Mazur:}{Measure de \sigma_{v} de^{243} Am}$ .
- B2/ S.M. Kalebin:
- Contrib. Total Neutron Cross Section Measurements on 241,243 244,245,246,248 Cm.
- B3 <u>M.S. Moore:</u> Status of Neutron Cross Sections of Actinides in the Resonance and Fast Energy Regions - Underground Nuclear Explosion Measurements.
- B4 <u>S. Igarasi:</u> Status of Measured Neutron Cross Sections of Actinides in the Fast Region.
- B4,B2/ Articles from the Soviet Journal of Nuclear Physics on the
   Contrib. Measurement of Fission Cross Sections of: Th229, Th230, Pa231, U232, Np237, Pu239, Bk249, Cf249.
- B5a <u>S. Yiftah, Y. Gur, M. Caner:</u> Status of Transactinium Isotope Evaluated Neutron Data in the Energy Range 10<sup>-3</sup> eV to 15 MeV.

B5a/ The Transactinides in the Main Evaluated Neutron Data Files Contrib. (CCDN).

- B5b J.E. Lynn: Theoretical Calculation of TND for Evaluation Purposes.
  B5b/ M. Caner, S. Yiftah: Contrib. A Survey of Cross Section Evaluations Methods for Heavy Isotopes.
  B6 S.A. Baranov, A.G. Zelenkov, V.M. Kulakov: The Experimental Investigation of the Alpha Decay of Actinide Isotopes.
  B6/ List of Compilations and Evaluations of TND. Contrib.
- B7 C.W. Reich:

Status of Beta- and Gamma-Decay and Spontaneous Fission Data from Transactinium Isotopes.