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ANALYSIS OF SAMPLE AND FUEL PIN IRRADIATION EXPERIMENTS CARRIED OUT IN THE PHENIX REACTOR USING JEF2 BASIC NUCLEAR DATA

R. SOULE E. FORT G. RIMPAULT

Commissariat à l'Energie Atomique
Centre d'Etudes de CADARACHE
Service de Physique des Réacteurs et du Cycle
Département d'Etudes des Réacteurs
13108 St PAUL-LEZ-DURANCE Cedex FRANCE

I - INTRODUCTION

In connection with radioactive waste transmutation studies, there is renewed interest in the nuclear data for minor actinides and fission products.

In EUROPE and particularly in FRANCE, multitemperature libraries have been produced from the **JEF-2.2** evaluations /1/ (Joint Evaluated File Version 2.2), for the most important isotopes and also for the minor actinides (Np, Am, Cm) and the fission products (^{99}Tc, ^{129}I, ^{135}Cs) for which transmutation is being envisaged.

The validation of these libraries is now underway, based both on :
-the analysis of a wide range of integral experiments /2/,
-and especially concerning waste transmutation, the analysis of sample or fuel pin irradiation experiments carried out in the PHENIX reactor.

The irradiation experiments investigated are:

- the **PROFIL 1** and **2** experiments consisting of irradiations of samples of pure separated isotopes placed in a standard subassembly in the first row of the inner core of PHENIX and far away from neutronic perturbations, in order to obtain clean irradiation conditions.

- the **TRAPU** experiment consisting of the irradiation of mixed-oxide pins that contained plutonium of different isotopic compositions which were heavily charged in minor plutonium isotopes. These pins were placed in standard PHENIX subassemblies and irradiated in positions close to the center of the reactor.

This paper presents the preliminary results of the analysis of these experiments using the Joint Evaluated File **JEF2.2**, and the conclusions drawn concerning the capture and (n,2n) cross-sections of a number of major and minor actinide isotopes, comparing with the analysis reported in /3/ and performed with the **JEF-1** basic data.

II - EXPERIMENTAL BACKGROUND

II - A Irradiated Fuel Analysis

In the **TRAPU** experiment, three types of plutonium pins were used as indicated in Table I. Higher quantities of secondary actinides were studied to obtain more accurate data.Standard pins were placed in standard PHENIX subassemblies and irradiated during six cycles in positions close to the center of the reactor.
Unirradiated samples of the same fuel were also analysed, to provide data on the fuel before irradiation.

-Table I-

Isotopic Composition of the three TRAPU Fuel Pins

Plutonium Isotope Compositions (%)					
Experiment	^{238}Pu	^{239}Pu	^{240}Pu	^{241}Pu	^{242}Pu
TRAPU-1	0.1	73.3	21.9	4.0	0.7
TRAPU-2	0.8	71.4	18.5	7.4	1.9
TRAPU-3	0.2	34.0	49.4	10.0	6.4

II - B Experimental Techniques

After irradiation, small samples (20mm high) were cut from the experimental pins (both fuel and clad) and put into a solution. The objective of this analysis was to determine the fuel composition by nuclide. Neodynium-148 was used as a burn-up indicator since it is a stable fission product with a small capture cross-section, and it enables determination of the number of fissions that have taken place in the sample.

Mass spectrometry was then used, with simple or double isotopic dilution and well-characterised tracers. All of the analysis results are presented as ratios of concentrations. Since all the concentrations can be related directly or indirectly to the ^{238}U content, the fuel composition before and after irradiation can be compared, taking into account, by calculation, the ^{238}U consumption, which is always small (a few percent).

II -C- Accuracy of the Measurements

The experimental techniques described in Sec. II-B give the nuclide concentration ratios shown in Table II. The table also shows the global estimated accuracies for each quantity. This accuracy estimate also accounts for the reproducibility of the measurements. However, in quoting the final results, we have introduced a supplementary uncertainty, called "representativity" uncertainty, which is based on the consistency of the results obtained for a set of samples.

-Table II-

Measured Atomic Ratios and Estimated Experimental Accuracies

Measured Atomic Ratio	Accuracy at 2σ (DR or r = DR/R %)	Measured Atomic Ratio	Accuracy at 2σ (DR or r =DR/R %)
$^{234}U/^{238}U$	DR = ± 0.0003	$^{144}Nd/^{148}Nd$	DR = ± 0.02
$^{235}U/^{238}U$	r = ± 0.3 %	$^{145}Nd/^{148}Nd$	DR = ± 0.02
$^{236}U/^{238}U$	DR = ± 0.0005	$^{146}Nd/^{148}Nd$	DR = ± 0.02
$^{237}Np/^{238}U$	r = ± 3.0 %	$^{150}Nd/^{148}Nd$	DR = ± 0.02
$^{239}Pu/^{238}U$	r = ± 1.0 %	$^{241}Am/^{239}Pu$	r = ± 2.0 %
$^{238}Pu/^{239}Pu$	DR = ± 0.05	$^{242m}Am/^{241}Am$	r = ± 1.0 %
$^{240}Pu/^{239}Pu$	DR = ± 0.03	$^{243}Am/^{241}Am$	r = ± 1.0 %
$^{241}Pu/^{239}Pu$	DR = ± 0.02	$^{244}Cm/^{239}Pu$	r = ± 3.0 %
$^{242}Pu/^{239}Pu$	DR = ± 0.003	$^{242}Cm/^{244}Cm$	r = ± 3.0 %
$^{148}Nd/^{238}U$	r = ± 1.5 %	$^{243}Cm/^{244}Cm$	r = ± 5.0 %
$^{143}Nd/^{148}Nd$	DR = ± 0.02	$^{245}Cm/^{244}Cm$	r = ± 5.0 %

II -D- Separated Nuclei Sample Analysis

The most accurate experimental technique for obtaining information on the integral capture cross-section is to determine the variation in composition that results from high-flux irradiation of a pure sample. This method can be used for all the isotopes for which the descendant, obtained via neutron capture, is stable or has a long radioactive period.

One or two standard pins, with pure separated isotope capsules (46 in **PROFIL-1**, 2*42 in **PROFIL-2**) have been irradiated in a standard subassembly in the first row of the inner core of PHENIX. They were placed far away from neutronic perturbations, in order to obtain clean irradiation conditions. The samples were inside two stainless steel containers, as shown in Fig. 1.

The **PROFIL-1** pin is shown in Fig. 2. Table III lists the separated isotopes irradiated in the two experiments. The **PROFIL-1** irradiation was done during the first three cycles of Phenix; the **PROFIL-2** irradiation lasted four cycles.

The samples were analysed using the techniques described in Sec II-B. Again, the uncertainty in the variation in the number of atoms due to irradiation is of the order of $\pm$1% or less.

-Table III-

Separated Isotopes Irradiated in PROFIL Experiments

Experiment	Th	U	Np	Pu	Am	Cm
PROFIL-1		235		238	241	
				240		
				241		
				242		

PROFIL-2	232	233	237	238	241	244
		234		239	243	
		235		240		
		238		242		

III - FUEL IRRADIATION EXPERIMENT ANALYSIS

III- A- How to Isolate Basic Data

All the fuel irradiation experiment analyses were based on accurate evolution calculations, starting from the experimental values of the initial concentrations. In the present work, data from the new evaluated data file **JEF2.2** /1/ were used in the analysis. Standard procedures (based on two-dimensional diffusion codes) for computing flux distributions during irradiation and standard burn-up codes were used to calculate the irradiated isotope concentration variations. Corrections were also applied for variable spectrum effects and environmental perturbations.

It is well known that the calculated final concentrations depend on both the basic data and on the uncertainty in the irradiation history, as modeled in the calculations. The main effect of the irradiation history uncertainty can be suppressed by using the experimental data on the total fluence τ received by the irradiated pins or samples. Therefore, the experimentally measured concentrations of Neodynium fission products after irradiation were used in both the **PROFIL** and **TRAPU** experiments to correctly normalise the evolution calculations to the total absolute fluence.

Table IV shows the calculation/experiment ratio (C/E) values obtained in this way, for the **TRAPU** pin compositions at the end of irradiation. In particular, Table IV contains the results obtained by using:

- the **JEF-1** evaluated file /3/, see columns a)

- the **JEF2.2** evaluated file /1/, see columns b)

The global uncertainty (in %) is indicated in columns c) of Table IV.

-Table IV-

C/E Values of Final Concentrations in the TRAPU Experiments

^{238}U = 100	TRAPU-1		TRAPU-2		TRAPU-3	
	a)	b) c)	a)	b) c)	a)	b) c)
^{234}U	0.98	0.98 ±2.5	1.00	1.00 ±1.3	1.04	1.04 ±1.0
^{235}U	0.99	1.01 ±0.3	1.01	1.03 ±0.2	1.01	1.03 ±0.2
^{236}U	0.98	0.93 ±0.5	1.00	0.95 ±0.4	0.99	0.95 ±0.3
^{237}Np	0.91	0.74 ±6.8	0.90	0.74 ±3.3	0.85	0.73 ±3.2
^{238}Pu	1.02	0.99 ±0.9	1.00	1.02 ±0.4	0.99	1.03 ±0.4
^{239}Pu	1.00	1.02 ±0.4	0.98	1.01 ±0.3	0.98	1.01 ±0.3
^{240}Pu	0.99	1.00 ±0.4	0.98	0.98 ±0.3	0.98	0.99 ±0.3
^{241}Pu	1.03	1.06 ±0.4	1.00	1.01 ±0.3	1.02	1.04 ±0.3
^{242}Pu	1.08	1.12 ±0.5	1.03	1.06 ±0.4	1.01	1.03 ±0.3
^{241}Am	0.95	0.98 ±3.0	0.96	0.98 ±3.6	0.97	0.98 ±2.1
^{242m}Am	1.36	1.04 ±3.6	1.41	1.07 ±4.0	1.36	1.03 ±2.5
^{243}Am	1.08	1.10 ±3.6	1.05	1.06 ±4.0	1.08	1.09 ±2.5
^{242}Cm	0.96	1.04 ±2.4	0.95	1.01 ±2.6	0.94	1.01 ±2.1
^{243}Cm			1.13	0.76 ±2.7	1.13	0.76 ±2.6
^{244}Cm	1.03	1.04 ±2.0	1.15	1.15 ±2.2	1.16	1.17 ±1.7

From the data of Table IV, the following preliminary remarks can be made, anticiping some conclusions from Sec. III-B:

1. The sensitivity study suggests that the ^{238}U (n,2n) mean cross-section parameter (including both microscopic cross-section data and the weighting spectrum) is responsible for the discrepancy in the final concentrations of ^{237}Np. This suggestion is also confirmed by Fig. 3 where are plotted the ^{238}U (n,2n) microscopic cross-sections issued from:

- the experimental values published by **FREHAUT** and al./4/,
- the pointwise values included in the **JEF-1** file,
- the pointwise values included in the **JEF2.2** file.

2. The difference in the C/E values for the ^{242}Pu concentration in the three **TRAPU** experiments is related to a corresponding difference in sensitivity of the ^{242}Pu concentrations in the same experiments, the sensitivity being maximum for **TRAPU-1** (+30%), intermediate for **TRAPU-2** (+20%) and minimum for **TRAPU-3** (+10%). This result is also a confirmation of the **PROFIL** experimental trend, which indicates that the **JEF2.2** data overestimate the ^{241}Pu radiative capture cross-section.

3. The C/E values for the ^{241}Pu concentration in the three **TRAPU** experiments are also a confirmation of the **PROFIL** experimental trend, which indicates that the **JEF2.2** data overestimate the ^{240}Pu radiative capture cross-section.

4. For the other results, concerning mainly minor actinides, the analysis of the C/E results is still now under investigations by sensitivity studies.

Besides these first considerations of the **JEF2.2** data, it must be noted that the calculated value of the postirradiation fuel concentration also depends on the basic data ensemble as a whole, and the only way to obtain separate information about the basic data themselves is to use results from different experiments in a statistical adjustment procedure.

In contrast, in the specific case of the **PROFIL** pure separated isotope irradiations (see Table III), the very simple decay schemes allow, in most cases, considering the irradiated sample concentrations to be dependent on a few parameters, and the results can be analysed directly in terms of average cross-section values (generally capture or (n,2n)) /5/.

The experimental values of the average cross-sections obtained in this manner constitute a very powerful synthesis of the irradiation results and allow for the easiest use of the experimental information.

III - B The General PROFIL Analysis

The first part of the PROFIL result analysis is quite similar to the general fuel irradiation experiment interpretation. In fact, very accurate evolution calculations concerning the concentration of each element in the irradiated sample were also carried out. For a very pure separated isotope sample of atomic mass A, the discrepancy between the experimental and calculated values of the quantities

$$\frac{\Delta N_{A+1}}{N_A} = \frac{N_{A+1}(\tau)}{N_A(\tau)} - \frac{N_{A+1}(0)}{N_A(0)}$$

and

$$\frac{\Delta N_{A-1}}{N_A} = \frac{N_{A-1}(\tau)}{N_A(\tau)} - \frac{N_{A-1}(0)}{N_A(0)} \qquad (1)$$

where $N_A(0)$ and $N_A(\tau)$ are the atom number density of an isotope of mass A before and after irradiation, respectively, can be practically considered to be the direct consequence of the uncertainty in the following integral rates, which in turn are easily related to the evolution calculation input data :

$$R_c(A) = \bar{\sigma}_c(A).\ \tau$$

and

$$R_{n,2n}(A) = \bar{\sigma}_{n,2n}(A).\ \tau \qquad (2)$$

where

τ = fluence received by the sample during the entire irradiation,

$\bar{\sigma}_c(A)$, $\bar{\sigma}_{n,2n}(A)$ = capture and (n,2n) average cross-sections of isotope A.

The less important effects on N_{A+1}/N_A and N_{A-1}/N_A due to parameters other than those appearing in Eqs.(2) are evaluated by sensitivity studies and are included as part of the method uncertainties in the final results.

As is generally done in fuel irradiation experiments, the total fluence $\mathcal{F}$ can be obtained from a large number of experimental results concerning the Neodynium fission product concentration in the **PROFIL** analysis. Thus, the irradiation history represented in the evolution calculation can be coherently normalised to the actual experimental value of $\mathcal{F}$. Moreover, in the **PROFIL** interpretation, information on the absolute fluence is obtained from a large number of Neodynium measurements in irradiated pure ^{235}U samples: Since the yields for ^{235}U fission are by far the best known, the uncertainty in the fluence normalisation of the **PROFIL** analysis is particularly small.

In this way,it is easy to see that the C/E ratios related to the two quantities in Eqs.(1) can be considered to be C/E ratios related to the reaction rate ratios (i.e., "spectral indexes") $\sigma_c(A)/\sigma_f(^{235}U)$ and $\sigma_{n,2n}(A)/\sigma_f(^{235}U)$, at the locations corresponding to the sample positions. Furthermore, adding the uncertainties in the local spectra computation to the Doppler effect evaluation and to the concentration evolution effect on the average cross-sectons, we can consider the same C/E ratios to be related to the spectral indexes in a more esily computed, infinite fuel medium spectrum (the so-called "fundamental-mode" spectrum) at room temperature.

It is worth noting that, in any case, the ratios between the two different PROFIL capture or (n,2n) reaction rate results are largely independent of the absolute fluence normalisation and the ^{235}U fission yield evaluation.

The results for the **PROFIL-1** and **PROFIL-2** irradiations are shown in Table V in terms of the capture and (n,2n) spectral indexes in the fundamental mode, according to the general **PROFIL** analysis procedure.

-Table V-

C/E Values for the PROFIL Experiments Using JEF-1, and JEF-2.2 Data

Data Type	JEF-1	JEF-2.2	Uncertainty
			(%)
$\bar{\sigma}_c$ (^{235}U)	0.97	0.95	± 1.4
$\bar{\sigma}_c$ (^{238}U)	0.96	0.95	± 1.6
$\bar{\sigma}_c$ (^{237}Np)	0.90	0.94	± 4.1
$\bar{\sigma}_{n,2n}$(^{237}Np)	1.19	1.19	± 15.0
$\bar{\sigma}_c$ (^{238}Pu)	0.95	0.97	± 3.0
$\bar{\sigma}_c$ (^{239}Pu)	0.97	0.99	± 1.8
$\bar{\sigma}_{n,2n}$(^{239}Pu)	1.38	0.59	± 11.0
$\bar{\sigma}_c$ (^{240}Pu)	1.06	1.12	± 1.6
$\bar{\sigma}_{n,2n}$(^{240}Pu)	0.83	0.85	± 14.0
$\bar{\sigma}_c$ (^{241}Pu)	1.11	1.21	± 3.7
$\bar{\sigma}_c$ (^{242}Pu)	1.16	1.31	± 3.5
$\bar{\sigma}_c$ (^{241}Am)	1.03	1.04	± 1.4
$\bar{\sigma}_c$ (^{243}Am)	0.94	0.89	± 5.0

The performance of the **JEF2.2** data is fairly coherent with the performance of the **JEF-1** data. Nevertheless, we can note some deteriorations on the C/E results concerning:

- the capture cross-sections of ^{240}Pu, ^{241}Pu and ^{242}Pu,

- the (n,2n) cross-section of ^{239}Pu.

Other improvements are needed for the capture data of all Plutonium isotopes. We have seen that the overestimation of the ^{240}Pu and ^{241}Pu radiative capture is confirmed by the **TRAPU** results.

It is worth noting the excellent result obtained using both the **JEF-1** and **JEF2.2** data for the ^{241}Am radiative capture cross-section. This result was obtained with a more specific analysis procedure, described in Sec.IV.

Statistical adjustments , based on sensitivity studies, have been performed concerning the radiative capture cross-sections of the major actinides and have confirmed the trends shown by the analysis of the **PROFIL** experiments /6/.

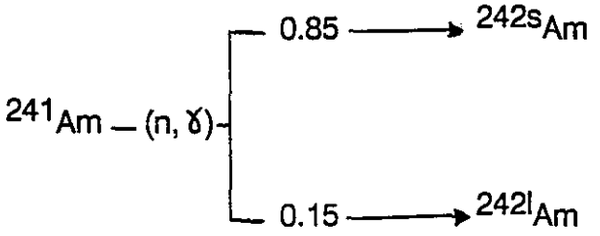
IV - ^{241}Am Sample Analysis

The decay chain for ^{241}Am neutron radiative capture (see Fig 4) has two different branches, each characterised by its own branching ratio, the first of which involves the population of the two isomeric states ^{242s}Am and ^{242l}Am (where s and l stand for "short-lived" ($T_{1/2}$ = 16h) and "long-lived" ($T_{1/2}$ = 141 yr), respectively).

It is evident that the calculated isotopic concentration ratio $^{242l}Am/^{241}Am$ depends not only on the capture ratio of the ^{241}Am nuclide, but also on the capture reaction isomeric cross-section ratio. The $^{238}Pu/^{241}Am$, $^{242}Cm/^{241}Am$, and $^{242}Pu/^{241}Am$ ratios depend not only on the ^{241}Am capture rate and on the corresponding isomeric ratio, but also on the ^{242}Am decay branching ratio, which is in turn better known than the former.

If the branching ratios used for the evolution calculation were the "true" ones, the decay scheme would depend on only one parameter (the ^{241}Am capture rate), and for the same reason all the C/E values corresponding to the four above concentration ratios should give the same result. On the contrary, if the different C/E values should be inconsistently dispersed, it is necessary to modify (within the appropriate range of uncertainty) the branching ratio values used in the calculations, in order to arrive at the required consistent C/E values.

Table VI shows the results of the C/E comparison obtained using the branching ratio values indicated in Fig.4: The C/E values are in marked disagreement. Table VII shows the much better consistency of the results obtained using the values of Fig.5. Thus, the experimental branching ratio for the ^{241}Am capture reaction in the fast spectrum used in the **PROFIL** irradiations can be established as:



and the associated absolute uncertainty is < 1%, since a 1% change in the 0.15 value of the long-lived branch affects the concentration ratio $^{242l}Am/^{241}Am$ by ~7% (i.e., 0.01/0.15).

TABLE VI

C/E Values for the Different 241Am Sample Isotope Concentration Ratios Obtained With the Branching Rate Ratios of Fig.4

Isotope Concentration Ratio	C/E on $\sigma_c(^{241}Am)^{a,b}$
$^{238}Pu/^{241}Am$	1.03 ±2.5[c]
$^{242}Pu/^{241}Am$	1.03 ±2.0
$^{242l}Am/^{241}Am$	1.40 ±0.5

[a] Average of various samples

[b] C/E values are in absolute values, while uncertainties are relative and given in %

[c] Uncertainty from sample analysis

TABLE VII

C/E Values for the Different 241Am Sample Isotope Concentration Ratios Obtained with the Branching Rate Ratios of Fig.5

Isotope Concentration Ratio	C/E on $\sigma_c(^{241}Am)$ [a,b]
238Pu/241Am	1.09 ± 2.5[c]
242Pu/241Am	1.09 ± 2.0
242lAm/241Am	1.04 ± 0.5
Average	1.04 ± 1.1

[a] Average of various samples
[b] C/E values are in absolute values, while uncertainties are relative and given in %
[c] Uncertainty from sample analysis

When the different C/E values become consistent, the average C/E value can be associated to the last free parameter of the decay chain. In the present case, C/E = 1.04 ± 1.1% can be considered to be the local C/E value associated with the **JEF2.2** $\sigma_c(^{241}Am)/\sigma_f(^{235}U)$ reaction rate ratio value used in the evolution calculations.

This same C/E value is reported in Table IV with a larger uncertainty, including the components associated with the passage from the true local spectrum to the fundamental-mode mean value.

V CONCLUSIONS

The irradiation experiments performed in PHENIX are a powerful source of information on the cross-sections of major and minor actinides.

They provide an experimental data base for the validation of basic data files.

Besides capture, fission and (n,2n) cross-sections, branching ratios can also be determined

The high accuracy of the experiments also allows us to define residual uncertainties, which can be used to reduce the uncertainties of relevant design parameters, such as the long-lived radioactive waste transmutation studies and strategies.

The C/E results obtained with the **JEF-2.2** data for the analysis of the irradiation experiments presented in this paper, are preliminary and some improvements are still necessary, especially by sensitivity studies and data adjustments

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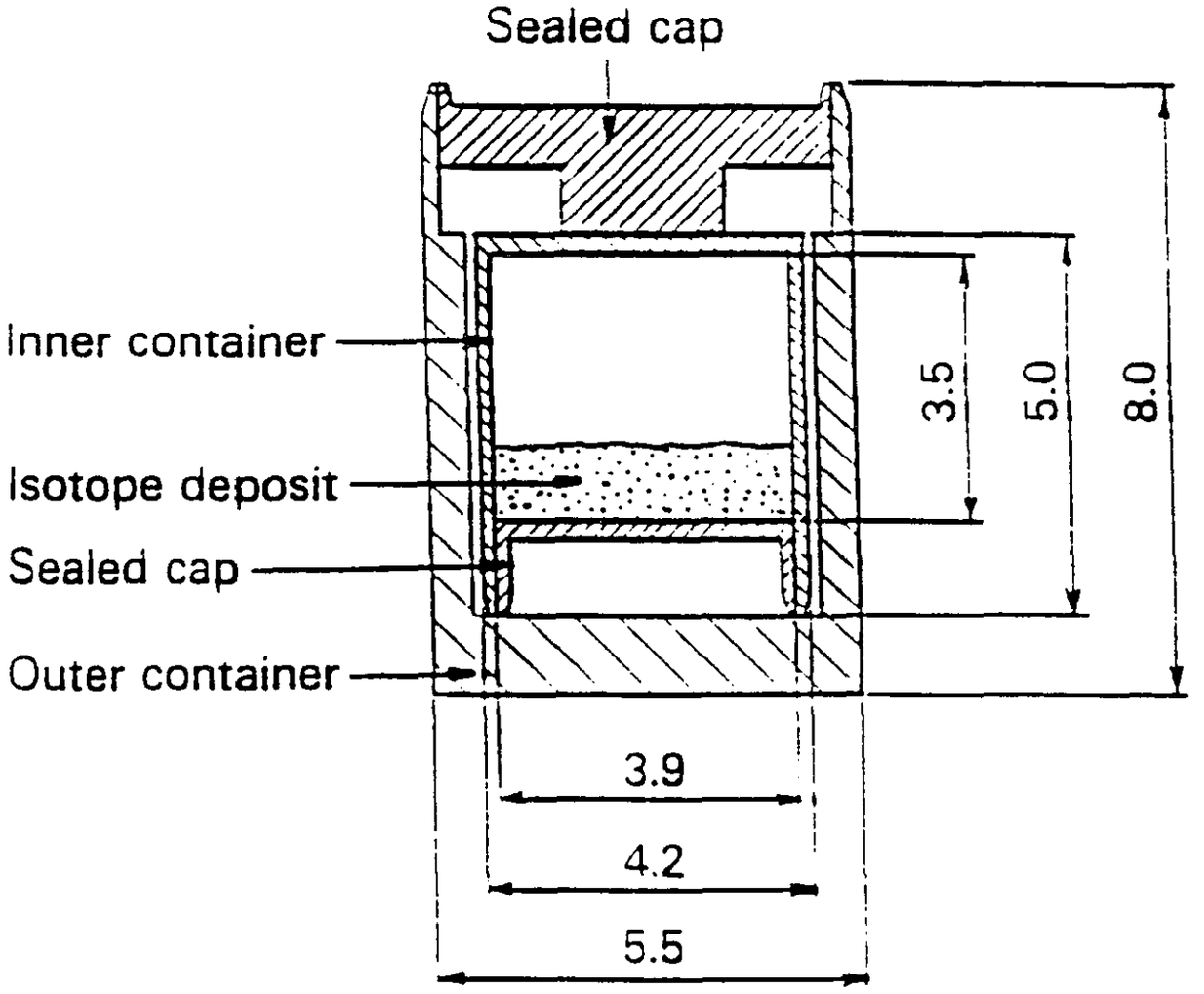
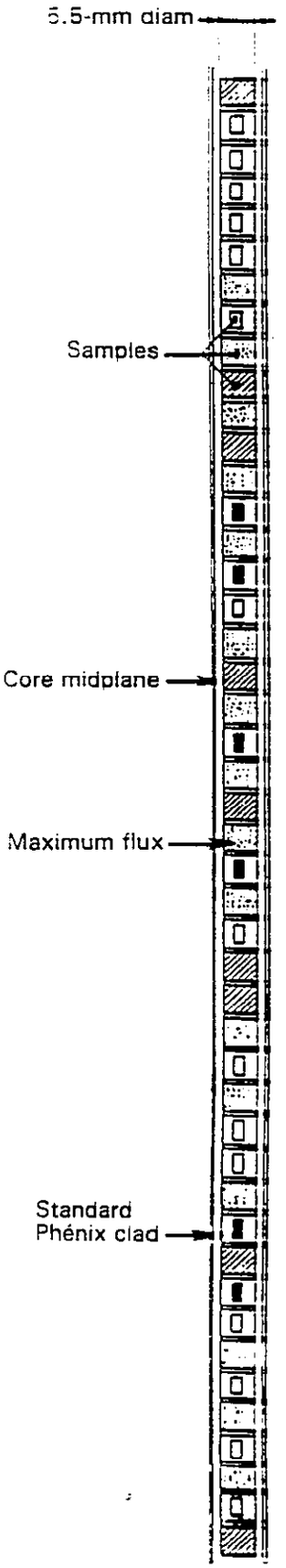


Fig. 1. Stainless steel double container for PROFIL irradiation. Dimensions are given in millimetres.



Position (mm)	Isotope	Chemical form	Mass (mg)
180	^{235}U	UO_2	22
170	^{145}Nd	Nd_2O_3	22
160	^{105}Pd	Palladium	21.7
150	^{133}Cs	CsF	23.4
140	^{101}Ru	Ruthenium	19.6
130	^{95}Mo	Molybdenum	21.5
120	^{238}Pu	PuO_2	14
110	^{97}Mo	Molybdenum	21.7
100	^{239}Pu	PuO_2	~10
90	^{235}U	UO_2	22.5
80	^{241}Am	AmO_2	10.0
70	^{238}U	UO_2	22.3
60	^{240}Pu	PuO_2	3.7
50	Natural lithium	LiF	3.8
40	^{242}Pu	PuO_2	29
30	Natural boron	Boron	1.1
20	^{149}Sm	Sm_2O_3	25.8
10	^{241}Pu	PuO_2	4.3
0	^{235}U	UO_2	24.1
−10	^{239}Pu	PuO_2	~10
−20	Natural boron	Boron	1.0
−30	^{240}Pu	PuO_2	6
−40	^{238}U	UO_2	23
−50	^{241}Pu	PuO_2	5
−60	Natural lithium	LiF	3.7
−70	^{239}Pu	PuO_2	~10
−80	^{101}Ru	Ruthenium	20
−90	^{235}U	UO_2	23.9
−100	^{238}U	UO_2	23.7
−110	^{240}Pu	PuO_2	6.3
−120	^{133}Cs	CsF	22.4
−130	^{242}Pu	PuO_2	27
−140	^{95}Mo	Molybdenum	21.9
−150	^{105}Pd	Palladium	20
−160	^{241}Pu	PuO_2	12.5
−170	Natural boron	Boron	1.3
−180	^{235}U	UO_2	24.5
−190	Natural lithium	LiF	4.2
−200	^{101}Ru	Ruthenium	19
−210	^{242}Pu	PuO_2	25
−220	^{145}Nd	Nd_2O_3	20.2
−230	^{238}Pu	PuO_2	12.6
−240	^{97}Mo	Molybdenum	19.7
−250	^{241}Am	AmO_2	13.7
−260	^{149}Sm	Sm_2O_3	22.9
−270	^{235}U	UO_2	25

Plutonium and americium

References

Uranium

Fission products

Fig. 2. PROFIL pin irradiation in Phénix.

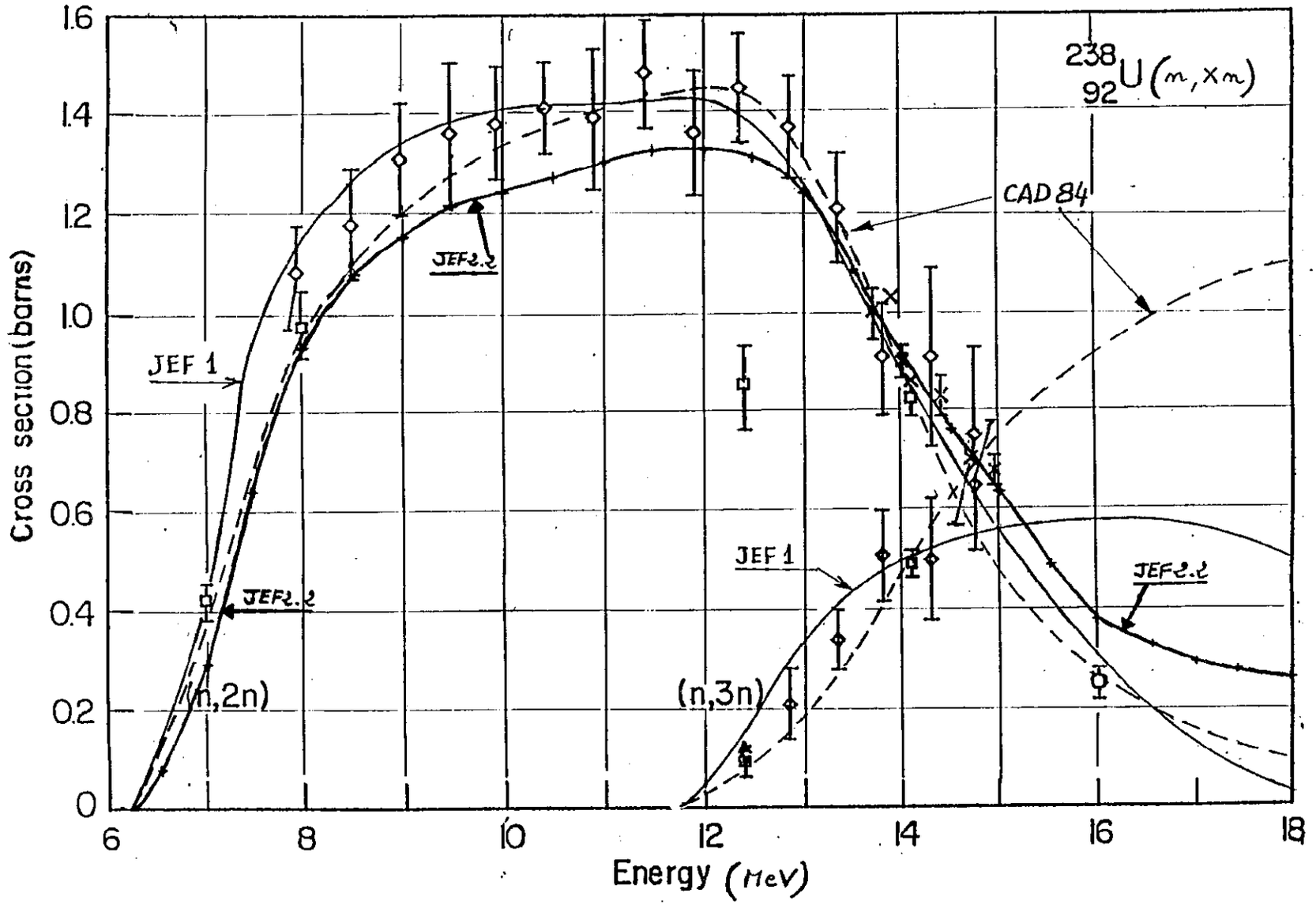


Fig. 3 - ^{238}U (n, Xn) cross-section

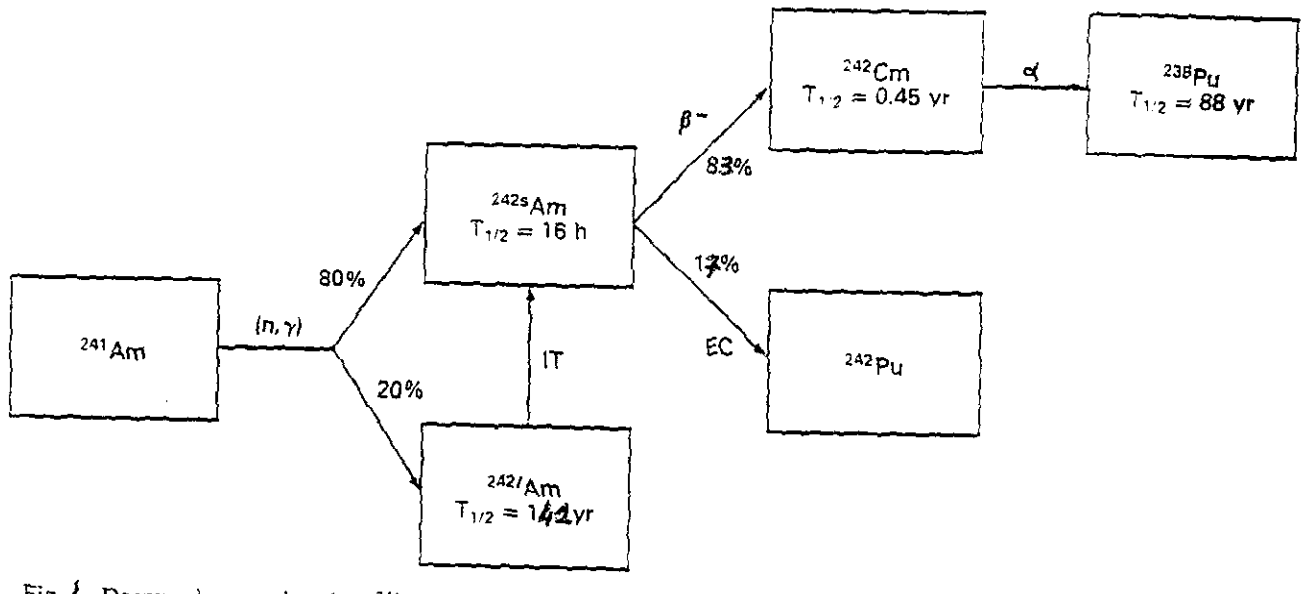


Fig. 4. Decay scheme related to ^{241}Am capture: original branching ratio values: EC = electron capture and IT = isomeric transition.

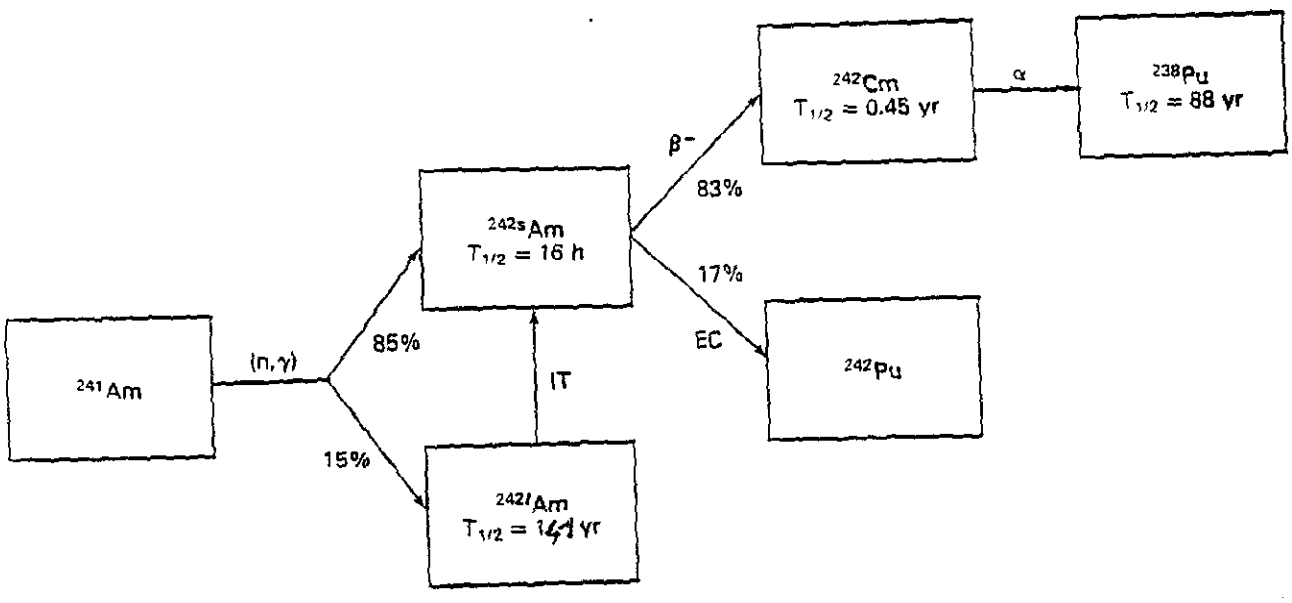


Fig. 5. Decay scheme related to ^{241}Am capture: modified branching ratio values.