Some Remarks about the ^{241}Am Capture Cross-Sections and Branching Ratio

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1. INTRODUCTION.

In the framework of the JEF2.2 benchmarking programme, some specific experiments are available to qualify cross-sections and nuclear data used in fission reactor calculations, and especially for Minor Actinide incineration studies. This paper is devoted to the nuclear data (branching ratio and decay constants) of the ^{241}Am capture cross-section. After a brief description of the physical phenomena related to the capture of one neutron by ^{241}Am and of the experimental data base we have, we present the parametric study which has been made to qualify these nuclear data.

2. DESCRIPTION OF THE EXPERIMENTS AND OF THE ^{241}Am CAPTURE RATE PHENOMENON.

Several experiments have been designed to measure the capture cross-sections of heavy nuclides involved in fission reactor design calculations:

- PROFIL 1 and 2, for fast reactors,
- ICARE S and R for epithermal reactors (High Conversion Light Water Reactors)
- SHERWOOD for standard PWRs.

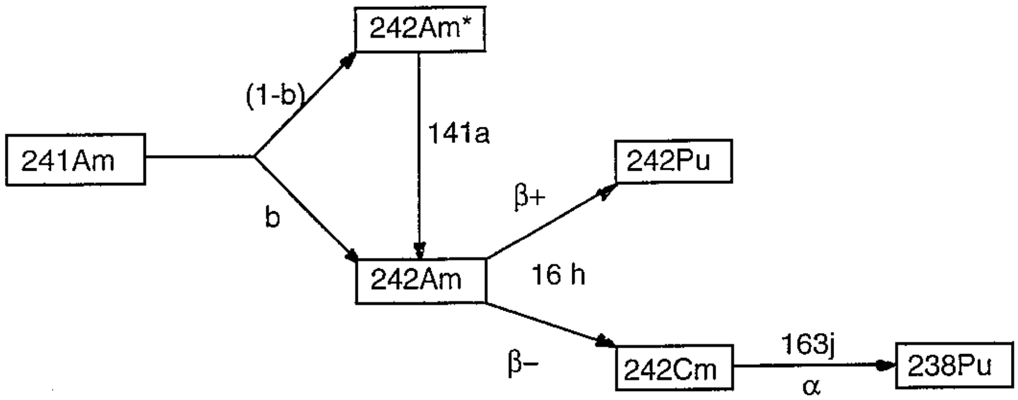
Each experiment was designed in order to obtain the fundamental mode in the zone where the experimental pin was irradiated. These experiments consisted in irradiating one or two experimental pins containing samples made of the actinide for which we wanted to measure the capture cross-section. The methodology used to derive the capture cross-section is as follow. Isotopic analyses before and after irradiation allow one to deduce the capture cross-section of the nuclide "*i*" using the formula :

$$\frac{N_{i+1}}{N_i}(\tau)-\frac{N_{i+1}}{N_i}(0)=\sigma_c^i.\tau.f(\tau) \qquad (1)$$

where :

- $N_{i+1}/N_i(\tau)$ = isotopic ratio after irradiation
- $N_{i+1}/N_i(0)$ = isotopic ratio before irradiation
- τ = fluence
- $f(\tau)$ = Calculated correction factor taking into account other reaction rates than capture
- σ_c^i : capture cross-section of the nuclide ***i***.

During each experiment, a specific sample was used to determine the capture cross-section of ^{241}Am. The major difficulty to interprete the experimental results comes from the complexity of capture of one neutron by ^{241}Am, which arises as follow :



If the radioactive decay of ^{242}Am is well-known (JEF2.2 recommends 17.3 % for the β^+ decay and 82.7 % for the β^- decay), the branching ratio "b" describing the capture phenomenon, depends of the energy of the neutron, and consequently depends of the spectrum in which the sample is irradiated.

3. PARAMETRIC STUDIES PERFORMED CONCERNING THE VALUE OF "b".

A specific analysis was performed for both PROFIL 1 and PROFIL 2, by D'ANGELO for the JEF1 nuclear data set. This study was reconducted by R. SOULE for the JEF2.2 evaluations : assuming that JEF2.2 decay constants of ^{242}Am are good, the calculations were made using b = 80 %, 85 % (PROFIL 1 and 2) and 87 % (PROFIL2) for the branching ratio. The results are summarized in the following tables :

Table 1 : PROFIL 1 Calculations.

C/E	SAMPLE 1		SAMPLE 2	
Isotopic Ratio	b = 80 %	b = 85 %	b = 80 %	b = 85 %
^{242}Am*/^{241}Am	1.390 ± 0.005	**1.042 ± 0.005**	1.425 ± 0.005	**1.069 ± 0.005**
^{242}Pu/^{241}Am	0.975 ± 0.020	**1.036 ± 0.020**	0.992 ± 0.020	**1.054 ± 0.020**
^{238}Pu/^{241}Am	0.962 ± 0.025	**1.022 ± 0.025**	0.957 ± 0.025	**1.016 ± 0.025**
^{242}Cm/^{241}Am	1.042 ± 0.010	**1.081 ± 0.010**	1.045 ± 0.010	**1.085 ± 0.010**

Table 2 : PROFIL 2 Calculations.

Isotopic Ratio	b = 80 %	b = 85 %	b = 87 %
^{242}Am*/^{241}Am	1.399 ± 0.003	**1.050 ± 0.003**	0.910 ± 0.003
^{242}Pu/^{241}Am	1.025 ± 0.020	**1.089 ± 0.020**	1.113 ± 0.020
^{238}Pu/^{241}Am	1.017 ± 0.017	**1.080 ± 0.017**	1.104 ± 0.017

These studies seem to indicate that the "best" branching ratio is around b = 0.85 : the C/E values obtained when using b = 0.85 are consistent and indicate a small over-estimation of the ^{241}Am capture cross-section in the fast energy range.

The same parametric study has been performed for the ICARE/S irradiated sample calculations. For this experiment, which is representative of a HCLWR spectrum (epithermal spectrum), the difficulty comes from the weights that we must take into account to "combine" fast, epithermal and thermal reaction rate contributions. S.F. MUGHABGHAB gives the following values in "BNL" Neutron cross-sections :

Resonance Integral : b = 86.3 ± 7.0 for ^{242}Am and (1-b) = 13.7 ± 1.4 % for ^{242}Am*
Thermal value : b = 90.8 ± 2.2 for ^{242}Am and (1-b) = 9.2 ± 1.0 % for ^{242}Am *

When weighting these branching ratios by the corresponding reaction rates, the parametric study gave a "fast" branching ratio of about b = 83 ± 5 % for the ^{242}Am and (1-b) = 17 ± 5 % for the ^{242}Am*. Furthermore, these calculations indicate an underestimation by about 20 % of the ^{241}Am capture cross-section in the epithermal energy range.

3. CONCLUSIONS

The qualification of the ^{241}Am capture cross-section is a very delicate problem which involves a good knowledge of the nuclear data related to this reaction rate, and especially of the (n,γ) branching ratio. These branching ratios are dependent on the energy of the captured neutron.
Some studies have been performed by the CEA to estimate the best values that we must use in irradiation experiment calculations and of course also in minor actinide incineration studies.
The results of both fast and epithermal experiments show a very good consistency for the branching ratio which must be used in the fast energy range : b = 85 ± 1 %.
However, in order to improve these results, a specific evaluation must be made on the ^{241}Am capture cross-section and the associated branching ratio depending on the incident neutron energy in the same way as it has been recommended in ENDF-B/V (Figure 1).

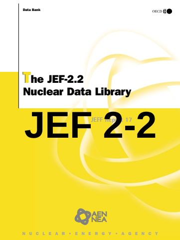


Figure 1.

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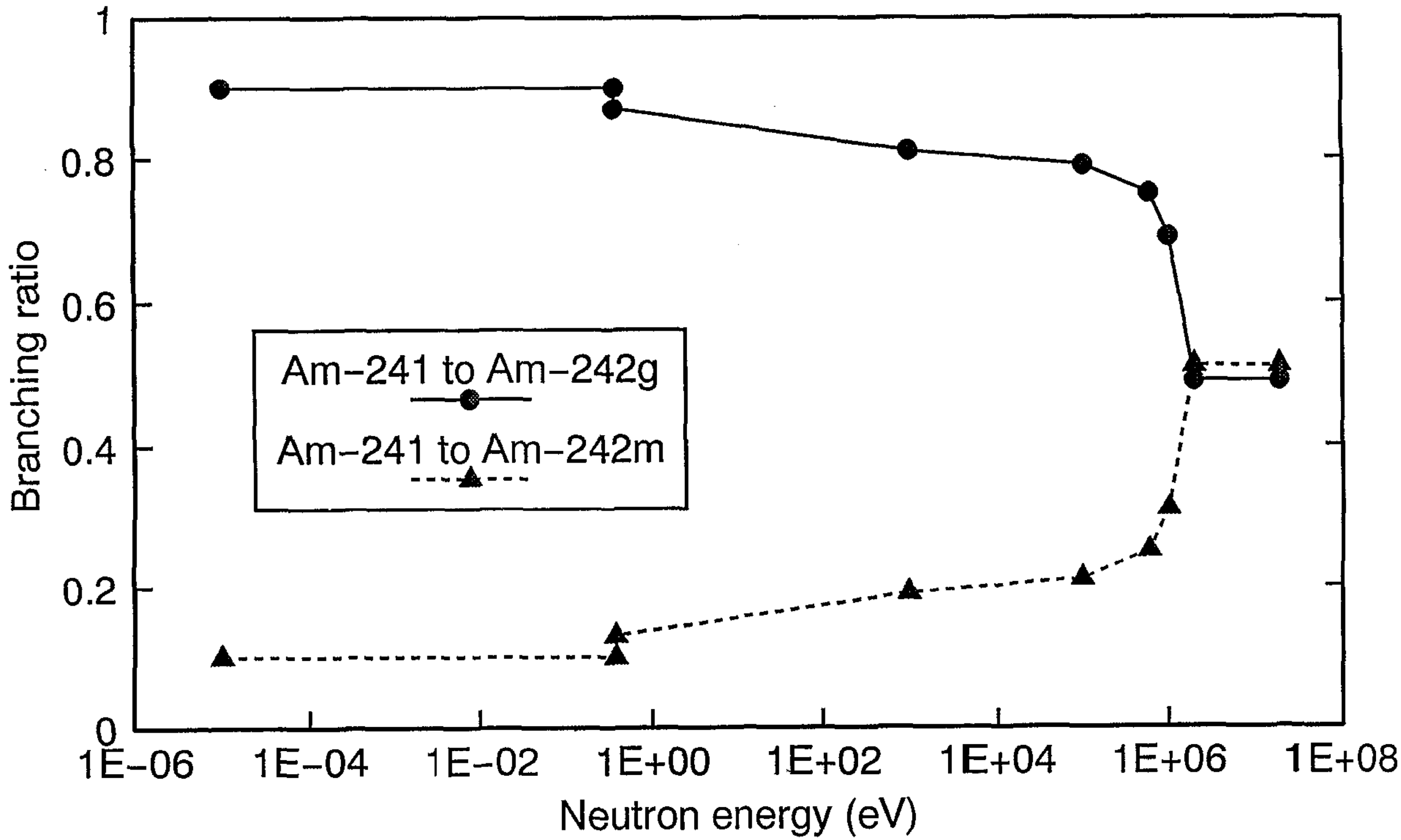

Branching ratio of Am-241 (n, γ) reaction