CONTRIBUTION TO THE VALIDATION OF JEF2 ACTINIDE NUCLEAR DATA

REANALYSIS OF FUEL IRRADIATION EXPERIMENTS IN PHENIX

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Roland SOULE, CEA, CE Cadarache, France

Introduction

A first analysis of the PROFIL and TRAPU experiments performed in Phénix reactor using the Joint Evaluated File JEF2.2 has been presented in /1/.

Some important discrepancies between Experiment and Calculations have been observed in particular :

- for the capture cross sections of Pu isotopic:
    - underestimation of ≈4% for ^{239}Pu,
    - overestimation of ≈10% for ^{240}Pu, ≈ 20% for ^{241}Pu and ≈ 15% for ^{242}Pu,
- for the capture cross section of ^{235}U (underestimation of ≈7%),
- for the capture cross section of ^{238}U (underestimation of ≈4%),
- for the (n,2n) cross section of ^{239}Pu (underestimation of ≈40%).

These integral informations have been included in a set of about ~~280~~ 350 integral data of different types (critical masses, material buckling, spectral indexes,...) belonging to a large range of spectral hardness (fast core, transmission and thermal cores) to produce, by the way of a statistical adjustment procedure, the adjusted **ERALIB1** library /2/.

In this paper we present the reanalysis of the PROFIL and TRAPU experiments based on the new adjusted ERALIB1 library. The results obtained in this way show the quality of the statistical adjustment for the major U and Pu isotopes and indications for evaluated data modifications of several minor actinides are presented.

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 ± 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		

Expériences PROFIL Comparaison Calcul/Expérience

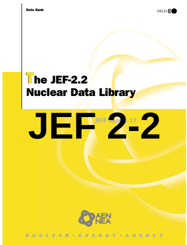
C/E values (a)

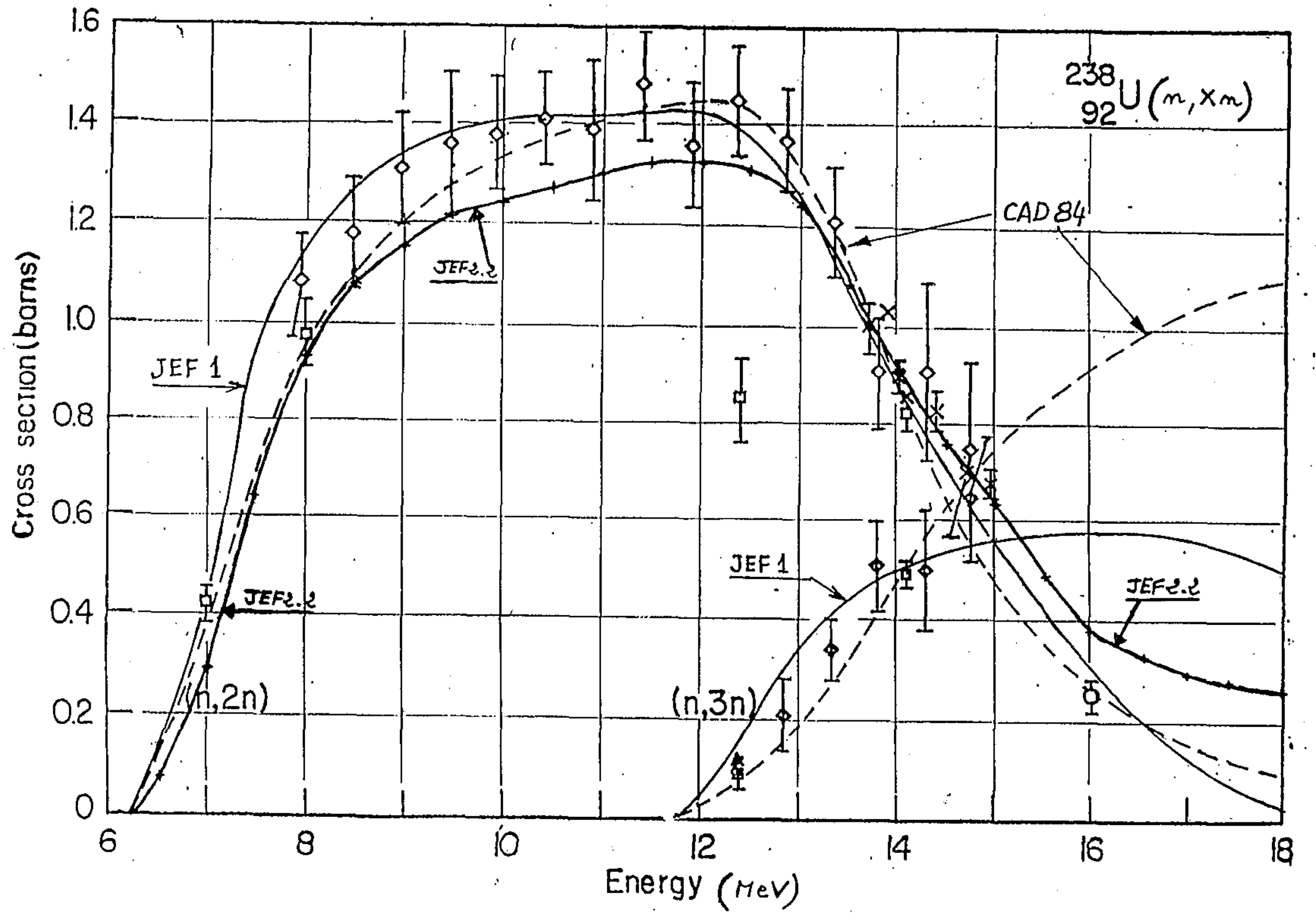
Data Type (b)	JEF2.2	ERALIB1	Uncertainties (%)
σ_c ^{233}U		0.93	± 3.0
σ_c ^{234}U		1.01	± 3.0
$\sigma_{n,2n}$ ^{234}U		0.94	± 5.0
σ_c ^{235}U	0.93	0.97	± 1.2
$\sigma_{n,2n}$ ^{235}U	0.96	0.97	± 3.5
σ_c ^{238}U	0.96	0.95	± 1.6
σ_f ^{238}U	1.00	0.99	± 1.4
σ_c ^{239}Pu	0.96	0.99	± 2.1
σ_f ^{239}Pu	1.00	1.00	± 2.2
$\sigma_{n,2n}$ ^{239}Pu	0.62	0.81	± 10.6
σ_c ^{240}Pu	1.11	1.00	± 1.6
$\sigma_{n,2n}$ ^{240}Pu	1.00	0.99	± 14.1
σ_c ^{241}Pu	1.22	1.05	± 3.6
$\sigma_{n,2n}$ ^{241}Pu	1.04	1.01	± 4.1
σ_f ^{241}Pu	0.98	0.95	± 3.3
σ_c ^{242}Pu	1.15	1.01	± 2.8
$\sigma_{n,2n}$ ^{242}Pu		0.97	± 3.5
σ_f ^{242}Pu	0.94	0.92	± 8.6
σ_c ^{238}Pu	0.95	0.96	± 2.8
σ_c ^{241}Am	1.04	1.04	± 1.0*
σ_c ^{243}Am	0.96	0.96	± 5.0
σ_c ^{237}Np	0.97	0.96	± 3.6
$\sigma_{n,2n}$ ^{237}Np	1.20	1.20	± 4.7

(a) C/E values are in absolute values
(b) All the results are average reaction rate ratios (spectral indexes) related to the ^{235}U fission rate

Expérience TRAPU Comparaison Calcul/Expérience

^{238}U = 100.	TRAPU-1			TRAPU-2			TRAPU-3		
	JEF2.2	ERALIB1	Unc (%)	JEF2.2	ERALIB1	Unc (%)	JEF2.2	ERALIB1	Unc (%)
^{234}U	0.98	0.98	± 2.5	1.00	1.00	± 1.3	1.04	1.04	± 1.0
^{235}U	1.01	1.01	± 0.3	1.03	1.03	± 0.2	1.03	1.03	± 0.2
^{236}U	0.91	0.94	± 0.5	0.93	0.96	± 0.4	0.93	0.96	± 0.3
^{237}Np	0.78	0.78	± 6.8	0.77	0.78	± 3.3	0.75	0.75	± 3.2
^{238}Pu	0.99	0.99	± 0.9	1.01	1.01	± 0.4	1.01	1.01	± 0.4
^{239}Pu	1.02	1.01	± 0.4	1.00	1.00	± 0.3	1.00	1.00	± 0.3
^{240}Pu	0.99	1.00	± 0.4	0.97	0.98	± 0.3	0.99	1.00	± 0.3
^{241}Pu	1.04	1.01	± 0.4	1.01	1.00	± 0.3	1.03	1.01	± 0.3
^{242}Pu	1.11	1.04	± 0.5	1.05	1.02	± 0.4	1.03	1.01	± 0.3
^{241}Am	0.98	0.97	± 3.0	0.98	0.98	± 3.6	0.98	0.98	± 2.1
^{242m}Am	1.03	1.01	± 3.6	1.06	1.05	± 4.0	1.02	1.01	± 2.5
^{243}Am	1.07	0.90	± 3.6	1.03	0.88	± 4.0	1.06	0.92	± 2.5
^{242}Cm	1.02	1.00	± 2.4	0.99	0.98	± 2.6	0.99	0.98	± 2.1
^{243}Cm				0.71	0.68	± 2.7	0.70	0.68	± 2.6
^{244}Cm	0.98	0.83	± 2.0	1.09	0.92	± 2.2	1.10	0.95	± 1.7
^{148}Nd	1.01	1.00	± 0.3	1.00	0.99	± 0.2	1.00	0.99	± 0.2





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

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