DIRECTION DE L'ENERGIE NUCLÉAIRE
DIRECTION DU CENTRE DE CADARACHE
DÉPARTEMENT D'ÉTUDES DES RÉACTEURS
SERVICE DE PHYSIQUE DES RÉACTEURS ET DU CYCLE
LABORATOIRE D'ÉTUDES DE PHYSIQUE

NT	**SPRC/LEPH – 05/204**	**0**	**5H64**	**A-DONUT-02-01**		**1/23**
NATURE	CHRONO UNITE	INDICE	UNITE	EOTP	CLASSEMENT UNITE	PAGE

Note Technique

TITRE : **BESOINS EN DONNÉES NUCLÉAIRES POUR LES ACTINIDES PAR COMPARAISON AVEC DES EXPÉRIENCES DE COMBUSTIBLES ET D'ECHANTILLONS IRRADIÉS**

TITLE : **NUCLEAR DATA NEEDS FOR ACTINIDES BY COMPARISON WITH POST IRRADIATION EXPERIMENTS**

AUTEUR(S) : G. Noguere, E. Dupont, J. Tommasi, D. Bernard

RÉSUMÉ : L'interprétation des expériences à haut burnup réalisées auprès des réacteurs CRUAS et DAMPIERRE et la nouvelle analyse des expériences PROFIL et PROFIL-2 (irradiations d'échantillons) réalisées dans le réacteur rapide PHENIX ont confirmé ou délivré de nouvelles tendances intégrales sur les réactions (n,γ) et (n,2n) de certains actinides importants pour l'industrie nucléaire. Les résultats obtenus fournissent l'opportunité de proposer des mesures et des évaluations dont l'objectif sera d'améliorer les données nucléaires et d'assurer une meilleure cohérence physique entre le domaine des résonances résolues et le "continuum" (hautes énergies). Les tendances intégrales pour ^{233,234,238}U, ^{238,239,240,241,242}Pu, ^{232}Th, ^{241}Am, ^{237}Np et ^{244}Cm calculées avec JEFF-3.0 sont comparées aux données différentielles EXFOR.

ABSTRACT : The interpretation of high-burnup irradiation experiments, carried out in the CRUAS and DAMPIERRE power reactors, and the new analysis of the PROFIL and PROFIL-2 sample irradiation experiments, carried out in the fast reactor PHENIX, confirmed or delivered new integral trends on (n,γ) and (n,2n) reactions of specific actinides of interest for the nuclear industry. These results provide the opportunity to propose experimental and evaluation activities with the aim of improving nuclear data and ensuring a better physics consistency between the resolved resonance range and the "continuum" region. Integral trends for ^{233,234,238}U, ^{238,239,240,241,242}Pu, ^{232}Th, ^{241}Am, ^{237}Np and ^{244}Cm calculated with JEFF-3.0 are compared with the EXFOR data.

MOTS CLÉS : DONNEES NUCLEAIRES, ACTINIDES, PROFIL, PHENIX, CRUAS, DAMPIERRE

	Rédacteur	Vérificateur	A.Q.	Approbateur
Nom et visa	G. Noguere	C. De Saint Jean	A. Bernard	Chef du LEPH J.M. Ruggieri
Date	28/09/2005			

Cadre de réalisation du document
DOB .... DSOE
Domaine .... Simulation
Projet .... DONUT
Contrat .... collaboration JEFF

CLASSIFICATION				
DR	CC	CD	SD	Sans
				■

INDICE	OBJET DES RÉVISIONS	DATE	N° CHRONO
0	EMISSION INITIALE	28/09/2005	05/204

Table of Contents

1 Post-Irradiation Experiments 4

2 Low energy range of the (n,γ) Reactions 7
- 2.1 Microscopic Data 7
- 2.2 Plutonium 9
- 2.3 Americium 10
- 2.4 Neptunium 11
- 2.5 Curium 11

3 High Energy Range of the (n,γ) Reactions 12
- 3.1 Microscopic Data 12
- 3.2 Plutonium 14
- 3.3 Americium 15
- 3.4 Neptunium 15
- 3.5 Thorium 16
- 3.6 Curium 16

4 (n,2n) Reactions 17
- 4.1 Uranium 17
- 4.2 Plutonium 18

5 Measurement and Evaluation Issue 18
- 5.1 Uranium 18
- 5.2 Plutonium 19
- 5.3 Neptunium 19
- 5.4 Americium and Curium 20

6 Conclusions 20

References 21

Nuclear Data Needs for Actinides by Comparison with Post Irradiation Experiments

The JEFF project made heavy use of integral data, such as Post-Irradiation Experiments (PIE) carried out in thermal reactors [1], to improve specific evaluations of interest for the nuclear industry. By contrast, little improvements in the high energy range (Unresolved Resonance Range) are based on PIE performed in fast spectra. In this context, the latest interpretations with the JEFF-3.0 library of PIE carried out in the CRUAS and DAMPIERRE PWRs [2] and in the fast reactor PHENIX (PROFIL and PROFIL-2 experiments) [3] provide the opportunity to propose evaluation activities which aim of improving nuclear data and ensuring a physics consistency between the resolved resonance range and the high energy range.

The focus of the present study is to check the evaluations of specific reactions - (n,γ) and (n,2n) - for the main actinides of interest for the nuclear industry. For practical purpose, the discussion is restricted to isotopes of interest for the PROFIL experiments ($^{233,234,238}U$, $^{238,239,240,241,242}Pu$, ^{232}Th, ^{241}Am, ^{237}Np and ^{244}Cm). In this work, integral trends provided by APOLLO-2.5 and ERANOS-2.0 based JEFF-3.0 calculations are compared to the microscopic trends provided by the differential data reported in the EXFOR database and by the evaluations available in the latest version of the European library JEFF-3.1.

1 Post-Irradiation Experiments

Integral trends on the main reactor relevant actinides are presented in Tables 1 and 2 in term of C/E values. These results were obtained from the interpretation of independent Post-Irradiation Experiments with the JEFF-3.0 library.

The trends in the epi-thermal energy range are based on high-burnup irradiation experiments carried out in the CRUAS and DAMPIERRE power reactor [2]. The fuel inventory prediction was calculated with the deterministic APOLLO-2.5 code.

For the fast energy range, C/E results were obtained from the interpretation of the PROFIL (PROFIL and PROFIL-2) sample irradiation experiments performed in the PHENIX demonstration fast reactor (respectively in 1974 and 1979). The PROFIL experiments have been especially designed to collect accurate integral data. These experiments contained rods with a large number of samples ($\sim$ 130) containing almost pure actinides and fission products isotopes, separated into small quantities of several milligrams. The analysis of the data was performed with the ERANOS-2.0 code system [3, 4].

After irradiation, the composition of the spent fuel (CRUAS and DAMPIERRE experiments) or samples (PROFIL experiments) is determined by Isotope Dilution Inductively Coupled Plasma Mass Spectroscopy (ID-ICP-MS). The physical variables of interest for the interpretation are the

Table 1 - Fuel inventory prediction in high-burned UOx and MOx spent fuel [2]. The C/E-1 (%) values were calculated with the APOLLO-2.5 code by using the JEFF-3.0 neutron library.

Isotopic Ratio	CRUAS (UOx spent fuel)		DAMPIERRE (MOx spent fuel)	
	C/E-1 (%)	C/E-1	C/E-1 (%)	C/E-1
	56.8 GWd/tHM	69.2 GWd/tHM	52.6 GWd/tHM	58.9 GWd/tHM
$^{234}\text{U}/^{238}\text{U}$	-2.6 ± 2.0	-4.3 ± 2.4	-1.1 ± 2.0	-0.7 ± 2.2
$^{235}\text{U}/^{238}\text{U}$	-2.5 ± 6.7	2.3 ± 7.0	-0.2 ± 2.3	-0.8 ± 2.5
$^{236}\text{U}/^{238}\text{U}$	-1.4 ± 1.1	-2.1 ± 1.2	-3.1 ± 1.3	-2.7 ± 1.3
$^{237}\text{Np}/^{238}\text{U}$	1.7 ± 2.8	-0.5 ± 2.6	-2.2 ± 2.6	-1.7 ± 2.5
$^{238}\text{Pu}/^{238}\text{U}$	0.5 ± 4.2	-0.8 ± 3.6	-2.7 ± 1.4	-1.4 ± 1.9
$^{239}\text{Pu}/^{238}\text{U}$	7.4 ± 2.8	9.9 ± 3.4	1.7 ± 2.2	4.8 ± 2.2
$^{240}\text{Pu}/^{238}\text{U}$	2.6 ± 1.4	3.5 ± 0.9	1.1 ± 0.9	3.3 ± 1.0
$^{241}\text{Pu}/^{238}\text{U}$	4.1 ± 2.4	6.4 ± 2.7	0.0 ± 0.9	1.6 ± 1.3
$^{242}\text{Pu}/^{238}\text{U}$	2.3 ± 4.1	-0.8 ± 2.9	2.1 ± 1.3	3.0 ± 1.3
$^{241}\text{Am}/^{238}\text{U}$	29.3 ± 11.6	17.4 ± 6.1	15.2 ± 3.9	16.5 ± 3.7
$^{242m}\text{Am}/^{238}\text{U}$	-9.1 ± 7.1	-12.5 ± 6.8	-18.9 ± 2.6	-20.2 ± 3.5
$^{243}\text{Am}/^{238}\text{U}$	7.8 ± 5.5	4.4 ± 4.0	3.8 ± 2.0	5.7 ± 1.7
$^{243}\text{Cm}/^{238}\text{U}$		-16.1 ± 6.5	15.2 ± 1.8	-15.8 ± 2.3
$^{244}\text{Cm}/^{238}\text{U}$	6.3 ± 9.9	1.5 ± 6.6	-2.8 ± 3.8	-1.0 ± 3.4
$^{245}\text{Cm}/^{238}\text{U}$		17.8 ± 9.3	4.8 ± 4.2	10.0 ± 3.7
$^{246}\text{Cm}/^{238}\text{U}$		-8.3 ±10.0	-12.5 ± 7.5	-8.7 ± 6.9
$^{247}\text{Cm}/^{238}\text{U}$		4.6 ±12.2	-4.2 ± 9.2	2.7 ± 8.5

concentration ($n_{\alpha,i}$) and ($n_{\alpha,f}$) of the isotope α at the beginning and at the end of the irradiation period. In order to reach an optimal exploitation of the results with a reliable uncertainty analysis, the sensitivity coefficients S_{α,σ_β} of the final concentration of nuclide α to the cross sections of nuclide β must be known. In a first approximation, the evaluation of the sensitivities may be seen as the first derivative of a given concentration at a given time with respect to the cross sections of interest. The variation of the concentration $\Delta n_\alpha = n_{\alpha,i} - n_{\alpha,f}$ with respect to the cross section σ_β may be expressed as follows:

$$\frac{\delta(\Delta n_\alpha)}{\Delta n_\alpha} = S_{\alpha,\sigma_\beta}\,\Delta(\sigma_\beta) \qquad \text{with} \qquad \Delta(\sigma_\beta) = \frac{\delta\sigma_\beta}{\sigma_\beta} \tag{1}$$

Example of sensitivity coefficients (S_{α,σ_β}) calculated by J. Tommasi [5] for the interpretation of the PROFIL-2 experiment are presented in Figure 1 in a 33 group energy mesh. In practice, the composition of the samples is expressed in term of isotopic ratios $E = n_{\alpha,f}/n_{\beta,f}$ and the variation of the concentration may be calculated as follows:

$$\frac{\delta\left(\Delta\left[\frac{n_\alpha}{n_\beta}\right]\right)}{\Delta\left[\frac{n_\alpha}{n_\beta}\right]} \equiv 1 - \frac{1}{C/E} \tag{2}$$

in which C/E stands for the ratio between the calculated and the experimental isotopic ratios.

Table 2 - Results of the interpretation of the PROFIL and PROFIL-2 sample irradiation experiments performed with the ERANOS-2.0 code [4]. The C/E-1 (%) values were obtained with the JEFF-3.0 neutron library.

Isotopic Ratio	Sample	PROFIL C/E-1 (%)	PROFIL-2 C/E-1 (%)	Weighted Average
^{234}U/^{235}U	^{235}U	-2.1 ± 1.6	0.0 ± 0.4	-1.0 ± 1.5
^{235}U/^{238}U	^{235}U	1.1 ± 4.8	0.0 ± 0.4	0.6 ± 2.4
^{236}U/^{235}U	^{234}U		0.5 ± 0.4	0.4 ± 0.1
	^{235}U	0.4 ± 0.2	0.4 ± 0.1	
^{239}Pu/^{238}U	^{235}U	0.8 ± 0.8		1.4 ± 0.8
	^{238}U	1.1 ± 0.3	2.3 ± 0.2	
^{237}Np/^{238}U	^{238}U		-6.7 ± 2.9	-6.7 ± 2.9
^{240}Pu/^{239}Pu	^{238}U	0.1 ± 0.1	−2.2 ± 0.6	-1.6 ± 1.2
	^{239}Pu	−2.1 ± 0.2	−2.3 ± 0.1	
^{238}Pu/^{239}Pu	^{239}Pu	-19.1 ± 4.1	-26.7 ± 12.6	-22.9 ± 6.6
^{239}Pu/^{238}Pu	^{238}U	1.9 ± 0.8	3.5 ± 0.1	2.7 ± 1.1
^{241}Pu/^{240}Pu	^{239}Pu	4.4 ± 1.1	3.6 ± 0.4	4.3 ± 0.7
	^{240}Pu	5.2 ± 0.5	3.9 ± 0.6	
^{242}Pu/^{241}Pu	^{240}Pu	16.6 ± 2.7	11.5 ± 2.9	12.4 ± 3.7
	^{241}Pu	9.3 ± 0.7		
^{243}Am/^{242}Pu	^{242}Pu	20.9 ± 1.8	14.7 ± 2.8	17.8 ± 4.4
^{238}Pu/^{237}Np	^{237}Np		-6.0 ± 1.3	-6.0 ± 1.3
^{242m}Am/^{241}Am	^{241}Am	8.7 ± 1.9	7.6 ± 0.2	8.2 ± 1.0
^{238}Pu/^{241}Am	^{241}Am	5.5 ± 1.6	10.7 ± 1.9	7.3 ± 2.7
^{242}Pu/^{241}Am		6.3 ± 1.5	9.7 ± 2.6	
^{242}Cm/^{241}Am		4.5 ± 1.2		
^{244}Cm/^{243}Am	^{242}Pu	0.0 ± 6.7		0.0 ± 6.7
^{245}Cm/^{244}Cm	^{244}Cm		-2.6 ± 0.3	-2.6 ± 0.3
^{233}U/^{232}Th	^{232}Th		-16.4 ± 7.6	-16.4 ± 7.6
^{234}U/^{233}U	^{233}U		-8.5 ± 0.1	-8.5 ± 0.1
^{235}U/^{234}U	^{233}U		2.8 ± 1.9	2.9 ± 0.9
	^{234}U		3.1 ± 0.2	

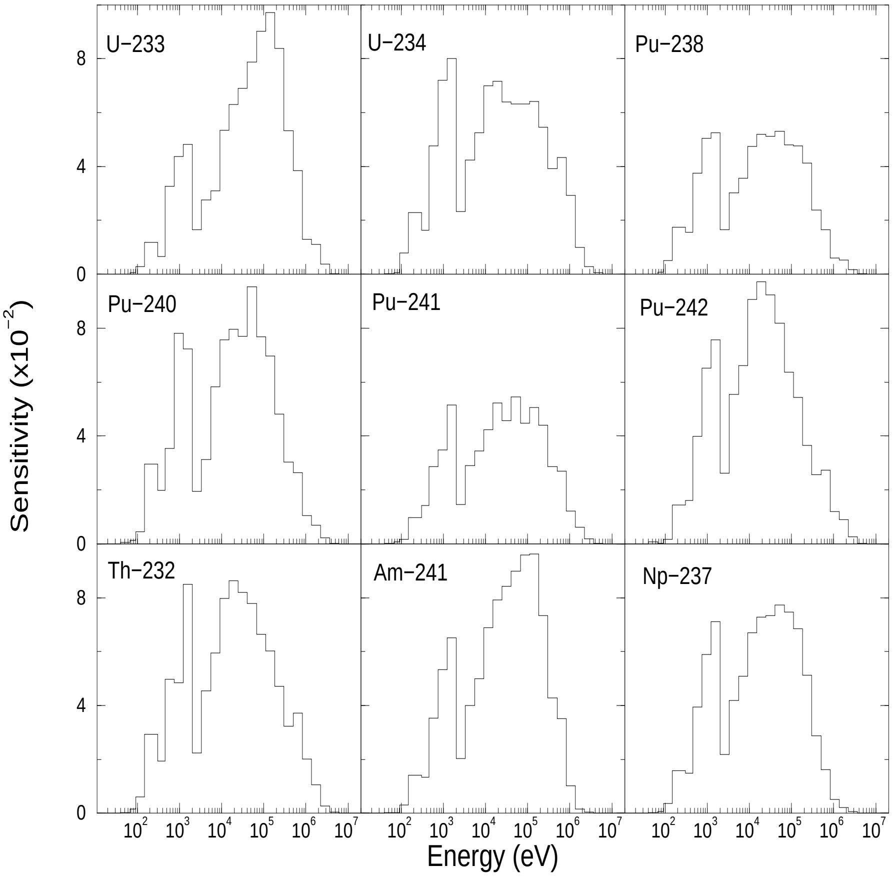

Figure 1 - Examples of sensitivity profiles (PROFIL-2 experiment) of the C/E results to the capture cross section calculated with the ERANOS-2.0 system [5].

2 Low energy range of the (n,γ) Reactions

On the basis of the conclusions of the JEF-2.2 validation studies [6], extensive works have been done to improve the modeling of the low neutron energy cross section of some reactor relevant isotopes. In this work, we report microscopic trends on the capture cross section of isotopes of interest for the uranium and thorium fuel cycle (^{233}U, ^{234}U, ^{238}Pu, ^{240}Pu, ^{241}Pu, ^{242}Pu, ^{241}Am, ^{237}Np, ^{244}Cm and ^{232}Th (n,γ) reactions). For comparison, integral trends provided by the interpretation with JEFF-3.0 of the CRUAS and DAMPIERRE experiments [2] are given in Table 1.

2.1 Microscopic Data

The number of relevant experimental capture data reported in EXFOR are rather limited. Three groups of isotopes can be distinguished.

- There is no experimental capture data for the low neutron energy range – first resonance(s) – of the ^{234}U, ^{238}Pu, ^{240}Pu, ^{242}Pu, ^{241}Am and ^{232}Th (n,γ) reactions. For these isotopes, European evaluations (JEFF-3.0 and JEFF-3.1) are compared with the experimental total cross sections available in EXFOR (Figure 2).

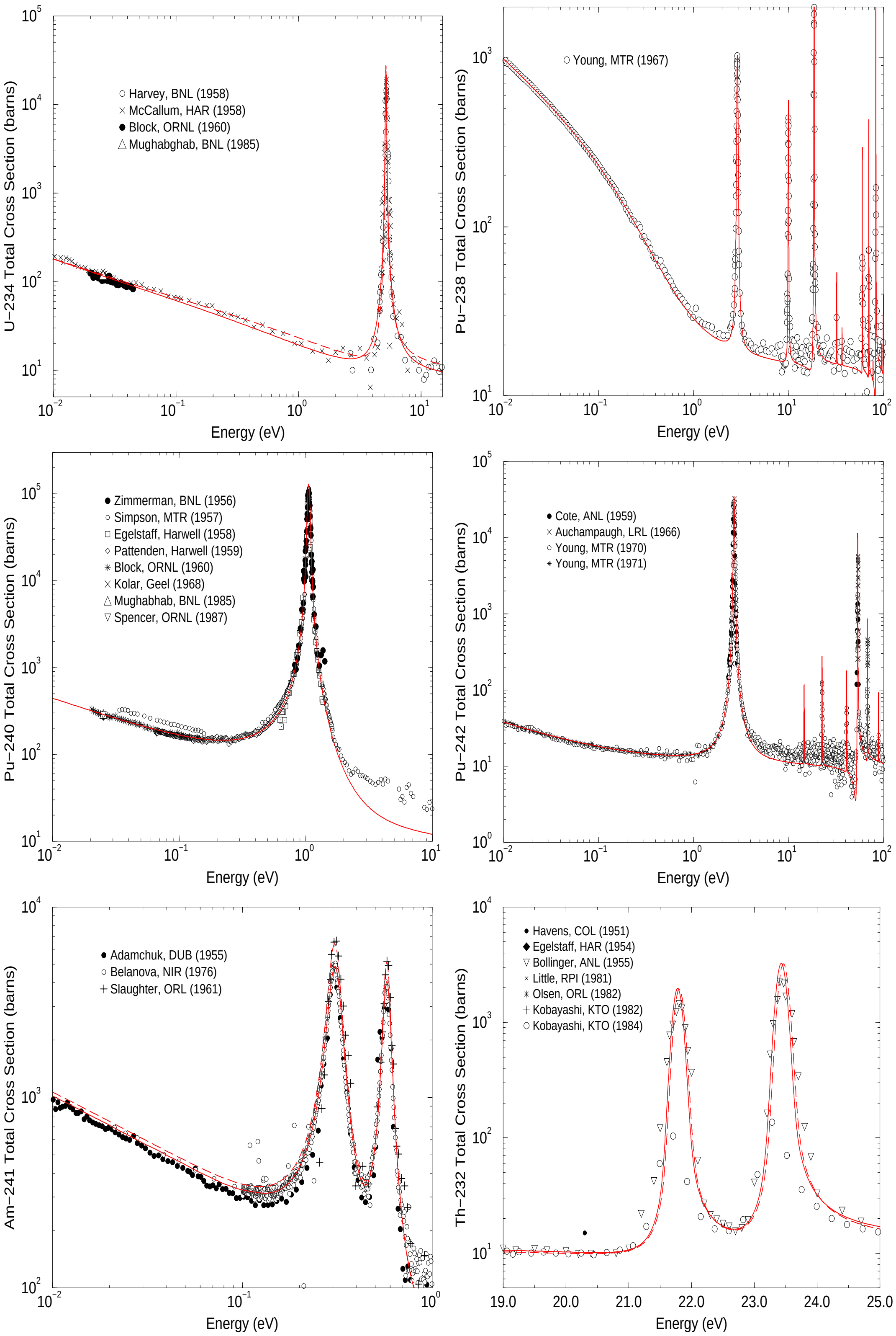

Figure 2 - Low neutron energy of the ^{234}U, ^{238,240,242}Pu, ^{241}Am and ^{232}Th total cross sections. The JEFF-3.0 (solid line) and JEFF-3.1 (dashed line) library are compared with the experimental data available in the EXFOR database (for ^{238,240,242}Pu, the JEFF-3.1 and JEFF-3.0 curves are similar).

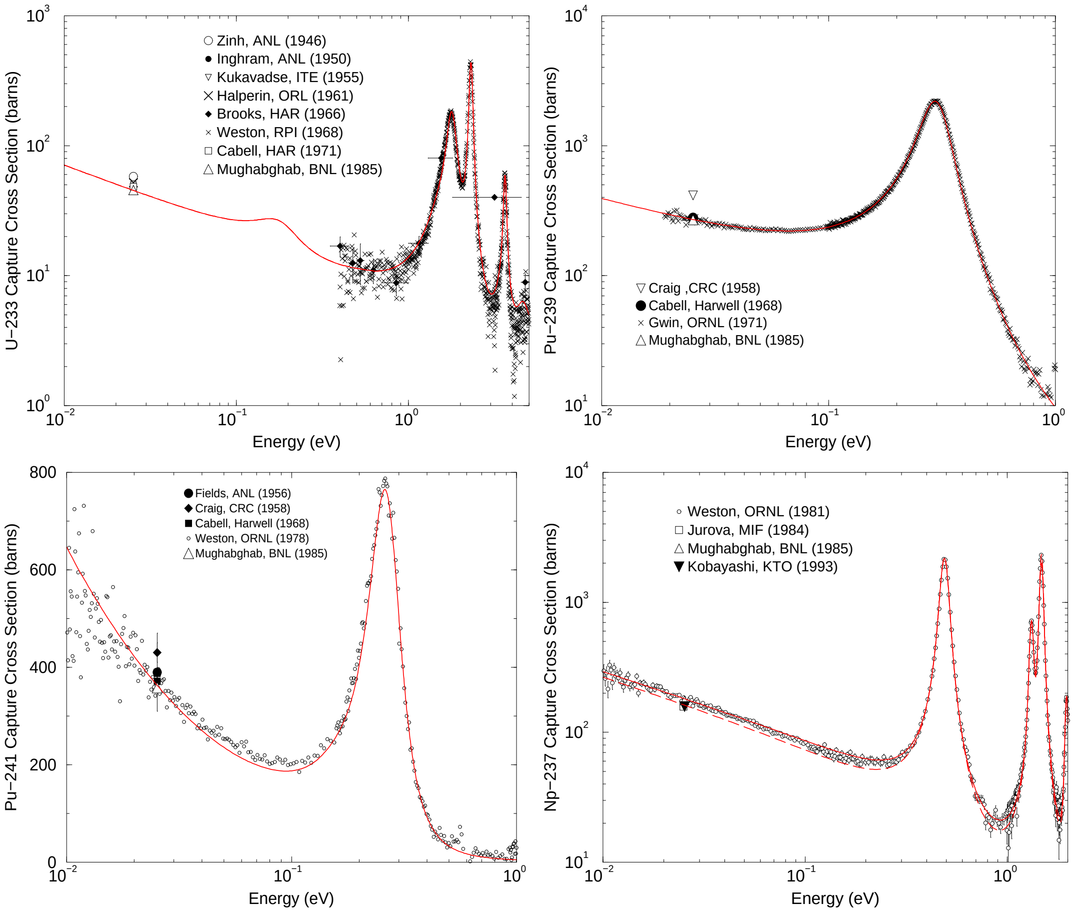

Figure 3 - Low neutron energy of the ^{233}U, ^{239,241}Pu and ^{237}Np capture cross sections. The JEFF-3.0 (solid line) and JEFF-3.1 (dashed line) library are compared with the experimental data available in the EXFOR database (for ^{233}U, ^{239}Pu and ^{241}Pu the JEFF-3.1 and JEFF-3.0 curves are similar).

- Few capture cross sections have been reported in the EXFOR database covering the low neutron energy range of the ^{233}U, ^{239}Pu, ^{241}Pu and ^{237}Np (n,γ) reactions (Figure 3). However, for each isotope, a single one capture data set describe the first resonance(s). These measurements have been performed or co-supervised by Weston.

- For ^{244}Cm, neither experimental capture nor total cross sections covering the first resonances have been reported in the database. Very little experimental work have been performed on these isotopes because of sample procurement and handling problems. The low neutron energy capture cross sections, available in the JEFF-3.0 and JEFF-3.1 libraries, are shown in Figure 4.

2.2 Plutonium

A closer inspection of the average results reported in Table 1 indicate a slight overestimation of the ^{239}Pu build-up. Owing to the multiple source of uncertainties, this trend has to be taken with care. In JEFF-3.1, the decrease of the ^{238}U(n,γ) resonance integral by 0.7% [7] will reduce this remaining overestimation of the ^{239}Pu prediction.

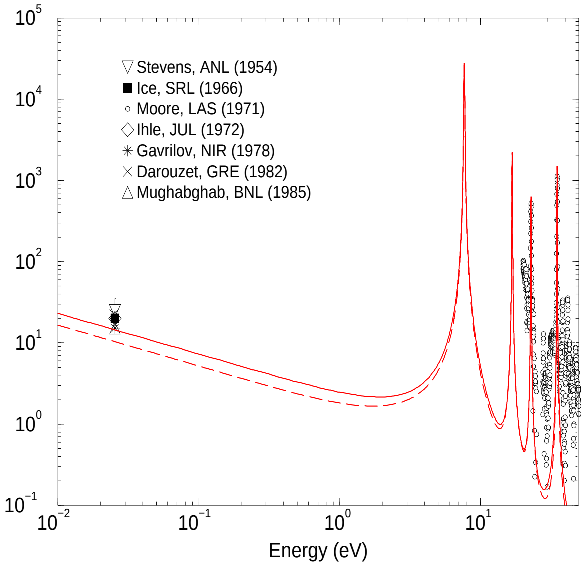

Figure 4 - ^{244}Cm capture cross sections recommended in JEFF-3.0 (solid line) and JEFF-3.1 (dashed line). (n,γ) or (n,tot) data covering the first resonances have never been reported in the EXFOR database.

The low energy range of the ^{240,241}Pu neutron cross sections have been improved for JEFF-3.0 by accounting for integral trends given by PIE performed in French pressurized water reactors. Evaluations of the thermal and epi-thermal energy range have been done in collaboration with the Oak Ridge National Laboratory (ORNL) [8, 9]. As a result, related isotopic ratios are accurately predicted, and C/E discrepancies are lower than 3%. Thanks to the increase of the capture area of the strong s-wave at 0.26 eV of ^{241}Pu, the longstanding ^{242}Pu underestimation in JEF-2.2 based calculations is significantly reduced with JEFF-3.0.

For ^{242}Pu and ^{238}Pu, the evaluations of the epi-thermal energy range available in JEFF-3.0 (=JEFF-3.1) date back respectively to 1987 and 1989. Below 10 eV, the modeling of the ^{238}Pu cross sections is based on the total cross section measured by Young in 1967 [10]. For ^{242}Pu, the evaluation of the Resolved Resonance Range takes into account neutron and radiation widths compiled in Reference [11]. The poor agreement between the European evaluation and the EXFOR data is shown in Figure 5.

2.3 Americium

For ^{241}Am, the PIE results based on JEFF-3.0 calculations predict a strong underestimation of about 16% of its effective capture cross section. In order to reduce the C/E discrepancy reported in Table 1, the thermal and epi-thermal energy ranges have been revisited at the CEA/DEN of Cadarache [12]. The resonance parameters have been improved by considering the total cross section measured by Slaughter [13] (Figure 2). Parallel increase of the ^{241}Am isomeric ratio to ^{242m}Am has been performed to improve the build-up prediction of ^{242m}Am. These modifications have been included in the latest version of the European library (JEFF-3.1).

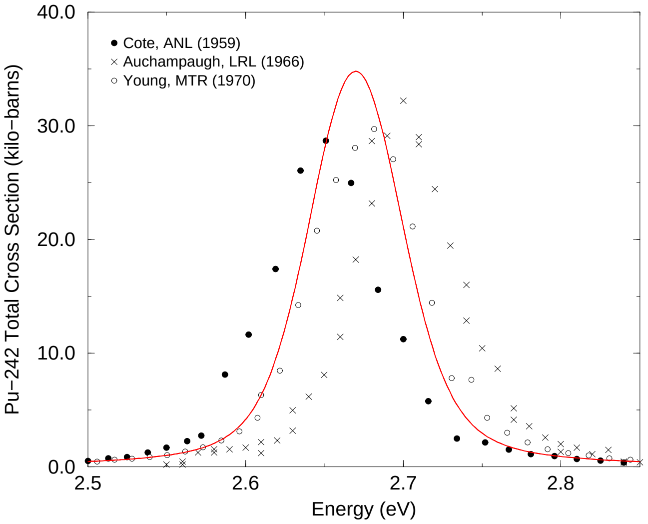

Figure 5 - Comparison between the EXFOR data and the ^{242}Pu total cross sections (solid line) of JEFF3.0 (=JEFF-3.1).

2.4 Neptunium

Integral results do not stress any significant problem in the modeling of the low energy range of the ^{237}Np(n,γ) cross section. The concentration in MOx spent fuel is perfectly calculated with the APOLLO-2.5 code and JEFF-3.0 data.

In JEFF-3.1, cross sections in the thermal and sub-thermal energy ranges have been modified (Figure 3). The JEFF-3.0 has been replaced by the Japanese evaluation. The latter accounts for a thermal capture cross section of about 161.7 b, which is consistent with the values reported by Jurova (158±4 b) [14] and Kobayashi (158 ± 3 b) [15]. By contrast, these thermal capture cross sections are $\sim$ 12% lower than the JEFF-3.0 value ($\sim$ 181 b), which was confirmed by the Mini-Inca experiment (180±5 b) [16]. Specific studies are needed to explain the recurring discrepancies between European and Japanese experimental thermal capture cross sections.

2.5 Curium

For the curium isotopes, the evaluations in JEFF-3.0 and those proposed in JEFF-3.1 remain a patchwork of a large variety of experimental or evaluated works. Because of its radiotoxicity, direct measurements of the ^{244}Cm (18.1 years) cross sections are extremely difficult. Reliable integral trends are suitable to perform its evaluation.

For JEFF-3.1, the ENDF/B-VI evaluation has been selected to replace the JEFF-3.0 evaluation (Figure 4). In the American work, the parameters of the bound level and of the first resonance were modified within reasonable experimental limits to provide a satisfactory agreement with integral data and production studies.

In a general way, the nuclear properties at low energies of the main actinides of interest for the thermal reactors have been significantly improved. However, for specific isotopes (curium and americium), accurate experimental data are still required to improve the Reich-Moore description of the first resonances.

3 High Energy Range of the (n,γ) Reactions

In this section, we discuss the integral trends provided in the high energy range by the PROFIL experiments. A new interpretation of the experimental results has been performed with the ERANOS-2.0 code system and the JEFF-3.0 neutron library. Experimental set-up and integral trends are presented in Reference [3]. Extensive comparison between the integral and microscopic trends for the fission products are given elsewhere [19]. The focus of this work is to discuss the ERANOS based JEFF-3.0 results obtained for the main actinides (uranium, plutonium, neptunium, americium and curium). Integral trends in terms of variation on the cross section are listed in Table 3.

Table 3 - Average integral trends provided by the interpretation of the PROFIL and PROFIL-2 experiments with JEFF-3.0. The $\Delta(\sigma)$ values stand for the integral trends in terms of variation on the cross section (in %). The positive (negative) signs indicate that the JEFF-3.0 cross section is overestimated (underestimated).

Isotopes		Reaction	$\Delta(\sigma)$
U	233	(n,γ)	- 9.4 ± 0.1
	234	(n,γ)	+ 2.6 ± 0.8
	238	(n,2n)	- 7.2 ± 3.3
Pu	238	(n,γ)	+ 2.3 ± 0.9
	239	(n,γ)	- 1.7 ± 1.3
	239	(n,2n)	- 32.4 ± 12.1
	240	(n,γ)	+ 4.0 ± 0.6
	241	(n,γ)	+ 11.7 ± 3.1
	242	(n,γ)	+ 14.6 ± 3.1
Th	232	(n,γ)	- 19.0 ± 10.5
Am	241	(n,γ)	+ 6.2 ± 2.3
Np	237	(n,γ)	- 5.6 ± 1.3
Cm	244	(n,γ)	- 2.5 ± 0.3

3.1 Microscopic Data

The quality of the capture data available in the EXFOR database depends on the isotope under investigation and on their applications in the conventional nuclear industry. From a closer inspection of the EXFOR data, three groups of isotopes can be distinguished:

- Several experimental capture data for the ^{239,240,242}Pu, ^{232}Th, ^{241}Am and ^{237}Np (n,γ) reactions are available in the EXFOR database. The comparison with the European libraries is shown in Figure 6.

- Isotopes for which a single one capture data set is available are shown in Figure 7. However, the data sets for the ^{241}Pu and ^{244}Cm (n,γ) reactions cannot be used to accurately describe the Unresolved Resonance Range above 10 keV. The ^{238}Pu(n,γ) data set reported by Silbert [20] has to be corrected with an appropriate normalisation factor of about 0.6 to become in good agreement with the PROFIL calculations. For ^{233}U, the data from Hopkins [21] are consistent

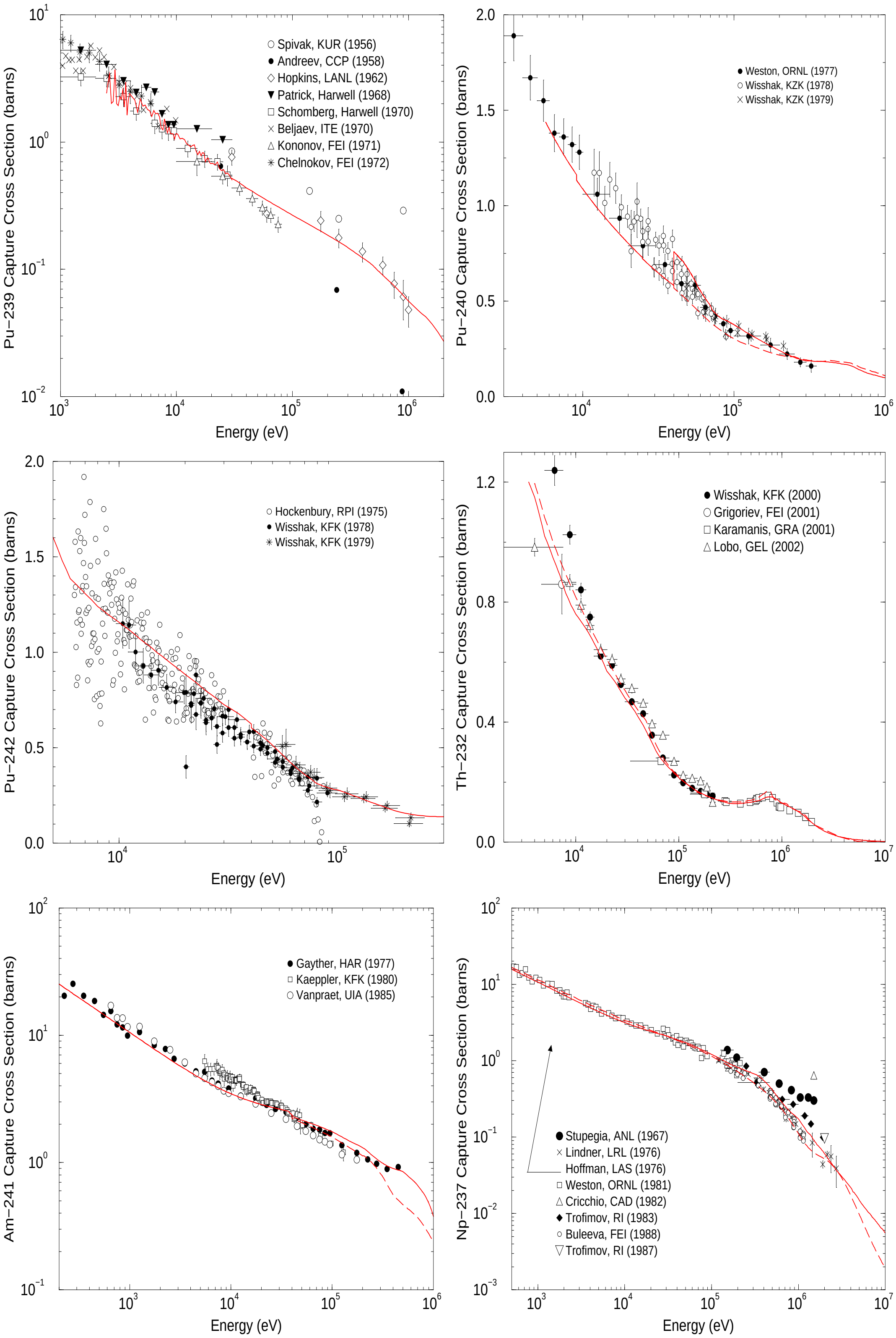

Figure 6 - High energy range of the ^{239,240,242}Pu, ^{232}Th, ^{241}Am and ^{237}Np capture cross sections. The JEFF-3.0 (solid line) and JEFF-3.1 (dashed line) neutron library are compared with the capture data reported in the EXFOR database (for ^{239,242}Pu, the JEFF-3.0 and JEFF-3.1 curves are similar).

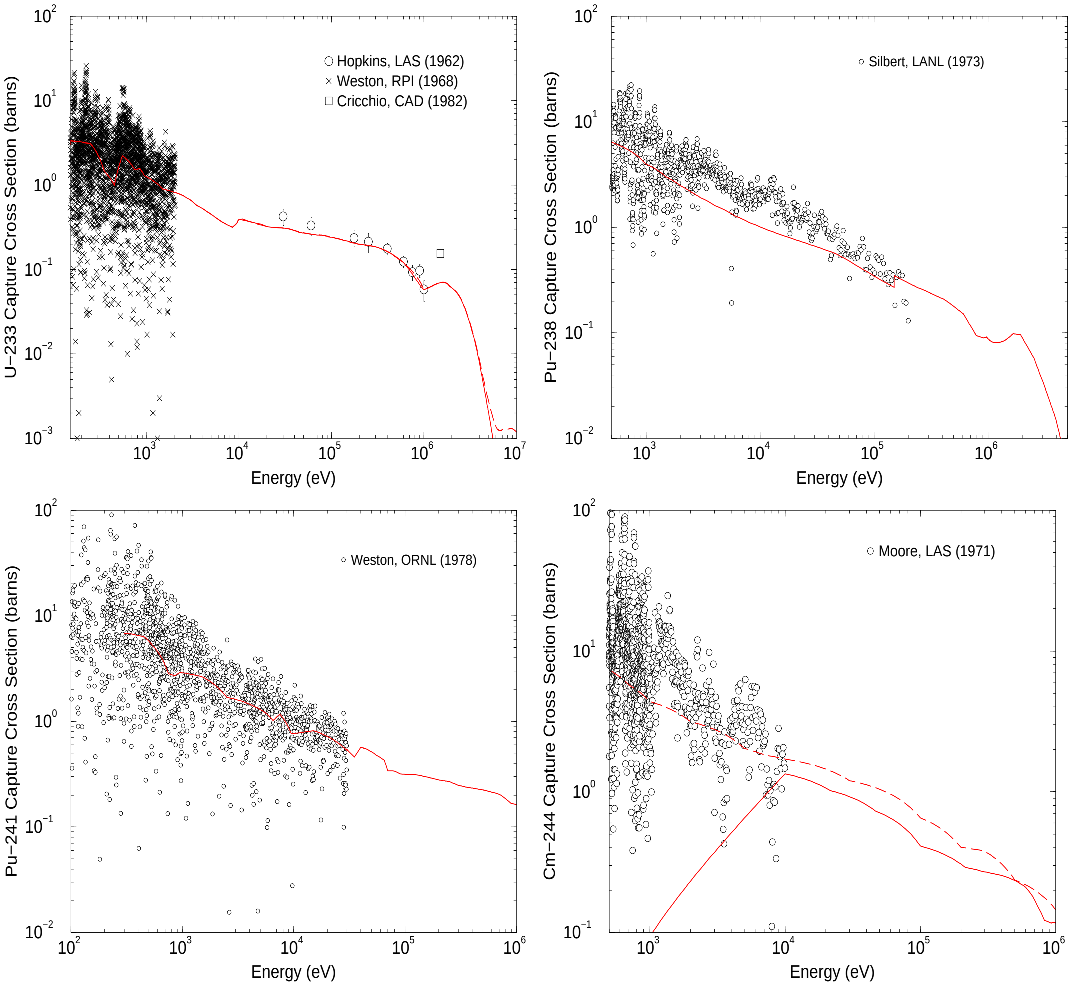

Figure 7 - High energy range of the ^{233}U, ^{238,241}Pu and ^{244}Cm capture cross sections. The JEFF-3.0 (solid line) and JEFF-3.1 (dashed line) neutron library are compared with the capture data reported in the EXFOR database (for ^{233}U, ^{238}Pu and ^{241}Pu the JEFF-3.0 and JEFF-3.1 curves are similar).

with the PROFIL predictions. However, the accuracy of these data is poor (from ±15% to ±27%).

- For ^{234}U, capture data have never been reported in the EXFOR library. Figure 8 compares the European evaluations JEFF-3.0 and JEFF-3.1.

3.2 Plutonium

For ^{239}Pu, the PROFIL experiments does not stress any significant mistakes on the statistical modeling of the (n,γ) reaction. The slight discrepancy on the C/E value reported in Table 3 remains within the experimental accuracy.

Extensive work have been done to improve the description of the ^{240}Pu cross sections in JEFF-3.0. The Unresolved Resonance Range has been revisited at the CEA/DEN of Cadarache [22], and the continuum region has been studied at the CEA/DAM of Bruyère le Chatel. However, around 40 keV, the continuity between the statistical treatment of the Unresolved Resonance

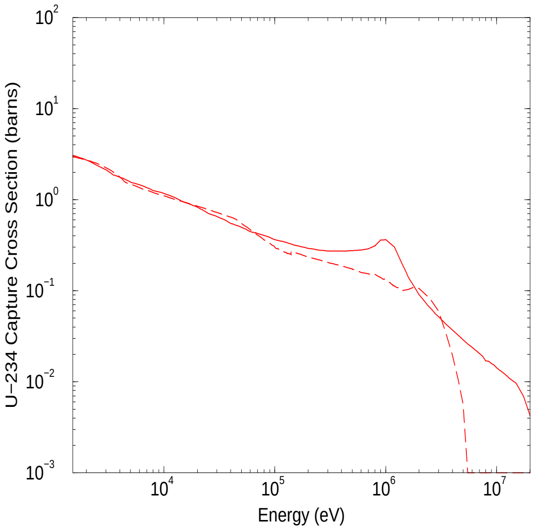

Figure 8 - ^{234}U capture cross sections recommended in the JEFF-3.0 (solid line) and JEFF-3.1 (dashed line) neutron library. capture data covering the Unresolved Resonance Range have never been reported in the EXFOR database.

Range (Hauser-Feshbach model) and the optical model calculations was not correctly described (Figure 6). The PROFIL experiments point out this mistake. As shown in Figure 1, the sensitivity of the C/E result to the high energy (n,γ) cross section reaches a maximum around $\sim$ 40 keV ($\simeq$ mean value of the ^{240}Pu sensitivity profile). This mistake has been corrected for JEFF-3.1.

The PROFIL experiments suggest that the ^{242}Pu effective capture integral from JEFF-3.0 (=JEFF-3.1) is overestimated by 15%. The three EXFOR data sets, shown in Figure 6, confirm the PROFIL trend while the modeling of the average capture cross section from 6 keV to 210 keV was previously performed on the same data sets and a complete check of all reaction channels above 40 keV has been made with improved optical model calculations [23]. This surprising result confirms the need for a better physics consistency between the resolved resonance range and the continuum region.

3.3 Americium

The PROFIL experiments suggest to decrease by 6% the ^{241}Am capture cross section recommended in JEFF-3.0. Around 100 keV (maximum of the high energy sensitivity profile, Figure 1), the JEFF-3.0 curve is systematically above the experimental data. For JEFF-3.1, it has been decided to keep the average resonance parameter of JEFF-3.0 (=JEF-2.2) and to replace the description of the high energy range (above 40 keV) by the evaluation performed by Maslov [24]. With this modification, the (n,γ) cross section becomes consistent with the data reported in EXFOR (Figure 6) and agrees with the PROFIL trend.

3.4 Neptunium

According to the PROFIL interpretation, the ^{237}Np(n,γ) cross section in JEFF-3.0 is underestimated by 5.6 ± 1.3%. Within the 10 keV to 100 keV neutron energy range (maximum of the sensitivity profile, Figure 1), the average resonance parameters have been determined on a

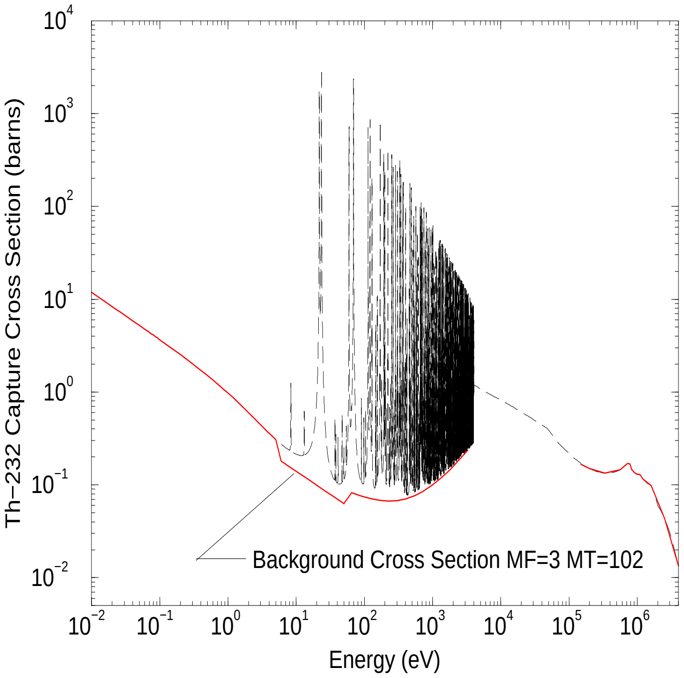

Figure 9 - ^{232}Th capture cross section (dashed line) together with the background cross section (solid line) introduced in the JEFF-3.1 evaluation.

single one data set reported by Weston [25]. The new evaluation of the Unresolved Resonance Range available in JEFF-3.1 is based on the same data set (Figure 6). Therefore, no significant modifications of the C/E values, reported in Table 3, are expected with JEFF-3.1.

3.5 Thorium

For ^{232}Th, the PROFIL interpretation based on JEFF-3.0 shows a strong underestimation of the (n,γ) cross section. A wide number of data sets are available in the EXFOR database. Only data reported in the library after 2000 are shown in Figure 6. One can observe that, within the [10 keV-100 keV] energy range (maximum of the sensitivity profile in the high energy range, Figure 1), the JEFF-3.0 curve is systematically below the experimental data. With JEFF-3.1, the C/E discrepancy could be slightly reduced. In the Unresolved Resonance Range [4 keV-150 keV], the evaluation performed by Maslov [26] agrees with the data measured by Wisshak [27].

In Figure 9, one can observe that the end of the resolved resonance range in JEFF-3.1 is not correctly described. This behaviour is due to the addition of a highly questionable background cross section. Significant improvements of the ^{232}Th evaluation could be reached by accounting for the latest data measured at the IRMM [28], n_TOF-CERN [29] and VdG facility of the Nuclear Centre of Bordeaux-Gradignan (CENBG) [30]. Above 10 keV, the capture data reported by Lobo et al [28] are roughly up to 15% higher than the (n,γ) cross section of JEFF-3.0.

3.6 Curium

The PROFIL-2 experiment has delivered an integral information on a single curium. The variation $\Delta(\sigma)$ reported in Table 3 was deduced from the isotopic ratio ^{245}Cm/^{244}Cm measured in an

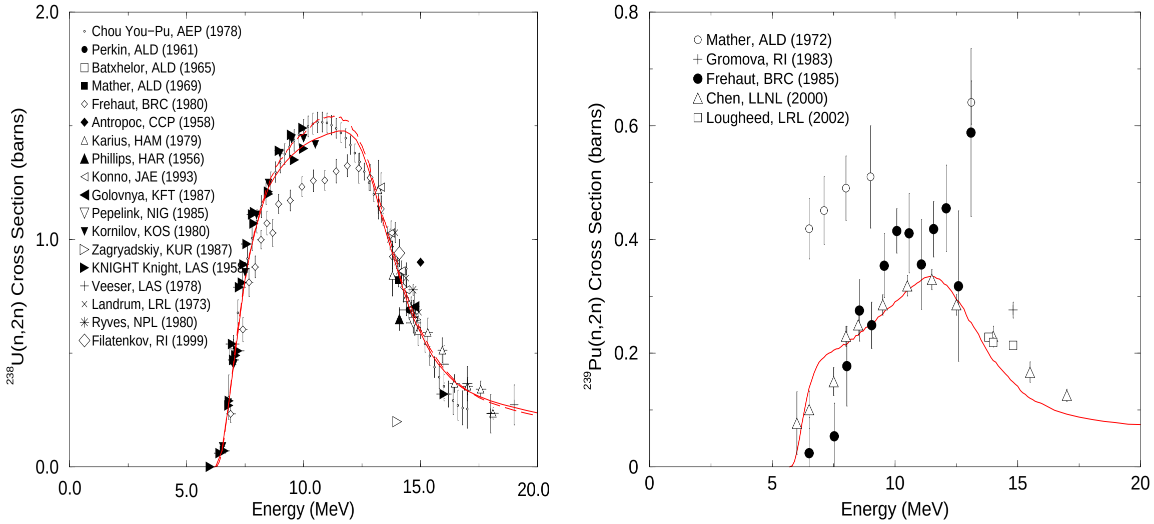

Figure 10 - ^{238}U and ^{239}Pu (n,2n) experimental cross sections available in EXFOR. The solid (dashed) line represents the JEFF-3.0 (JEFF-3.1) evaluation (for ^{239}Pu, the JEFF-3.0 and JEFF-3.1 curves are similar).

enriched ^{244}Cm sample. The C/E value obtained with JEFF-3.0 is close to unity. The discrepancy is lower than 3% (Table 2). However, this good result should be taken with care because of the wrong description of the Unresolved Resonance Range in JEFF-3.0 (Figure 8). The modeling of the cross section above the resonance range has been improved in JEFF-3.1 (=ENDF/B-VI.8). New ERANOS-2.0 calculations are needed to validate this significant modification.

4 (n,2n) Reactions

For the PROFIL experiments, measurements of the isotopic ratios ^{237}Np/^{238}U and ^{238}Pu/^{239}Pu provide an estimate of the trends on the (n,2n) cross sections of ^{238}U and ^{239}Pu. The results are presented in Table 3. The quoted accuracies does not take into account the slightly poor prediction of the neutron flux at high energy. Above a few MeV, the accuracy of the calculations is deteriorated by the uncertainty related to other nuclear data and threshold reactions. These trends are meaningful in a restricted energy range that corresponds to the maximum of the sensitivity profile. The latter reaches $\sim$ 85% of the total sensitivity within the 6 MeV to 10 MeV energy range.

4.1 Uranium

For uranium, the trend on the (n,2n) reaction has been deduced from a single measurement of the ^{237}Np/^{238}U ratio in a ^{238}U sample. The PROFIL result suggests to increase the (n,2n) cross section by $\sim$ 1.072. The low accuracy of the present prediction ($\pm$0.033) accounts for uncertainties provided by the chemistry analysis. Around 10 MeV, the integral trends agree with the upper experimental values and confirm the increase of about $\sim 3 - 4\%$ recommended in the JEFF-3.1 evaluation (Figure 10). The (n,2n) cross section included in the latest version of the European library is based on GNASH calculations [31] which have been validated with the data reported by Frehaut [32], re-normalized by a factor 1.10.

4.2 Plutonium

By contrast, the PROFIL experiments point out a strong underestimation of the ^{239}Pu(n,2n) cross section. The PROFIL results suggest to increase the JEFF-3.0 (=JEFF-3.1) evaluation by a factor 1.32 ± 0.12 (Figure 10). Owing to the poor accuracy of the PROFIL results and of the data available in EXFOR or in the literature [33], no relevant trend can be distinguished from the present microscopic and integral data.

Around 6 MeV, the behaviour of the (n,2n) cross section is not well defined. Within the 8 MeV to 10 MeV energy range, the uncertainty on the data measured by Frehaut [34] ranges from 10% to 40%. The data reported by Mather deviate significantly from the expected systematics. The data extracted from Reference [33] are model-dependent. They have been deduced from partial γ-ray cross sections measured with the GEANIE detector [35]. At high neutron energy, the latter become consistent with the data reported by Lougheed [36]. The European evaluation, based on BRC calculations [37], are in good agreement with the GEANIE data.

5 Measurement and Evaluation Issue

A summary of the needs in term of *measurements* or *evaluation* is given in Table 4. The list takes into account the experimental campaigns and evaluations which are planned or in progress. For example, evaluation of ^{232}Th is in progress in the frame of the CRP on Evaluated Nuclear Data for Th-U Fuel Cycle [38], and a wide experimental campaign on the ^{241}Am isotope is planed in the frame of a European collaboration between the Institute for Reference Materials and Measurements (IRMM), the Budapest Neutron Centre (BNC) and the CEA/DEN of Cadarache.

In a near future, the number of experimental data for actinides will increase significantly in EXFOR. Indeed, the last five years were the opportunity for the nuclear data community to carry out extensive measurements of these isotopes, through ADS and nuclear waste management related projects. The proceedings of the ND2004 conference, held in Santa Fe in 2004, give a wide overview of the experimental works that could be useful for future evaluations.

5.1 Uranium

For ^{233,234}U, capture data covering the Unresolved Resonance Range and the continuum region are needed. For the epi-thermal energy range (probably up to 10 keV), analysis of high-resolution Time-Of-Flight capture measurements carried out in the frame of the n_TOF collaboration is in progress.

Final evaluations could consider other experimental data. The ^{233,234}U fission cross sections have been measured at the n_TOF-CERN facility with the PPAC and FIC detectors [39, 40]. The low neutron energy range of the ^{234}U(n,f) reaction has been investigated at the High-Flux ILL Reactor and at the GELINA facility. High precision measurements of neutron induced fission of ^{233}U have been conducted by the Nuclear Centre of Bordeaux-Gradignan (CENBG) [41]. The last one we can mentioned is the high-resolution transmission measurements of ^{233}U performed at the ORELA facility.

About recent evaluation works, analysis of the ^{233}U Resolved Resonance Range have been performed at ORNL [42] and the high energy range has been revisited by Maslov. Systematic modeling of the high energy range of the uranium isotopes from $^{232-241}$U have been performed by the T16 Nuclear Physics Group of the Los Alamos National Laboratory (LANL). A similar work has been performed at the CEA/DAM of Bruyere-Le-Chatel.

Table 4 - Summary of the needs related to the (n,γ) and (n,2n) reactions of the main actinides of interest for the uranium and thorium fuel cycles. The needs are expressed in term of *evaluation* or *measurements*. The acronyms RRR, URR and Cont stand respectively for the Resolved Resonance Range, the Unresolved Resonance Range and the Continuum region. The experimental or evaluation works which are planned or in progress are indicated with a circle ($\circ$).

Isotopes		Reaction	Comments	RRR	URR	Cont.
U	233	(n,γ)	measurements	○	×	×
	234	(n,γ)	measurements	○	×	×
Pu	238	(n,γ)	measurements	×	×	×
	239	(n,2n)	measurements			×
	241	(n,γ)	measurements		×	×
			evaluation		×	×
	242	(n,γ)	measurements	×		
			evaluation	×	×	
Th	232		evaluation	○	○	○
Am	241	(n,γ)	measurements	○	○	
Np	237		evaluation	×	×	×
Cm	244	(n,γ)	measurements			○

5.2 Plutonium

Concerning the plutonium isotopes, the consistency of the evaluation vary according to their respective importance for the nuclear industry. The priority for the conventional applications is ^{239}Pu, ^{240}Pu, ^{241}Pu, ^{242}Pu and ^{238}Pu. The last one becomes important for the nuclear waste studies.

Owing to the poor accuracy of the EXFOR data, new measurements covering a wide energy range are needed to get reliable data for ^{238}Pu. Accurate measurements of the ^{239}Pu(n,2n) reaction is requested to solve the large discrepancy pointed out by the PROFIL results ($\sim$ 30%). Data are needed to described the ^{241}Pu(n,γ) reaction in the Unresolved Resonance Range and the continuum regions (the low energy range has been re-visited for JEFF-3.0). For ^{242}Pu, data in the Resolved Resonance Range is suitable in order to get reliable prior average resonance parameters for the statistical analysis of the Unresolved Resonance Range.

About evaluation activities, new modeling of the ^{241}Pu and ^{242}Pu capture, fission and elastic cross sections is needed to solve the huge discrepancies pointed out by the PROFIL experiments. For ^{242}Pu, the evaluation has to take into account the measurements carried out at the GELINA facility in the frame of the n_TOF collaboration. For ^{240}Pu, the European evaluation could be re-visited with the recent low temperature transmission data measured at the IRMM.

5.3 Neptunium

New evaluation of ^{237}Np is suitable to solve the ambiguity between the microscopic and integral trends (PROFIL experiments) introduced by the JEFF-3.1 library. The Evaluation work will account for the total cross section measured at the GELINA facility [44] as well as the fission and capture measurements carried out in the frame of the n_TOF collaboration [39, 45].

5.4 Americium and Curium

For the americium and the curium isotopes, wide experimental campaigns are planned or in progress. High-resolution Time-Of-Flight measurements have been performed at the n_TOF-CERN facility. The latter will provide relevant data for the ^{241}Am(n,f), ^{243}Am(n,f), ^{245}Cm(n,f) and ^{243}Am(n,γ) reactions. Measurements of the fast neutron-induced fission and capture of curium (^{242,243,244}Cm) and americium (^{241,243}Am) are conducted by the CENBG [43]. For the low and high energy range of ^{241}Am, a series of measurements are planned in the frame of a European collaboration between IRMM, the Budapest Neutron Centre (BNC) and the CEA/DEN of Cadarache.

The new interpretation with JEFF-3.1 of specific PIE will provide key information to refine the needs in terms of *measurement* and *evaluation* for ^{241,243}Am and $^{243-247}$Cm.

6 Conclusions

For the epi-thermal energy range, integral trends provided by the CRUAS and DAMPIERRE experiments confirm the overall improvements of the spent fuel inventory prediction based on JEFF-3.0. The increase of the ^{235}U capture resonance integral (by about 6%), of the ^{241}Pu capture area (0.26 eV) and of the ^{238}U(n,2n) reaction are the main corrections that improve the ^{236}U-^{237}Np-^{238}Pu and ^{242}Pu-^{243}Am build-up predictions. With JEFF-3.1, owing to the decrease by 0.7% of the ^{238}U capture resonance integral, of the increase of the ^{241}Am capture area at 0.3 eV and of the increase of the ^{241}Am isomeric ratio to ^{242m}Am, we expect to reduce the remaining discrepancy on the ^{239}Pu and on the ^{241,242m}Am build-up predictions.

For the high energy range, the PROFIL calculations based on JEFF-3.0 have demonstrated the needs for improving the Unresolved Resonance Range of ^{233}U, ^{240}Pu, ^{241}Pu, ^{242}Pu, ^{232}Th, ^{241}Am and ^{237}Np as well as the modeling of the ^{238}U and ^{239}Pu (n,2n) cross sections. The modifications in JEFF-3.1 of the ^{240}Pu and ^{241}Am capture cross sections and of the ^{238}U(n,2n) reaction agree with the PROFIL trends. Those for the ^{232}Th, ^{237}Np and ^{233}U capture cross sections will have a minor impact on the PROFIL calculations. The significant modifications of the ^{234}U and ^{244}Cm evaluations have to be validated with new ERANOS-2.0 calculations. Integral results for the ^{238}Pu, ^{241}Pu and ^{242}Pu capture cross sections and for the ^{239}Pu(n,2n) reaction will be unchanged with the JEFF-3.1 evaluations. The C/E values for the ^{233}U(n,γ), ^{241}Pu(n,γ), ^{242}Pu(n,γ), ^{239}Pu(n,2n) and ^{232}Th(n,γ) reactions will still deviate significantly from unity (from 9% to 30%).

The crude comparison between the needs in terms of integral trends and the EXFOR data shows the lack of accurate experimental data which could be included in the evaluation procedure. The EXFOR data can be separated into two groups of isotopes. For ^{242}Pu, ^{232}Th and ^{237}Np the experimental data available in EXFOR (or still under analysis) are (or will be) sufficient to perform new evaluations, while for the ^{239}Pu(n,2n) reaction and for the ^{241}Pu, ^{238}Pu, ^{241}Am, ^{233}U and ^{234}U capture cross sections, the experimental data does not exist in EXFOR or are not accurate enough to reach an accurate modeling of the epi-thermal and/or high energy ranges.

For the next release of the European library, a special care has to be given to the modeling of plutonium (^{241}Pu and ^{242}Pu), americium (^{241}Am), curium ($^{243-247}$Cm) and isotopes of interest for the thorium fuel cycle (^{232}Th, ^{233}U and ^{234}U).

References

[1] A. Santamarina et al., *Rapport de qualification R1 du produit APOLLO-2.5/CEA93.V6*, Technical Report CEA-SPRC/LEPh/02-003 (2002).

[2] D. Bernard et al. *JEF-2.2/JEFF-3.0 improvements on spent fuel inventory prediction in LWRs.*, JEF/DOC 1043 (2004).

[3] J. Tommasi et al., *Analysis of the PROFIL and PROFIL-2 sample irradiation experiments in PHENIX for JEFF-3.0 nuclear data validation*, Nucl. Sci. Eng., in press.

[4] J. Tommasi, *Synthèse des interprétations des expériences PROFIL et PROFIL-2 d'irradiation d'isotopes séparés dans PHENIX*, NT-DER/SPRC/LEPH-04/216 (2005).

[5] J. Tommasi, *Interprétation de l'expérience PROFIL-2 d'irradiation d'isotopes séparés dans PHENIX*, NT-DER/SPRC/LEPH-04/216 (2005).

[6] *The JEF-2.2 nuclear data library*, OCDE, NEA Databank, JEFF Report 17 (2000).

[7] H. Derrien et al., *Evaluation of* ^{238}U *resonance parameters from 0 to 20 keV*, ND2004, Santa Fe, New Mexico, Sept. 26 - Oct. 1 (2004).

[8] H. Derrien et al., *Reevaluation and validation of the* ^{241}Pu *resonance parameters in the energy range thermal to 20 eV*, Nucl. Sci. Eng. 150, 109 (2005).

[9] O. Bouland and H. Derrien, Nucl. Sci. Eng. 127, 2, 105 (1997).

[10] T.E. Young et al., *Neutron total and absorption cross section of* ^{238}Pu, Nucl. Sci. Eng. 30, 355 (1967).

[11] S.F. Mughabghab, M. Divadeenam and N.E. Holden, *Neutron Cross Sections: Neutron Resonance Parameters and Thermal Cross Sections* (Academic Press, New York, 1981).

[12] O. Bouland and D. Bernard, *Revised evaluation of* ^{241}Am, JEF/DOC-1086 (2005)

[13] G.G. Slaughter et al., *High-resolution total cross section measurements on* ^{237}Np *and* ^{241}Am, Bull. of the American Phys. Soc. 6, 70 (1961).

[14] L.N. Jurova et al. *Integral radiation capture cross sections in thermal and resonance energy region for* ^{230}Th, $^{231,232,233}Pa$, ^{236}U, ^{237}Np, All-Union Conf. on Neutron Physics, Kiev (1983).

[15] K. Kobayashi et al., *Measurements of thermal neutron cross section and resonance integral for the* $^{237}Np(n,\gamma)$ *reaction*, Report JAERI-M-94-019 (1993).

[16] A. Letourneau et al., *The minor actinide transmutation incineration potential studies in high intensity neutron fluxes*, ND2004, Sante Fe, New Mexico, Sept. 26 - Oct. 1 (2004).

[17] M.S. Moore and G.A. Keyworth, *Analysis of the fission and capture cross section of the curium isotopes*, Phys. Rev. C, 1656 (1971).

[18] J.C. Browne et al., *Fission cross section for curium-245 from 0.01 to 35 eV*, Nucl. Sci. Eng. 65, 166 (1978).

[19] E. Dupont, J. Tommasi and G. Noguere, *Contribution à l'analyse des besoins d'évaluation des produits de fission en spectre rapide*, NT-DER/SPRC/LEPH-05/203 (2005).

[20] M.G. Silbert and J.R. Berreth, *Neutron capture cross section of plutonium-238*, Nucl. Sci. Eng. 52, 187 (1973).

[21] J.C. Hopkins and B.C. Diven, *Neutron capture to fission ratios in U-233, U-235, Pu-239*, Nucl. Sci. Eng. 12, 169 (1962).

[22] O. Bouland, *Re-evaluation of the* ^{240}*Pu cross section in the unresolved resonance range*, PHYSOR, Seoul, Korea, Oct 7-10 (2002).

[23] A. Tudora, *Evaluation of* ^{242}*Pu data for the incident neutron energy range 5-20 MeV*, JEF/DOC-768 (1998); see also G. Vladuca et al., Report NEA/SEN/NSC/WPPR(96)5. (1996).

[24] V. Maslov et al., International Nuclear Data Committee, IAEA, INDC(BLR)-5 (1996)

[25] L.W. Weston and J.H Todd, *Neutron capture cross section of* ^{237}*Np*, Nucl. Sci. Eng. 79, 184 (1981).

[26] Maslov V. et al., Report JAERI-Research 01-0XX (2001).

[27] K. Wisshak et al., *Neutron capture cross section of* ^{232}*Th*, Nucl. Sci. Eng. 137, 183 (2001).

[28] G. Lobo et al., *Measurement of the* ^{232}*Th neutron capture cross section in the region 5 - 150 KeV*, Nucl. Sci. Tech., sup. 2, 429 (2002).

[29] G. Aerts et al., *Measurement of the neutron capture cross section of* ^{232}*Th at the n_TOF-CERN facility*, ND2004, Santa Fe, New Mexico, Sept. 26 - Oct. 1 (2004).

[30] D. Karamanis et al., *Neutron radiative capture cross section of* ^{232}*Th in the energy range from 0.06 to 2 MeV*, Nucl. Sci. Eng. 139, 282 (2001).

[31] M.J Lopez-Jimenez et al., *Overview of recent Bruyère-Le-Chatel actinide evaluations*, ND2004, Santa Fe, New Mexico, Sept. 26 - Oct. 1 (2004).

[32] J. Frehaut et al. *Status of (n,2n) cross section measurements at Bruyeres-le -Chatel*, Symposium on Neutron Cross Sections from 10-50 MeV, Upton, Long Island, May 12-14 (1980).

[33] H. Chen et al., *The* ^{239}*Pu(n,2n)*^{238}*Pu cross section inferred from the IDA calculations and GEANIE measurements*, Lawrence Livermore National Laboratory Report, UCRL-ID-141663 (2000).

[34] J. Frehaut et al., *(n,2n) cross sections of* ^{2}*H and* ^{239}*Pu*, Int. Conf. on Nuclear Data for Basic and Applied Science, Santa Fe (1985).

[35] J.A. Becker, *GEANIE Measurements of neutron-induced partial* γ*-ray cross sections for nuclear data*, ND2004, Santa Fe, New Mexico, Sept. 26 - Oct. 1 (2004).

[36] R.W. Lougheed et al., ^{239}*Pu and* ^{241}*Am (n,2n) cross-section measurements near* $E(n) = 14$ *MeV*, Rad. Acta, 90, 833 (2002).

[37] P. Romain et al., *The n+*^{239}*Pu system, new data evaluation and validation methods*, Nucl. Sci. Tech., Supp 2, 164 (2002).

[38] P. Schillebeeck and A. Trkov, *Evaluated Nuclear Data for Th-U Fuel Cycle*, International Nuclear Data Committee, IAEA, INDC(NDS)-468 (2004).

[39] L. Tassan-Got et al. *Fission of actinides induced by neutrons at n_TOF*, ND2004, Santa Fe, New Mexico, Sept. 26 - Oct. 1 (2004).

[40] V. Vlachoudis et al., *Measurements of fission cross section of actinides with the FIC detector at the n_TOF facility*, ND2004, Santa Fe, New Mexico, Sept. 26 - Oct. 1 (2004).

[41] C. Grosjean, *Mesure de la section efficace de fission de* ^{233}U *et des actinides mineurs induite par neutrons rapides*, PhD Thesis, CENBG, Bordeaux (2005).

[42] L.C. Leal et al., *Recent cross sections evaluations in the resolved resonance region at the Oak Ridge National laboratory*, ND2004, Santa Fe, New Mexico, Sept. 26 - Oct. 1 (2004).

[43] G. Barreau et al., *Fission cross sections of short-lived minor actinides by means of transfer reactions*, Int. Workshop on Nuclear Data Needs for Generation IV Nuclear Energy Systems, Apr. 5-7 , Antwerpen (2005).

[44] V.Gressier, *Nouvelle détermination expérimentale des paramètres de résonances neutroniques de* ^{237}Np *en dessous de 500 eV*, Phd Thesis, Université de Paris Sud (1999).

[45] V. Vlachoudis, *The n_TOF facility at CERN: performances and first physics results*, ND2004, Santa Fe, New Mexico, Sept. 26 - Oct. 1 (2004).