

EXPERIMENTAL VALIDATION OF MAIN FISSION PRODUCTS AND ACTINIDE NUCLEAR DATA. IMPROVEMENTS FOR JEFF-3.

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ABSTRACT

This paper describes how the analysis of selected integral experiments has led to recommendations for improving the main fission product and actinide cross-sections mainly in the thermal and the resolved resonance range. An extensive validation work of the JEF2.2 library, based on integral experiments performed in France, has led to propose specific modifications on nuclear data for the forthcoming JEFF3.0 file. These changes significantly improve many aspects of the neutronics calculations and reduce the discrepancies observed with JEF2.2. Various kind of thermal integral experiments are used to provide meaningful and complementary results on nuclear data. Concerning the major actinides, this study presents an extensive integral validation of the LDWL U235 evaluation, points out the trends obtained for U238 cross-section and proposes a revision of the resonance parameters for Pu241. For the main Fission products, using the reactivity worth measurements of separated FP samples and the Post-Irradiation Experiments on PWR spent fuel, this work suggests JEF2.2 capture cross-section modifications of Cs133, Nd143, Sm149 and Europium isotopes that are consistent both with integral and differential measurements. This work has strongly contributed to the elaboration and the preliminary validation of the JEFF3.0 nuclear data file

1 INTRODUCTION

The JEF2.2 [1] nuclear data library has been used for nearly 10 years mainly in the European countries, providing fruitful results in many fields such as reactor design, criticality and safety studies. In parallel, an important effort of cross-section evaluation has been done to take users feedback into account. This paper presents a synthesis of the work performed at CEA to validate the most recent evaluations with integral experiments. The results are mainly based on an extensive neutronic experimental program performed in France which includes Post-Irradiation Experiments from PWR, lattice criticality measurements, spectral indices, sample reactivity worth and temperature effect measurements. This paper emphasizes the importance of the inclusion of integral experiment information in the nuclear data evaluation process.

2 INTEGRAL EXPERIMENT CALCULATION METHODS

To interpret thermal integral experiments, deterministic calculations were performed with the French multigroup transport code APOLLO2 [2]. 2D lattice and assembly calculational schemes were carefully defined to avoid significant biases. Much attention was paid to the calculation of the U238 resonant capture rate :

- 172 energy group structure XMAS (CEA93 library based on JEF2.2) ;
- Accurate space dependent self-shielding formalism based on the background matrix theory (4 rings per fuel pins in order to account accurately for the "rim effect");
- The solid state effect in the Doppler broadening of the U238 resonances in UO_2 lattices has been studied in detail. To take account of this effect in the APOLLO2 code (only gas model for doppler broadening), the fuel temperature is adjusted to the following effective temperature T_{eff} which preserves the U238 reaction rate [3]:

$$T_{eff} = T_{fuel} + 8.6 + \frac{3100}{T_{fuel}}$$

where T_{fuel} is the real fuel temperature in Kelvin. This formula has been derived from resonant reaction rate calculations performed on the actual cross-section shape measured on UO_2 samples at GELINA facility [4] ;

- Collision probability methods (Pij) for the flux calculation. Calculations of Pij can be performed in the 2D exact geometry. Several module using interface currents methods have also been introduced allowing the use of the UP1 approximation (interface angular fluxes are linearly anisotropic).

To check these models, the integral experiments were also analysed through reference calculations using the TRIPOLI4 continuous-energy Monte Carlo code [5]. The probability table method is used to handle the U238 self-shielding in the unresolved resonance range.

An accurate modelling for depletion calculations was defined [6]. The influence of sensitive physical parameters was studied in detail such as, irradiation history, radial distribution of the fuel temperature within the pellet and its variation during irradiation (calculated with METEOR thermo-mechanical code), concentration of soluble boron, "stretch-out" operating mode. At every burn-up step, the APOLLO2 flux calculations were validated against reference continuous-energy TRIPOLI4 calculations : for example, the U238 capture rate is calculated in the deterministic APOLLO2 route with an accuracy of $-0.1\% \pm 0.1\%$ 1σ (TRIPOLI4 statistical uncertainty) and the Pu239 absorption rate is calculated with a negligible bias $+0.2\% \pm 0.1\%$ at any burn-up.

Burnup of PIE samples was deduced from fluence indicators such as $\frac{Nd_{148}}{U_{238}}$, $\frac{Nd_{145}}{U_{238}}$, $\frac{Nd_{150}}{U_{238}}$ ratios. Experimental uncertainties were evaluated from a detailed sensitivity study [7].

3 VALIDATION OF THE U_{235} LDWL EVALUATION

The new U_{235} evaluation proposed by Leal, Derrien, Wright and Larson (LDWL) in August 1997 [8] has been adopted in the JEFF3.0 file and in the Release 5 of ENDF/B-VI. One of the major differences between this evaluation and the previous JEF2.2 one is the increase of the capture cross-section in the resolved resonance range.

3.1 CAPTURE RESONANCE INTEGRAL : MOX PWR IRRADIATED FUEL

U236 build-up in a MOX spectrum gives an accurate validation of the U235 capture resonance integral. Indeed, the $\frac{U_{236}}{U_{238}}$ isotopic ratio is mainly sensitive to the U235 epithermal capture up to 40 GWj/t (at higher burn-up, U236 build-up is also sensitive to the U236 capture cross section). In MOX spectra, about 70% of U235 captures occur in the resonance region.

The investigated MOX assembly was irradiated in the SLB1 reactor, a 900 MWe PWR with a 30% MOX fuel loading. MOX assemblies include three zones with different Pu enrichments : a central zone (2.9% Pu), an intermediate zone (4.4% Pu) and a peripheral zone (5.7% Pu) [7].

rods	P14	I13	I02	N13	L14	A04	Q14	Q17
cycle	1	2	2	3	3	1	2	3
zone	Interm	Central	Interm	Interm	Central	Periph	Periph	Periph
BU (MWd/t)	12868	28368	28453	41493	45005	9556	24664	37683
U6/U8 (JEF2)	-10.13	-7.10	-8.24	-7.84	-6.45	-7.76	-8.00	-5.63
uncert. 1σ	$\pm 3\%$	$\pm 2\%$	$\pm 2\%$	$\pm 1.5\%$	$\pm 1.5\%$	$\pm 4\%$	$\pm 3\%$	$\pm 3\%$
U6/U8 (LDWL)	-5.68	-2.65	-3.79	-3.39	-2.00	-3.31	-3.55	-1.18

Table 1: C/E-1 in % using JEF2.2 and LDWL cross-sections for SLB1 experiment

The C/E values given by the peripheral rods are less reliable than the central rods owing to the transition spectrum at the MOX/UOX boundary and to the local burnup knowledge. Table 1 demonstrates that the new LDWL capture resonance integral (about 6% larger than in JEF2.2) reduces the discrepancy in U236 build-up : the mean bias over central + intermediate rods (2 and 3 cycles) decreases from $-7.4\% \pm 2\%$ to $-2.9\% \pm 2\%$. These results confirm the strong $\langle \Gamma_\gamma \rangle$ increase proposed by the LDWL evaluation. A stronger correction of the $\langle \Gamma_\gamma \rangle$ JEF2.2 value could be supported by this integral information, however the $\langle \Gamma_\gamma \rangle = 40 \text{ meV}$ deduced from the SAMMY analysis is probably the maximum value supported by the differential measurements.

3.2 UOX SYSTEM REACTIVITY

A set of French UOX cores was analysed : UH1.2, MISTRAL1 and CAMELEON all performed in the EOLE facility. The interpretation of these experiments used deterministic core calculations in two dimensions (Measured axial buckling was used to simulate 3D effects). We also investigated VALDUC experiments (LEU-COMP-THERM-007 in ICSBEP) which involves three UOX lattices experiments performed in the framework of Safety-Criticality programme. To enlarge the experimental validation of well thermalised UOX lattices, the buckling measurements in various EOLE experiments (CRISTO1, CRISTO2) are also reported in Table 2.

A 3D Monte-Carlo calculation (with TRIPOLI4 code) of the N4 PWR (CHOOZ-B1) start-up configuration is also analysed [12]. In this fresh fuel core calculation, the heterogeneous geometry (pin, clad, moderator, fuel assembly) is represented without any geometrical approximations.

We also included the analysis of MARACAS (LEU-COMP-THERM-049) critical configurations, performed in VALDUC. This programme involved arrays of contiguous cubic cans (20*20*20) loaded with low enriched uranium powders with an H/U ratio of 2, 2.5 and 3, reflected with polyethylene. The results shown in Table 2 represent the C/E value averaged over the various configurations.

To extend the experimental validation of the reactivity in LWR lattices, relevant LEU bench-

Experiment name	Lattice Pitch (cm)	V_{H2O}/V_{UO2}	Bore (ppm)	uncert. σ (pcm)	(C-E) (JEF2.2) (pcm)	(C-E) (LDWL) (pcm)
MARACAS1	powder	H/U=2	0	340	+290(100)	-510(200)
MARACAS2	powder	H/U=2.5	0	420	+430(100)	-382(200)
MARACAS3	powder	H/U=3	0	370	+290(100)	-316(200)
UH1.2	1.26	1.25	569	200	+347(70)	-113
N4	1.26	1.4	1214	300	+397(15)	-188
MISTRAL1	1.32	1.75	294	200	+147(80)	-264
CAMELEON	1.26	1.80	610	300	+801	+378
VALDUC1	1.26	1.82	0	300	-326(50)	-619
VALDUC2	1.60	3.81	0	300	-143(50)	-236
VALDUC3	2.10	7.58	0	300	-379(50)	-384
CRISTO2	1.58	3.56	832	300	-161	-351
CRISTO2L	1.71	4.40	672	300	-4	-226
CRISTO1	1.86	5.46	750	300	+217	+94
ZPR-HiC10	1.24	0.96	0	400	-180(70)	-572
ZPR-HiC13	1.16	0.60	0	600	-310(70)	-980
VVER11.0	1.10	0.89	0	370	-50(40)	-550
VVER12.7	1.27	1.66	0	300	+250(50)	-60
KRITZ2:1	1.48	1.17	218	380	-460(50)	-730
KRITZ2:13	1.63	1.70	452	250	-310(50)	-470

Table 2: C/E discrepancies on Keff for french UOX regular cores and LEU benchmarks

marks from the ICSBEP handbook were analysed. The TRIPOLI4-JEF2.2 results from ZPR-HiC (High Conversion tight lattices), KRITZ2:1 and VVER experiments are summarised in Table 2.

From the results presented in Table 2, the following conclusions can be drawn :

- In the Eole regular UOX cores UH1.2, MISTRAL1 and CAMELEON, the reactivity is well predicted using the U235 LDWL evaluation : $-100 \text{ pcm} \pm 150 \text{ pcm}$ (on the other hand the previous calculations based on JEF2.2 gave a Keff overestimation by $+330 \text{ pcm}$).
- Concerning the LWR-type international ICSBEP benchmark experiments (VALDUC, ZPR-HiC, VVER, KRITZ2), we notice a good reactivity prediction by TRIPOLI4/JEF2.2 : $-150 \text{ pcm} \pm 150 \text{ pcm}$. The use of LDWL evaluation for U235 cross-section causes a clear underestimation of the multiplication factor of these LWR small cores : about $-400 \text{ pcm} \pm 200 \text{ pcm}$.

This reactivity underestimation problem for LEU-LWR has been demonstrated by several authors and becomes a major issue for the evaluated file projects. As shown in the next section, these results do not allow to reject the U235 LDWL evaluation though.

3.3 HIGHLY ENRICHED URANIUM HOMOGENEOUS SYSTEMS

In order to have the maximum U235 resonance integral effect and to avoid possible error superposition problems with U238 cross-sections, two sets of highly enriched U235 experiment

were chosen. The first one includes the two TOPSY reflected uranium hybrid systems UH3-NI and UH2-UR and the HISS system (k_{inf} experiment) [15]. Reactivity calculations were performed with TRIPOLI4 and the impact of LDWL evaluation was calculated with APOLLO2 (infinite medium calculation). The second set of investigated experiments corresponds to highly enriched uranium solution experiments from ICSBEP Handbook (UO_2F_2 and $UO_2(NO_3)_2$ solutions). Only light water moderated experiments have been selected. Gadolinium poisoned solutions were not chosen in order to avoid additional uncertainty linked to Gd content knowledge. Uranium concentration ranged from 20 gU/l to about 700 gU/l, covering a wide range of spectra.

For the selected experiments, the LDWL reactivity effect is linear with q_{inf} (slowing down density calculated in infinite medium without leakage) and with uranium concentration. An average C-E equal to +380 pcm is obtained with JEF2.2, the use of LDWL evaluation gives a slight underestimation of -150 pcm on average (see Figure 1). We have also included in this analysis two experiments (HEU-SOL-THERM-004) with deuterium moderator. These systems have a very small q_{inf} and a large LDWL impact.

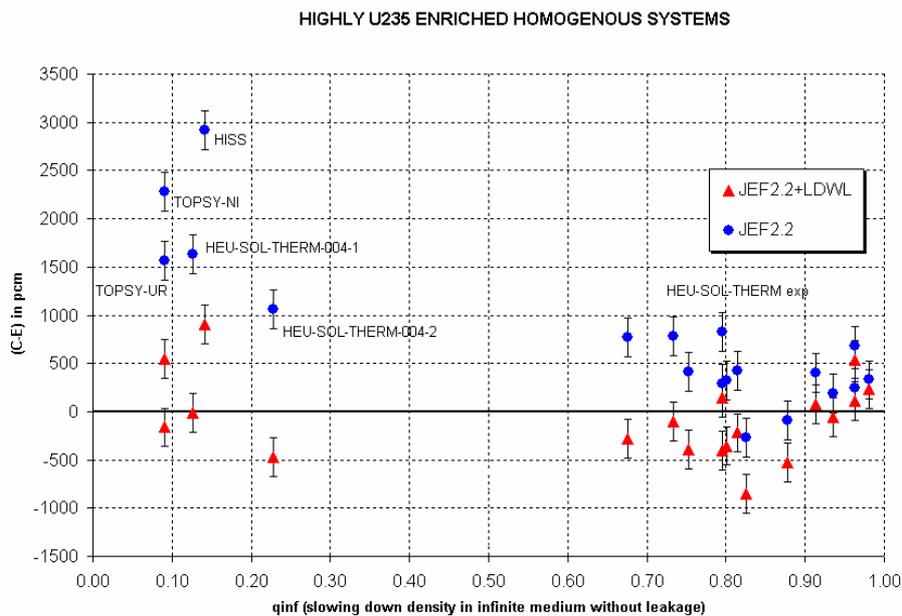


Figure 1: Reactivity (C-E) in pcm of highly enriched uranium experiments using JEF2.2 and LDWL evaluations.

We can conclude from this analysis that for highly enriched uranium systems the LDWL evaluation greatly improves the results, the overprediction of reactivity observed with JEF2.2 cross-sections being corrected. Considering also the clear improvement of the U236 build-up prediction, this evaluation was adopted in the JEFF3.0 file. It is emphasized that the LEU-LWR reactivity underestimation does not come from the LDWL evaluation but could come from the other nuclear data involved such as U238, O16 or H2O.

4 U238 CROSS-SECTION

The low energy part of the U238 evaluations (below 10 keV) is identical in the JEF2.2, ENDFB6 and JENDL3.2 libraries. The evaluation of the resonance region was revised by the NEANDC task force [20]. In this evaluation, much attention was paid to the capture cross-section evaluated by the task force which was not consistent with the integral measurements in the past. Compared to the previous evaluations, the JEF2.2 capture cross-section was significantly decreased. Two experiments were used in this work to validate the capture cross-section in the resolved resonance range : The direct measurement of the U238 capture rate in mock-up (Spectral index) and the build-up of Pu239 in PWRs.

4.1 SPECTRAL INDEX

The modified Conversion Ratio C^{U238}/F^{tot} or the spectral index $\sigma_c^{U238}/\sigma_f^{U235}$ were measured in the various experiments performed in the EOLE facility at CEA Cadarache. Two important experimental programs were chosen. The MISTRAL program [18] (MOX : Investigation of Systems which are Technically Relevant of Advanced Light water reactors), was devoted to advanced LWR loaded with 100% of MOX fuel. The ERASME program [19] was performed in 1985-1987 to study Light Water High Conversion Reactors loaded with mixed oxide fuel.

Four experimental configurations were investigated :

- **MISTRAL-1** : Regular enriched UO_2 lattice (3.7% U235 enrichment) involving about 750 fuel pins in a square pitch of 1.32 cm (moderation ratio equal to 1.76, H/HM=5.1). The criticality is achieved by adjusting the soluble boron concentration.
- **MISTRAL-2** : Regular 100% MOX lattice (7% Pu content) involving about 1600 fuel pins in a square pitch of 1.32 cm. Contrary to the previous experiment, no boron was added to the moderator. The criticality was obtained by adjustment of the radial critical size.
- **ERASME-S** : Tight hexagonal MOX lattice (11% Pu content, about 1500 MOX fuel pins) with a moderation ratio of 0.5
- **ERASME-R** : Realistic HCR hexagonal MOX lattice (11% Pu enrichment, about 1200 MOX fuel pins), with a moderation ratio equal to 0.9

Experiment name	Spectral index	C/E TRIPOLI4	exp. uncert.
MISTRAL1 UOX 3.7%	C^{U238}/F^{tot}	+2.2 %	± 2.0 %
MISTRAL2 MOX 7.0%	C^{U238}/F^{tot}	+2.3 %	± 1.5 %
ERASME-S	$\sigma_c^{U238}/\sigma_f^{U235}$	+1.6 %	± 2.3 %
ERASME-R	$\sigma_c^{U238}/\sigma_f^{U235}$	-0.2 %	± 2.1 %

Table 3: C/E in % on spectral index

From Table 3, we can conclude that the average overestimation of the U238 capture rate is :

$$(C - E)/E = +1.4\% \pm 1.0\%(1\sigma) \tag{1}$$

4.2 PU239 BUILD-UP IN PWR

Analysis of the Pu239 build-up from PWR spent fuel experiments can give a direct validation of U238 capture rate in the large resonances and in the thermal range (20% of the U238 capture rate). Three P.I.E experiments were investigated to that aim : 17*17 assemblies irradiated in BUGEY3, FESSENHEIM2 and GRAVELINES commercial reactors. In these experiments, removable pins are extracted from two assemblies at each inter-cycle shut-down, up to 5 irradiation cycles. The chemical assays are carried out on several rod cuts, at various heights in order to assess the axial variation effect of the water temperature.

NAME	cycle	U_{235} enrichment	maximum burn-up
Bugey3	3	3.1%	38 GWd/t
Fessenheim	5	3.1%	58 GWd/t
Gravelines	5	4.5%	60 GWd/t

Table 4: Characteristics of UOX spent fuel experiments

Experiment	20 GWj/t	40 GWj/t	50 GWj/t	60 GWj/t
Bugey	0.6	2.1		
Fessenheim			0.5	2.6
Gravelines	-0.7	0.4	1.0	1.4
exp. uncert. 1σ	$\pm 1.0\%$	$\pm 1.1\%$	$\pm 1.2\%$	$\pm 1.3\%$

Table 5: C/E-1 in % on $\frac{Pu_{239}}{U_{238}}$ using JEF2.2 cross-sections for UOX spent-fuel experiments [26].

The analysis results of the various $\frac{Pu_{239}}{U_{238}}$ chemical assays show that the U238 capture rate is slightly overestimated in JEF2.2 based calculations by $+1\% \pm 1\%(1\sigma)$.

This trend is confirmed by the PIE experiments performed in MOX fuels : chemical assays in the central zone (5-6% Pu content) of a MOX assembly irradiated in the SLB1 PWR reactor have shown a Pu239 build-up overestimation reaching $+3\% \pm 2\%$ after 3 irradiation cycles (BU = 45 GWd/t) [9].

From the irradiated fuel PIE experiments and the direct U238 capture measurements, we can conclude that the U238 capture rate is slightly over-estimated in UOX and MOX lattices : $1.2 \pm 0.7\%$ (1σ). Thus, the reduction of the U238 resonance capture should partly eliminate the current disagreement in Keff prediction for LWR UOX lattices.

4.3 U_{238} (N,2N) CROSS-SECTION

The U_{238} (n,2n) cross-section is important in LWR depletion calculations, because it influences greatly the formation of Np237 and Pu238.

Np237 formation : Table 6 shows that in French PWR assemblies, the Np237 build-up is underestimated. The formation of Np237 in UOX fuel is mainly governed by two mechanisms :

- the U_{235} capture : $U_{235} \xrightarrow{n,\gamma} U_{236} \xrightarrow{n,\gamma} U_{237} \xrightarrow{\beta^-} Np_{237}$

- the U_{238} (n,2n) reaction : $U_{238} \xrightarrow{n,2n} U_{237} \xrightarrow{\beta^-} Np_{237}$

The new U_{235} evaluation led to a reduction of this discrepancy, but there is still an underestimation which can be explained by the underestimation of the (n,2n) U_{238} cross-section.

Experiment	20 GWj/t	40 GWj/t	50 GWj/t	60 GWj/t
e=3.1%	-9.1	-1.9	-4.6	-0.6
e=4.5%	-2.3	-3.7	-5.3	-6.5
exp. uncert. 1σ	$\pm 3.4\%$	$\pm 3.4\%$	$\pm 3.4\%$	$\pm 3.2\%$

Table 6: C/E-1 in % on $\frac{Np_{237}}{U_{238}}$ using JEF2.2 cross-sections for UOX spent-fuel experiments.

Pu238 formation : In a UOX spectrum, Pu238 comes from three main processes :

- the U_{235} capture : $U_{235} \xrightarrow{n,\gamma} U_{236} \xrightarrow{n,\gamma} U_{237} \xrightarrow{\beta^-} Np_{237} \xrightarrow{n,\gamma} Np_{238} \xrightarrow{\beta^-} Pu_{238}$
- the U_{238} (n,2n) reaction : $U_{238} \xrightarrow{n,2n} U_{237} \xrightarrow{\beta^-} Np_{237} \xrightarrow{n,\gamma} Np_{238} \xrightarrow{\beta^-} Pu_{238}$
- the U_{238} (n, γ) through a complex chain of reactions which participates to the Pu238 formation (Cm242 decay)

At low burn-up, the U_{235} capture and U_{238} (n,2n) reaction contribute the most to the Pu238 build-up. The increase of the U_{235} capture in the resolved resonance range with the new LDWL evaluation improves the Pu238 prediction but it does not correct entirely the discrepancies (the effect of the new evaluation is to reduce the $\frac{Np_{237}}{U_{238}}$ by 2-3% at low burn-up in a UOX spectrum).

Experiment	20 GWj/t	40 GWj/t	50 GWj/t	60 GWj/t
e=3.1%	-8.7	-5.3	-4.0	-4.7
e=4.5%	-12.2	-10.6	-10.2	-10.2
exp. uncert. 1σ	$\pm 4.0\%$	$\pm 3.9\%$	$\pm 3.8\%$	$\pm 3.7\%$

Table 7: C/E-1 in % on $\frac{Pu_{238}}{U_{238}}$ using JEF2.2 cross-sections for UOX spent-fuel experiments.

Figure 2 compares the of U_{238} (n,2n) cross-sections from JEF2.2, ENDFB6.4 and BROND2.2. (JEF2.2 and JENDL3.2 (n,2n) cross-section are the same). The JEF2.2 evaluation is significantly lower than the other evaluations. Moreover, the comparison with the differential experimental data from the EXFOR database shows that the JEF2.2 evaluation is mainly based on a measurement performed by Frehaut et al. [21]. The other U_{238} (n,2n) differential measurements are significantly higher and more in agreement with ENDFB6 or BROND2.2 evaluation. The original data of Frehaut were recently renormalized [22] and are now in agreement with the other measurements. Therefore from the differential experiment results, it is demonstrated that the JEF2.2 (n,2n) cross-section is underestimated, in agreement with the integral information trend.

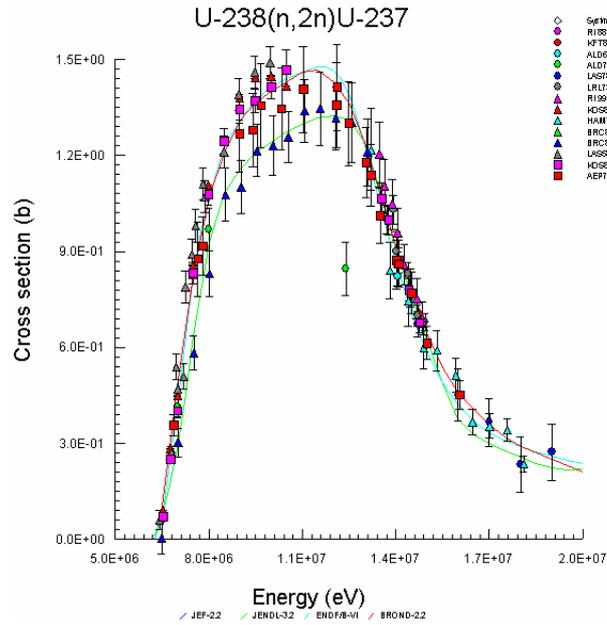


Figure 2: $(n,2n) U_{238}$ cross-section in the different evaluations and comparison with experimental data from EXFOR

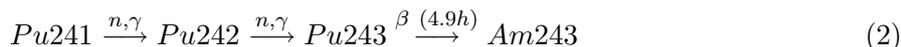
5 PU241 CROSS-SECTION

5.1 REVISION OF PU241 RESONANCE PARAMETERS UP TO 20 EV

In 1988, the Pu241 resonance parameters were evaluated in the neutron energy range from thermal to 300 eV by H. Derrien and G. De Saussure [23]. This evaluation was adopted in ENDF/B-VI and JEF2 libraries. After this work, a new fission cross-section measurement was performed by C. Wagemans et al. in 1991 [24] in order to check the shape of the fission cross-section in the thermal range. This measurement has demonstrated that the shape of the fission cross-section was compatible with the $1/v$ law contrary to the previous differential experiments. In order to take this information into account, the resonance parameters were revised in 1993 [25] in the energy range from 0.002 to 3 eV and adopted in a new release of ENDF/B-VI and in the JENDL3.2 file. Nevertheless, the experimental validation of the data through integral experiments (see [26] and [27]) has demonstrated that despite the reduction of C/E discrepancies given by the 1993 evaluation, the Pu241 capture cross-section is still significantly underestimated. To solve this problem, a new evaluation has been performed by H. Derrien [28] from thermal to 20 eV. This revision has led to a significant increase of the Pu241 capture cross-section in the 0.26 eV resonance compatible with the differential measurements.

5.2 EXPERIMENTAL VALIDATION USING POST-IRRADIATION EXPERIMENT

The French Post-irradiation Experiments (PIE) on PWR spent fuel give an accurate information on the average Pu241 cross-section in a PWR spectrum. The analysis of measured isotopic ratios Pu241/U238 and Pu242/U238 can be considered as a direct experimental validation of the Pu241 absorption and capture average cross-section respectively. Furthermore, the Am243 build-up is strongly related to the Pu241 capture cross-section (Pu242 formation) through the following process :



To assess the effect of the new Pu241 resonance parameters, the irradiated fuel experiment performed in the Gravelines PWR (UOX 17*17 assembly, 4.5% U235 enrichment) was analysed.

In a PWR spectrum, about 50% of the Pu241 capture and fission rate occurs in the 0.26 eV resonance (from 0.1 eV to 1 eV). About 35% of the reaction rate takes place in the 0.01 eV - 0.1 eV energy range (left wing of the 0.26 eV resonance). The rest of the reaction rate occurs above 1 eV and the contribution of the energy range below 0.01 eV is negligible.

Compared to the JEF2.2 evaluation, in the Gravelines spectrum at 10 GWd/t, the total Pu241 capture rate increases by 5.1 % and the fission rate decreases by about 2.7 %. The absorption reaction rate decreases slightly by 0.7 %.

Table 8 shows the C/E on Pu241, Pu242 and Am243 build up using JEF2.2 (1988 evaluation), JENDL3.2 (1993 evaluation) and the present revision (within the JENDL3.2 evaluation).

BU (GWd/t)	26.9	38.4	50.3	59.8
Pu241/U238 (JEF2.2)	-6.3 ± 2.3	-5.0 ± 1.8	-3.8 ± 1.6	-3.1 ± 1.6
Pu242/U238 (JEF2.2)	-10.5 ± 4.0	-9.7 ± 3.4	-8.8 ± 3.1	-8.6 ± 2.8
Am243/U238 (JEF2.2)	-18.0 ± 6.2	-11.6 ± 5.2		-8.8 ± 4.4
Pu241/U238 (JENDL3.2)	-5.4 ± 2.3	-3.8 ± 1.8	-2.5 ± 1.6	-1.6 ± 1.6
Pu242/U238 (JENDL3.2)	-9.0 ± 4.0	-7.9 ± 3.4	-6.8 ± 3.1	-6.4 ± 2.8
Am243/U238 (JENDL3.2)	-16.6 ± 6.2	-9.9 ± 5.2		-6.8 ± 4.4
Pu241/U238 (This work)	-5.9 ± 2.3	-4.4 ± 1.8	-3.1 ± 1.6	-2.4 ± 1.6
Pu242/U238 (This work)	-5.2 ± 4.0	-4.3 ± 3.4	-3.2 ± 3.1	-2.9 ± 2.8
Am243/U238 (This work)	-12.8 ± 6.2	-6.5 ± 5.2		-3.7 ± 4.4

Table 8: C/E-1 in % using JEF2.2, JENDL3.2 and the present Pu241 resonance parameters for the Gravelines experiment.

Table 8 shows that the decrease of the Pu241 absorption cross-section leads to an improvement of the Pu241 build-up prediction. The increase of the Pu241 capture cross-section in the present work has significantly reduced the discrepancy on the Pu242 formation. The correction of the Am243 prediction shows that the previous discrepancy was mostly linked to the underestimation of the Pu242 build-up. Furthermore, the large discrepancy observed in the Cm244 (-17% at 60 GWj/t) and Cm245 (-18% at 60 GWj/t) build-up prediction [26] is partly corrected with the new resonance parameters.

6 FISSION PRODUCTS

The following sections summarize the main trends obtained for Cs133, Nd143, Sm149 and Eu155 using the separated sample reactivity worth measurements in the Minerve reactor and the irradiated fuel analyses.

6.1 TRENDS GIVEN BY INTEGRAL EXPERIMENTS

In the Minerve reactor, three experiments were performed in the framework of Burn-up Credit program : R1-UO2 devoted to BUC investigation in storage pool and PWR-assembly transportation, R2-UO2 with a softer spectrum corresponding to the optimum moderation-ratio in a fuel dissolver and R1-MOX that was carried out to simulate a MOX spectrum. The description of these experiments is described in Reference [29]. Table 9 presents the C/E discrepancies on separated FP sample reactivity worth in the three Minerve configurations [30].

Fission Product	R1-MOX	R1-UO2	R2-UO2
Cs133	+6.5 ± 2.5%	+5.2 ± 2.5%	+4.8 ± 2.8%
Nd143	-0.7 ± 2.5%	-4.0 ± 2.5%	-6.0 ± 3.0%
Sm149	-6.2 ± 2.5%	-5.3 ± 2.4%	-4.2 ± 2.9%

Table 9: C/E-1 discrepancies in % on separated FP sample reactivity worth in the Minerve experiments with JEF2.2

The P.I.E give meaningful information about the Eu155 isotope (which was not oscillated in Minerve). Large discrepancies are observed on Europium and Gadolinium isotopes build-up with JEF2.2 library as shown in Table 10.

Burn-up GWd/t fuel	30	30	40	40	50	60
	UOX	MOX	UOX	MOX	UOX	UOX
154Eu/Cs137	+41%		+62%	+60%	+84%	+100%
154Eu/U238	+50%	+13%	+54%	+45%	+75%	+94%
154Gd/U238	+50%	+14%		+45%	+73%	
155Eu/U238	+9%	+62%	+14%	+20%	+16%	+18%
155Gd/U238	-1%	+54%	+4%	+10%	+8%	+12%
156Gd/U238					-22%	-15%

Table 10: C/E-1 in % discrepancies from French P.I.E for Eu and Gd isotopes with JEF2.2

The Minerve experiments and the P.I.E results for the studied FP suggest the following trends :

- Cs133 capture cross-section in the first resonance is overestimated (about 6%)
- Sm149 capture cross-section in the thermal and resonance range is under-estimated in JEF2.2 (about 5%)
- Nd143 thermal capture is under-estimated by about 3-5% in JEF2.2
- Eu154 and Eu155 capture resonance integral are not realistic (strong underestimation of the capture cross-section in the resolved range)

6.2 IMPROVEMENTS OF FP CAPTURE CROSS-SECTION

The information given by integral experiments allows to select the best evaluations amongst the most recent one. When the evaluations do not follow the trends given by integral experiments, specific modifications of the existing evaluation consistent with the differential measurements are proposed in the thermal and resolved range. A rigorous fit of available differential experiments using the integral information has not been performed in this study. A quick overview of the measurements (generally the thermal capture cross-section at 0.0253 eV and the resonance parameters of the first large resonance) has allowed us to find simple solutions to correct the cross-section.

Cs133 resonance integral : It can be seen in Table 11 that both JENDL3.2 and ENDFB6.7 follow the trend of the integral experiments, i.e a decrease of the resonance integral. Nevertheless, the decrease proposed by JENDL3.2 seems to be too strong.

	RI (b)	difference
JEF2.2	438	0 %
JENDL3.2	396	-9.6 %
ENDFB6.7	421	-3.9 %

Table 11: capture Resonance Integral (RI) given in the investigated evaluations

In addition, contrary to JEF2.2, the ENDFB6.7 and JENDL3.2 evaluations are based on the experiment carried out in 1990 by Nakajima et al. [31]. This experiment was performed with a very high energy resolution and is probably the best capture measurement available up to now in this energy range.

Nd143 thermal capture cross-section : The calculation of the Nd143 absorption rate in a thermal spectrum depends mainly on its thermal capture cross-section at 0.0253 eV. In order to correct the discrepancies observed in the Minerve experiment, we have investigated the thermal capture measurements available in the EXFOR database. The compilation of the measurements combined with the Minerve trends leads to a recommended value $\sigma_0^{Nd143} = 338 b$. This can be done easily in the evaluations by adjusting the Γ_n value of the Nd143 bound level.

Sm149 first large resonance : The Sm149 capture process in the Minerve spectrum mainly occurs in the first large resonance at about 0.1 eV. In JEF2.2, the increase of Γ_n of the first resonance by 3% is compatible with differential measurements and leads to a better agreement with the Minerve results. Moreover, it was checked that this modification induces a thermal capture value equal to $\sigma_0^{Sm149} = 41085 b$ consistent with the experimental data compiled in EXFOR.

Europium isotopes in JEF2.2 In JEF2.2, the cross-section in the resolved resonance range for Eu155 and Eu154 are not based on differential measurements (the evaluation was performed before the first transmission measurement). The resonance parameters were assigned randomly to respect average properties of these nuclei (average resonance spacing, average radiative and neutron width and their theoretical distribution). As shown in Table 10, these theoretical methods are not accurate enough to assess cross-section shape and can produce important discrepancies in depletion calculations.

The evaluations ENDFB6 and JENDL3.2 are very similar in the resolved resonance range (resonance parameters, thermal capture value and resonance integral). They are mainly based on the Anufriev et al. [32] transmission measurement. We have re-analysed the UOX and MOX P.I.E. experiments with the ENDFB6.7 cross-section for Eu155 and Eu154.

Burn-up GWd/t fuel	30 MOX	40 UOX	40 MOX	50 UOX	60 UOX
154Eu/Cs137		-3%		+1%	+4%
154Eu/U238	-8%	-7%	-8%	-4%	+1%
154Gd/U238	-7%		-7%	-4%	
155Eu/U238	+17%	+14%	+13%	+14%	+16%
155Gd/U238	+11%	+4%	+2%	+6%	+9%
156Gd/U238				-4%	+3%

Table 12: C/E-1 in % discrepancies from French P.I.E for Eu and Gd isotopes with JEF2.2 except Eu154 and Eu155 that come from ENDFB6.7 library

From the results presented in Table 12, it is shown that the Eu154, 154Gd, 155Gd and 156Gd build-up calculation is well improved. However, Eu155 build-up is still slightly overestimated, but the C/E agreement is no longer the cancellation of the two large biases on its formation (Eu154 capture) and its absorption. These results confirm that the resolved range evaluation for Eu154 and Eu155 based on the available differential measurements, mainly from Anufriev et al. are consistent with integral experiments.

7 CONCLUSION

The role of integral experiment is essential to the building process of a nuclear data library such as JEFF3.0. This paper has shown that the U_{235} evaluation proposed by Leal, Derrien, Wright and Larson improves the prediction of relevant experiments :

- The over-estimation of U236 build-up observed with JEF2.2 in the P.I.E of French PWR assemblies (UOX and MOX fuel cuts) is strongly reduced with the new U235 evaluation.
- The strong reactivity over-estimation in highly enriched uranium homogeneous systems is well corrected with LDWL.

However, the Keff prediction for Light Water Reactors and thermalized UOX lattices seems to be less satisfactory using the U235 LDWL evaluation. A trend to under-prediction is demonstrated, requiring to improve other nuclear data involved in the UOX lattices calculation. The analysis of integral experiments sensitive to the U238 cross-section in the epithermal range demonstrates a slight overestimation of the U238 capture rate that could correct partially the reactivity prediction. This is one of the most important issue that has to be adressed by the nuclear data community in the near future.

Furthermore, the irradiated fuel chemical assays are among the most accurate integral experiments that allows to give individual information on nuclide cross-sections. Their results have led to a revision of Pu241 resonance parameters and to a modification of important fission products in JEFF3.0.

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