

## QUALIFICATION OF THE $^{235}\text{U}$ LEAL DERRIEN LARSON EVALUATION USING FRENCH INTEGRAL EXPERIMENTS

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Contribution to the JEFF Working Group Meeting

15-16 December 1997, NEA Paris

### I. INTRODUCTION

It is now well known that the resonance region evaluation of  $^{235}\text{U}$  (of Leal and de Saussure) adopted in ENDF/B-VI and JEF2.2 nuclear data files is unsatisfactory. Several studies /1/ have demonstrated this evaluation could have an underestimation of about 10% for the capture cross section in the resonance energy range. Analysis concluded that the potential explanation is an underestimation of the mean capture width.

In consequence, it was decided to produce a new  $^{235}\text{U}$  evaluation. A first evaluation was proposed by Leal and Derrien (1995). Studies /2/ have tested it and concluded that the LD95 evaluation correct the capture cross section in the epithermal energy range but it is not fully satisfactory.

Leal Derrien and Larson /3/ propose a new  $^{235}\text{U}$  evaluation (LDL) which could be introduced in the future European library JEFF3.

This paper present the validation results based on the French critical experiments and PWR spent fuel analysis. The second section compares the different evaluations. The third section presents the reference scheme used in the APOLLO2 code. The fourth and the fifth describe the results of the validation.

### II. $^{235}\text{U}$ EVALUATIONS AND PROCESSING

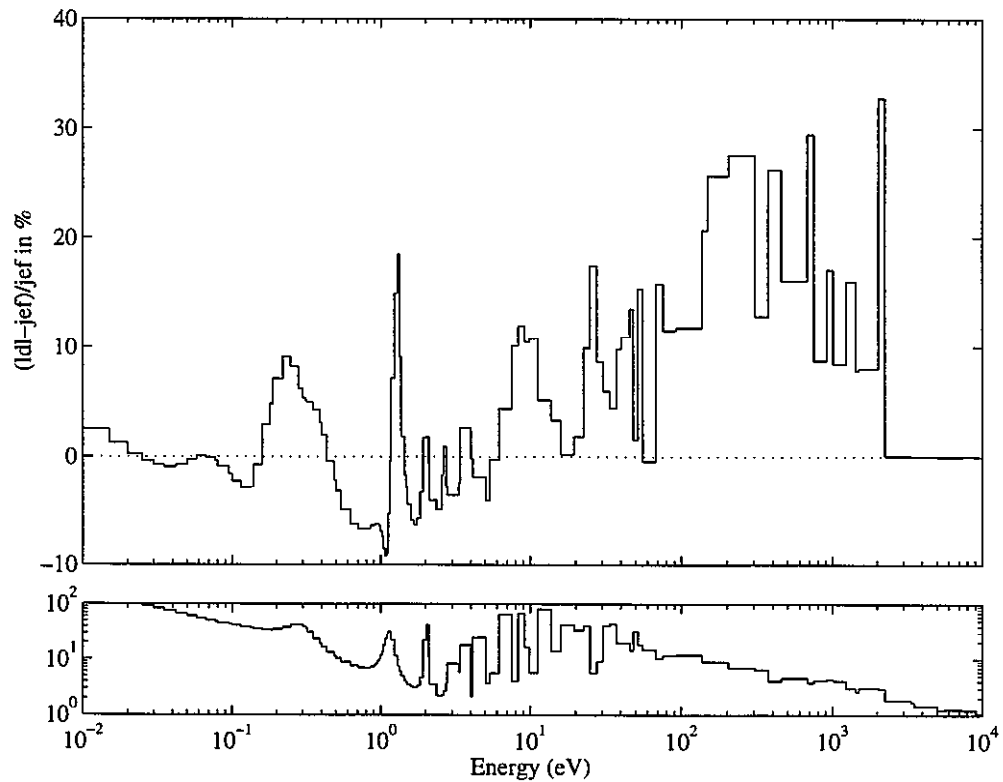
The main difference between the LDL evaluation and the previous (used in ENDF/B-6 and JEF2.2 nuclear data file) concerns the epithermal capture cross section. The LDL evaluation presents an increase about 22% in the energy range from 100 to 1000eV and 18% in [22.6eV, 454eV].

Table 1 : 2200 m/s values of the cross sections

cross-section	JEF2.2	ENDF/B-6	LDL
$\sigma_{0f}$	584.18	584.25	588.88
$\sigma_{0\gamma}$	98.76	98.96	98.66
$\sigma_{0n}$	15.48	15.46	15.67

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Figure 1 :  $^{235}\text{U}$  Evaluation Differences



The comparison of the fission and capture resonance integrals shows that the LDL evaluation is in excellent agreement with the value obtained from the integral measurements  $(0.513 \pm 0.015)$  /4/.

Table 2 : comparison of fission and capture resonance integrals (b)

	ENDF/B-V	JEF2.2	LDL
$I_\gamma$	138.97	132.88	140.49
$I_f$	281.92	278.61	276.04
$\alpha = I_\gamma / I_f$	0.493	0.477	0.509

We performed calculations using the recommended reference scheme and three different U235 nuclear data /5/.

- The first set is the JEF2.2 evaluation processed by NJOY .89 (previous version), it corresponds to the current CEA93 library of the APOLLO2 code.
- The second set is the same evaluation processed by the recent NJOY.94,
- The third set is based on the U235 LEAL DERRIEN LARSON's evaluation processed with NJOY.94.

In the second and the third sets, the fission spectrum is modified.

### III. APOLLO2 REFERENCE SCHEME

Calculations were performed with the French multigroup transport code APOLLO2 /6/. This code allows the use of several collision probability methods ( $P_{ij}$ ) to solve the integral equation. The  $P_{ij}$  calculations in the exact geometry can be performed and several modules using the interface current method have been introduced in APOLLO2 : RothxN formalism, UPk methods where  $P_{ss}$  transmission probabilities are computed in the exact geometry. In the UP0 method, the interface currents are assumed to be isotropic and in the UP1 to be linearly anisotropic.

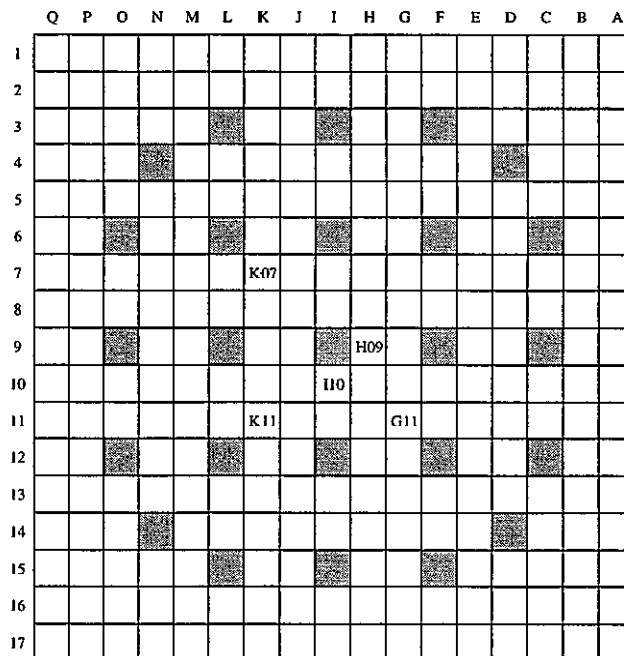
It is well-known that we need a calculational scheme without any bias in order to analyze nuclear data. We have defined a reference route using the APOLLO2 code. It is based on :

- 172 energy group library, allowing to simulate correctly in the whole spectrum
- a very accurate space dependent self-shielding formalism on the actual heterogeneous geometry. 6 rings in the rod are described in order to represent the Rim effect. The background matrix formalism allows the treatment of the Dancoff effect between different materials and/or resonant nuclide concentrations.  $P_{ij}$  are computed with the UP1 multicell model. We demonstrated that this model is enough in the self-shielding calculation, giving the same results that exact 2D  $P_{ij}$ .
- exact 2D  $P_{ij}$  to the flux calculation.

This calculation scheme was checked against TRIPOLI4 reference continuous-energy Monte Carlo calculations /7/.

### IV. ANALYSIS OF UO<sub>2</sub> PWR SPENT FUEL

The investigated UO<sub>2</sub> 17x17 assembly was irradiated in a standard 900 MWe PWR: BUGEY 3 /8/. The initial enrichment is 3.25% in U235. Several rods of this assembly have been extracted at increasing irradiation rates, and analyzed. These rods H09, G11, K07, K11, I10 are positioned in a central zone of the assembly.



For this study, we used only the measured fuel inventory at mid-plane of the core ( height 1900 mm). The rod cuts are 2cm long in a flat axial BU zone. The burn up of each sample is determined from the Nd148/ U238 ratio.

The following tables summarize the discrepancies between Calculation and Experiment on U235/U238 and U236/U238 isotopic ratio in the investigated.

The chemical analyses in fuel from rods H09 and G11 correspond to identical burn ups, so we have made an average on these two rods, in order to decrease the experimental uncertainty. The C/E comparison was also average for rods I10 and K11 extracted after 3 irradiation cycles.

**Table 3 : C/E -1 in % using U235-JEF2.2 evaluation, processing NJOY.89**

	number of cycle	Burn up (Mwd/t)	U235/ U238	U236/U238
<b>rods H09+G11</b>	1.5	20 200	1.77	-1.01
<b>rod K07</b>	2	24 000	3.35	-1.59
<b>rods I10+K11</b>	3	39 000	1.43	-1.31

**Table 4 : C/E -1 in % using U235-JEF2.2 evaluation, processing NJOY.94**

	number of cycle	Burn up (Mwd/t)	U235/ U238	U236/U238
<b>rods H09+G11</b>	1.5	20 200	1.83	-1.12
<b>rod K07</b>	2	24 000	3.41	-1.66
<b>rods I10+K11</b>	3	39 000	1.54	-1.37

**Table 5 : C/E -1 in % using U235 Leal Derrien Larson , processing NJOY.94**

	number of cycle	Burn up (Mwd/t)	U235/ U238	U236/U238
<b>rods H09+G11</b>	1.5	20 200	1.41	2.11
<b>rod K07</b>	2	24 000	2.90	1.55
<b>rods I10+K11</b>	3	39 000	0.74	1.81

Comparing the results in Table 3 and Table 4, we can observe very small differences on the U235 depletion and on the U236 build-up due to the recent version of NJOY. This arises mainly from the fission spectrum of U235. The effect of the decreased U238(n,2n) reaction rate with the NJOY.94 processing is significant only for Np237 and Pu238 build-up : -1% at 20 GWd/t.

The results shown in Tables 4 and 5 point out that the LDL evaluation improves the calculation of U235 depletion with irradiation : the Calculation -Experiment discrepancy is reduced from +1.54% with JEF2 library to +0.74% using LDL evaluation (rod cuts extracted after 3 cycles).

Concerning U236 build-up, Tables 4 and 5 show that the underestimation from JEF2 calculation is canceled using LDL evaluation, however the increase of U235 capture rate looks overestimated. In the spent fuel analysis from Gravelines 3 reactor /9/ (initial enrichment 4.5%), the JEF2 calculation show a higher underestimation of U236 build-up : the mean bias over the 17 rod cuts spanning the [27;62

Gwd/t]burnup range is:  $-4.3\% \pm 0.8\%$  ( $1\sigma$ ). Using the U235 LDL evaluation enables to reduce the Calculation-Experiment disagreement down to  $-1\%$ .

Tables 6 and 7 present the U235 averaged capture and fission cross sections and the  $\nu$  neutron number emitted by fission. These macrogroup data permit to quantify the cross section level LDL/JEF2 (version NJOY.94, Burn up 30 Gwd/t) in epithermal and thermal range.

**Table 6 : Resonance range ( 0.625 eV to 5 Kev):**

evaluation	capture cross section (barn)	fission cross section (barn)	$\nu$
JEF2.2	12.47	24.78	2.434
LDL	13.39	24.54	2.434
(LDL-JEF2.2)/JEF2.2	+7.37 %	-0.97 %	0%

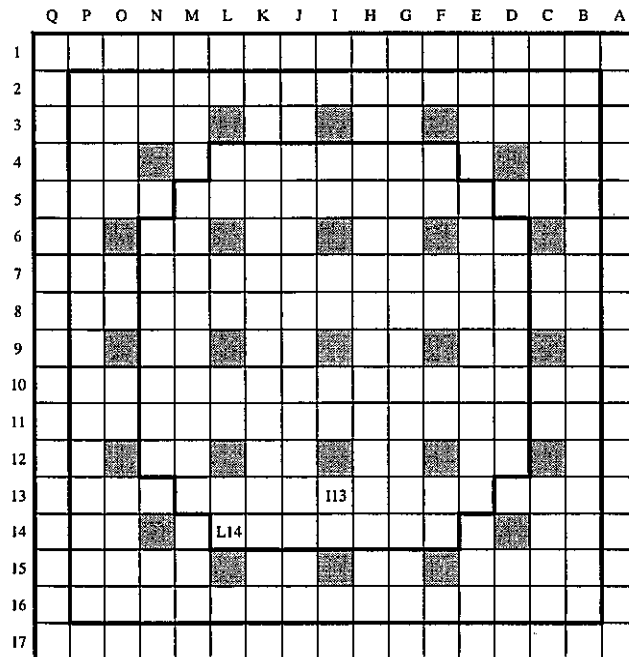
**Table 7 : thermal range ( 0.11 meV to 0.625 ev):**

evaluation	capture cross section (barn)	fission cross section (barn)	$\nu$
JEF2.2	43.48	252.32	2.438
LDL	43.81	251.00	2.437
(LDL-JEF2.2)/JEF2.2	+0.76 %	-0.52 %	-0.041%

Table 6 confirms that the major modification in the recent LDL evaluation is linked to the capture Resonance Integral, inducing a +7% increase in U235 epithermal capture rate. Since 40% of U235 capture rate happens in this resonance range, the LDL evaluation generates a 3% increase of U236 inventory in UO2 fueled lattices.

## V. ANALYSIS OF MOX PWR SPENT FUEL

The investigated 17x17 MOX assembly was unloaded from the SLB1 reactor. This PWR is devoted to Pu recycling, thus it corresponds to a mixed loaded core with 30% MOX assembly. Every MOX assembly is constituted by three zones of Plutonium enrichment, this enrichment is crescent from the peripheral zone towards the center zone. For our purpose, the most interesting analyzed rods are numbered I13 and L14: they are located in the central zone, therefore they are not under neutronic influence of the surrounding UO<sub>2</sub> fuel assemblies. The rods I13 was irradiated during two irradiation cycles and L14 during three cycles.



Three assembly calculations were carried out using the MOX reference calculation scheme and the three different U235 cross-section sets.

Tables 8, 9, 10 show the Calculation-Experiment comparison on U235 depletion and U236 build-up in the two « asymptotic » MOX pins.

**Table 8 : C/E -1 in % using U235-JEF2.2 evaluation, processing NJOY.89**

	number of cycle	Burn up (MWd/t)	U235/ U238	U236/U238
rod I13	2	30 000	0.94	-6.07
rod L14	3	45 000	2.08	-5.11

**Table 9 : C/E -1 in % using U235-JEF2.2 evaluation, processing NJOY.94**

	number of cycle	Burn up (MWd/t)	U235/ U238	U236/U238
rod I13	2	30 000	1.00	-6.13
rod L14	3	45 000	2.18	-5.15

**Table 10 : C/E -1 in % using U235 Leal Derrien Larson evaluation , processing NJOY.94**

	number of cycle	Burn up (MWd/t)	U235/ U238	U236/U238
rod I13	2	30 000	0.62	-1.69
rod L14	3	45 000	1.59	-0.70

The comparison between the two JEF2 calculations reported in tables 6 and 7 confirm that the effect of processing is very weak on major actinides.

Table 10 demonstrates that the Leal Derrien Larson evaluation improves C/E agreement on U235 depletion and reduces the discrepancy on U236 build-up within the experimental uncertainty margins : from  $-5.6\% \pm 1\%$  to  $-1\% \pm 1\%$  (90% confidence interval).

The following Tables present the U235 capture and fission cross sections and the neutron number emitted by fission ( $\nu$ ), averaged over thermal and epithermal ranges (burnup = 30 Gwd/t, processing NJOY.94).

**Table 11 : Resonance range ( 0.625 eV to 5 Kev):**

evaluation	capture cross section (barn)	fission cross section (barn)	$\nu$
JEF2.2	12.30	23.69	2.434
LDL	13.24	23.45	2.434
(LDL-JEF2.2)/JEF2.2	7.64 %	-1.01 %	0%

**Table 12 : Thermal range (0.11 meV to 0.625 eV):**

evaluation	capture cross section (barn)	fission cross section (barn)	$\nu$
JEF2.2	33.48	196.02	2.438
LDL	33.69	195.01	2.437
(LDL-JEF2.2)/JEF2.2	0.63 %	-0.51 %	-0.041%

The U236 build-up in MOX spectrum is a very powerful validation of the U235 Resonance Integral; due to the hardened spectrum, 70% of the U235 capture arises in resonance range. Therefore, the SLB1 irradiated fuel experiment allows us to conclude that the 7% increase of the (n, $\gamma$ ) Resonance Integral in the LDL evaluation is needed.

## **VI. BUCKLING MEASUREMENTS IN CRITICAL EXPERIMENTS**

A set of five French Critical Buckling Experiments was chosen for this experimental validation. A sixth one was selected from the US experiments in order to emphasize the tight lattice range.

The five first experiments were performed in the French Eole zero power reactor located at Cadarache. All these experiments were carried out in a LWR lattices, with a PWR-type fuel pins, the UO2 enrichments range from 3 to 3.5 per cent in <sup>235</sup>U. The water-to-UO2 volume ratios vary from Rmod=0.96 to Rmod=5.5. The lattice pitches are square. The experimental characteristics are summarized in Table 11.

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The Cristo III programme /10/ deals with hardened spectrum, such as those encountered in dissolver media poisoned with soluble gadolinium. The selected configuration corresponds to a very tight lattice, characterized by  $R_{mod}=96$ .

The Cameleon programme /11/ was built up to represent the existing PWRs, whereas Epicure /8/ was planned for studying mixed oxide core loading (Plutonium recycling in PWRs). For this study, only the reference configuration UH1.2 using UO<sub>2</sub> pins was selected.

The Cristo I, II programme, devoted to fuel storage criticality, has included a large set of experiments. Two regular lattices have been chosen for their accuracy and their significant energy dependent spectra being, mainly well-thermalized spectra.

HiC (high conversion experiments) are a part of the ZPR US programme. Core number 6 has been selected because it completes the undermoderated range, emphasizing the <sup>235</sup>U capture resonance integral.

Three types of calculations have been made for each critical experiment in order to separate the direct effect due to the evaluation from that of the data processing (cf. § II).

The APOLLO2 reference scheme calculation recommended for square pitch lattices has been used (cf. § III).

The results are summarized in Table 13

A 130 pcm discrepancy in terms of  $K_{inf}$  can be observed between the first two sets of results which correspond to the same JEF2.2 evaluation with a different processing (i.e. NJOY89 and NJOY94). For the reactivity, the difference in  $K_{eff}$  depends on the amount of leakage : the processing in the corrected version of NJOY induces a lower energetic fission spectrum, and consequently, a decrease of the migration area  $M^2$ . The fast fission factor decreased by 100 pcm in the case of CAMELEON experiment, and by 124 pcm in the case of UH12.

The comparison of the two right hand columns gives the actual impact of the evaluation for the  $k_{eff}$  multiplication factor calculation.

The first result is that the difference decreases with the slowing down density :

- For the low moderation ratio experiments (CRISTO3 and ZPR-HiC6), one can remark quite a large effect (-800 pcm in the case of CRISTO3 experiment). This is due to the dominant epithermal energy range in these intermediate spectrum experiments : the resonant capture contribution represents 57 % of the total <sup>235</sup>U capture.
- The effect for the medium moderation ratio experiments can be compared to the effect observed in the case of a PWR, and amounts to -400 pcm (the resonant capture component is 30% of the <sup>235</sup>U capture in the PWR mock-up experiment CAMELEON).
- The last two well-moderated experiments enhance the thermal effect, which is quite small :

$$\eta^{235} = \frac{v\sigma_f(^{235}\text{U})}{\sigma_a} \text{ value (averaged on the thermal spectrum of CRISTO2 experiments) is}$$

consistent between the JEF2.2 and the LDL evaluation :

$$\eta_{JEF2}^{235} = 2.082 \quad \text{and} \quad \eta_{LDL}^{235} = 2.080$$

### Conclusion

From these buckling measurements, it can be concluded that both the JEF2.2 and the LDL <sup>235</sup>U evaluation are satisfactory for well-thermalised lattices calculation. However, the French CRISTO3 experiment devoted to HCRs shows that the LDL evaluation is more suited to intermediate spectrum calculation. Furthermore, this conclusion is confirmed by the CAMELEON PWR mock-up experiment, where the C/E disagreement is reduced to the experimental uncertainty level.



**Table 13 : Qualification of the new Leal Derrien Larson (LDL)  $^{235}\text{U}$  evaluation  
using Buckling Measurements in Critical Experiments**

Experimental description						JEF2.2 NJOY89			JEF2.2 NJOY94			LDL NJOY 94		
Name	P	R <sub>mod</sub>	q	B <sup>2</sup>	σ	Keff-1	Kinf	M <sup>2</sup>	Keff-1	Kinf	M <sup>2</sup>	Keff-1	Kinf	M <sup>2</sup>
<i>Cristo3</i>	0.96	0.45	0.37	1.95	500	1470	1.13090	58.73	1307	1.12867	58.52	531	1.11997	58.49
<i>ZPR HiC6</i>	1.24	0.96	0.51	4.747	400	-12	1.19722	41.58	-87	1.19529	41.36	-604	1.18905	41.35
<i>UH1.2</i>	1.26	1.27	0.51	6.05	300	306	1.31438	51.30	309	1.31267	51.01	-181	1.30617	51.00
<i>Caméléon</i>	1.26	1.80	0.57	5.085	300	786	1.29618	56.26	818	1.29491	55.93	434	1.28981	55.90
<i>Cristo2</i>	1.58	3.56	0.76	3.575	300	-259	1.12008	34.40	-291	1.11872	34.12	-481	1.11658	34.12
<i>Cristo1</i>	1.86	5.46	0.89	-0.09	300	362	1.00069	32.44	231	0.99942	32.04	108	0.99818	32.19

Keff-1 : Calculation Experiment biais (pcm) on reactivity

P : Lattice Pitch (cm)

R<sub>mod</sub> : Water to UO<sub>2</sub> volume ratio

B<sup>2</sup> : Critical Buckling (10<sup>-3</sup> cm<sup>-2</sup>)

q : Slowing down density

σ : experimental uncertainty (pcm)

M<sup>2</sup> : Migration area (cm<sup>2</sup>)

## VII. CONCLUSION

The analysis of LWR reactivity experiments using buckling measurements pointed out a better Calculation-Experiment agreement on Keff value when using the U235 LDL evaluation, particularly in undermoderated lattices which are more sensitive to the Resonance Integral.

The analysis of PWR spent fuel demonstrates that the LDL evaluation improves the Calculation of U235 depletion versus burnup. Furthermore, the U236/U238 isotopic ratio in MOX lattice characterized by hardened spectrum indicates that U236 build-up is underestimated by -5% to -6% in depletion Calculation based on the JEF2 evaluation; on the other hand, the use of the proposed LDL evaluation cancels the Calculation-Experiment disagreement:

$$(C-E)/E = -1\% \pm 1\%$$

Thus, we can conclude that the increase of the U235 capture Resonance Integral in LDL evaluation is supported by integral experiments.

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