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Re-Estimation of Nuclear Data and Reliable Covariances using Integral Experiments. Application to JEFF3 Library

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Abstract - *Differential measurements of Nuclear Data (ND) are not enough accurate to meet the target-accuracy required by reactor design calculation. Therefore, integral experiments are needed to improve nuclear data files and to derive reliable covariance matrices. This paper describes the ND re-estimation process based on targeted experiments. The uncoupling between capture, fission and multiplicity data is obtained through French Post-irradiation experiments and critical regular LWR cores in EOLE. JEFF3.1.1 improvement trends as well as associated covariances are summarized for the main actinides.*

I. INTRODUCTION

Differential measurements of Nuclear Data (ND) are not enough accurate in order to meet the target-accuracy required by reactor design calculation [1]. Moreover, the uncertainties associated with the various ND evaluations are inconsistent. Therefore, integral experiments are needed to improve nuclear data files and to derive reliable covariance matrices.

Experimental validation of international ND files is generally based on critical K_{eff} measurements [2], using mainly the International Handbook of Benchmark Experiments ICSBEP [3]. Although it is satisfactory for Criticality-Safety calculation codes [4], this validation is insufficient for Reactor Physics; indeed, nuclear data of minor actinides ($^{238-241-242}$Pu, ^{237}Np, $^{241-243}$Am, $^{244-245}$Cm) have to be validated, particularly for fuel depletion and cycle length calculations [5]. Thus, spent fuel chemical assays also have to be considered in the validation process.

The international library JEFF3.1.1 [6] is currently used in France both in the safety-criticality package CRISTAL [7] and in the EDF [8] and AREVA [9] LWR calculation packages. Therefore, it is essential to validate the main evaluations of this library and to determine the associated realistic uncertainties.

In order to improve the JEFF3.1.1 library and to obtain reliable covariance matrices, targeted experiments are required. Critical experiments such as ICSBEP benchmarks are useful; however the rigorous uncoupling between capture, fission and multiplicity data can be met only through the addition of Post-irradiation experiments (PIE).

From the French experimental database including critical LWR cores and PWR spent fuel chemical analyses, using the ND re-estimation method, this study provides the ND improvement trends as well as the JEFF3.1.1 covariance matrices for the main nuclides.

II. THE INTEGRAL EXPERIMENTAL DATABASE

The selected K_{eff} and Buckling measurements in Low-Enriched UO_2 lattice experiments cover a wide range of moderation ratios. Most of them are LWR-type and have been carried out in the EOLE zero-power reactor at CEA-Cadarache. They are completed by the ZPR-HiC under-moderated experiment, and the LCT-007 experiments carried out at CEA-Valduc, from the ICSBEP handbook. The main characteristics of these LWR-UOX type cores are summarized in Table I, from the harder spectrum (CRISTO3 tight lattice) to well thermalized spectrum (LCT-007 case 4).

Concerning the reactivity of MOX lattices, the EPICURE-MH1.2 core in EOLE was used (MOX 7%Pu fuel pins in 1.26 cm pitch lattice).

Table I. K_{eff} measurements in LWR regular lattices included in the ND re-estimation

Experimental Programme	Lattice pitch (cm)	V_{H2O}/V_{UO2}	Enrichment U235 wt%
CRISTO-3	0.96	0.45	3.3%
ZPR-HiC Al-clad	1.24	0.96	3.0%
ZPR-HiC SS-clad	1.24	0.96	3.0%
EPICURE	1.26	1.25	3.7%
MISTRAL1	1.32	1.75	3.7%
CAMELEON	1.26	1.80	3.5%
LCT-007 case 1	1.26	1.82	4.8%
CRISTO-2 'tight'	1.58	3.56	3.0%
CRISTO-2 'large'	1.71	4.40	3.0%
CRISTO-1	1.86	5.46	3.0%
LCT-007 case 2,3,4	1.60 – 2.52	3.8 – 11.5	4.8%

To uncouple Capture / Fission / Multiplicity, and derive reliable trends on nuclear data, we used isotopic ratios measured in French PWR spent fuels. These PIE experiments on fuel rod cuts consist of measuring the relative concentration of nuclides after irradiation. The samples to be analyzed are extracted from the assembly at each end of cycle. After cooling and transportation, samples are dissolved in a hot acid solution and analyzed mainly by mass spectrometry. These measurements provide isotopic ratios N^{i} / N^{U238} of the isotope i for several burn-up values (the calculated integrated fission rate is normalized to the measured N^{Ndj}/N^{U8} neodymium indicators).

Measured actinides are : $U^{234,235,236}$, Np^{237}, $Pu^{238,239,240,241,242}$, $Am^{241,242m,243}$, $Cm^{243,244,245,246}$. Each experimental value can be considered as an individual integral measurement : more than 400 C/E values from UOX, Enriched Reprocessed Uranium URE and MOX assemblies have been used. Burnup of UOX fuels ranges from 12 up to 85 GWd/t, with the ALIX fuels analyzed after 5, 6 and 7 irradiation cycles [10]. Table 2 summarizes the P.I.E measurements included in the ND re-estimation.

Table II. Spent Fuel measurements used in the ND re-estimation

Fuel **UOX** PWR name	U enrich^t U235 wt %	Number of samples	Burnup range (GWd/t)
Bugey-3	3.10%	7	20 – 39
Cruas-4 URE	3.56%	6	12 – 36
Gravelines-2	4.50%	4	26 – 60
Gravelines-3	4.50%	4	50 – 61
Grav-5 ALIX	4.50%	4	64 – 85
Fuel **MOX** PWR name	Pu enrich^t Pu wt %	Number of samples	Burnup (GWd/t)
Dampierre-2	5.3%	2	52 ; 60
	6.7%	2	53 ; 58
Tricastin-1	9.8%	2	43 ; 56

III. CALCULATION METHODS AND C/E RESULTS

To avoid calculation biases linked to deterministic calculations, K_{eff} values of LWR experimental benchmarks were computed by 3D continuous-energy TRIPOLI4 Monte-Carlo code [11]. An example of TRIPOLI4 geometry is shown in Figure 1 for the Valduc experiment.

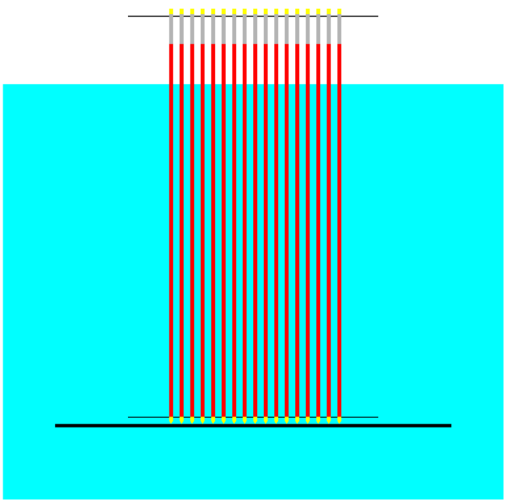

Fig. 1. TRIPOLI4 axial cut-off of the LCT-007 core n°4.

Calculation-Experiment comparison, based on JEFF3.1.1 library, is presented in Table III versus the slowing-down density (neutron fraction slowed below thermal cut-off E=4eV).

Table III. C/E bias in LWR UO_2 lattices

Experimental Programme	Slowing-down Q∞	C-E [pcm]	Exp Unc. 1σ [pcm]
CRISTO-3	0.37	+520	700
ZPR-HiC Al-clad	0.47	-420	500
ZPR-HiC SS-clad	0.49	+ 98	500
EPICURE	0.51	+330	280
MISTRAL1	0.53	+220	220
CAMELEON	0.57	+290	300
LCT-007 case 1	0.59	-210	140
CRISTO-2 'tight'	0.75	-200	400
LCT-007 case 2	0.78	-100	100
CRISTO-2 'large'	0.80	- 80	400
CRISTO-1	0.89	-110	400
LCT-007 case 3	0.92	-250	100
LCT-007 case 4	0.95	-300	100

Isotopic ratios calculations were performed with the deterministic transport code APOLLO2.8 [12] and its CEAv5 library also based on JEFF3.1.1. The SHEM 281-group [13] structure is used, that accounts for detailed description of the first main resonances and avoid self-shielding formalism approximations. 281-group assembly calculations are performed in the exact-2D geometry using the Method Of Characteristics (Figure 2). The validation of this *SHEM-MOC* scheme was carried out against TRIPOLI4 on extensive PWR numerical benchmarks, which demonstrated that APOLLO2.8 calculations are within 0.3% accuracy for the flux in any fuel pin.

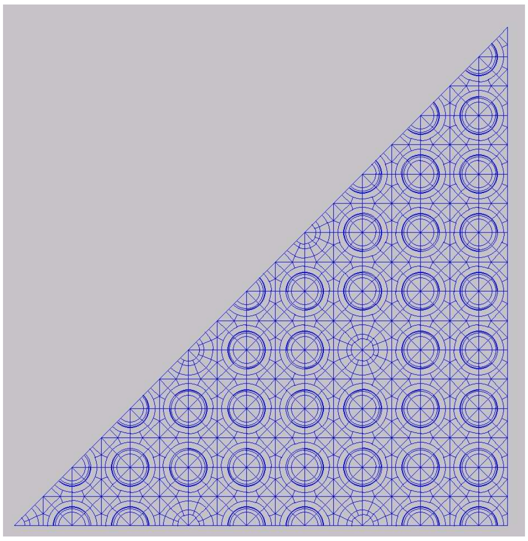

Fig. 2. APOLLO2.8 spatial mesh for PWR 17x17 assembly.

PIE depletion calculations, as well as K_{eff} predictions, are involved in the experimental validation of PWR parameters; therefore the C/E values are reported in the Validation Report of APOLLO2.8 [14].

IV. THE RE-ESTIMATION PROCESS

We developed the RDN code (Nuclear Data Re-estimation) based on a non-linear regression method.

q integral measurements are described by the random vector Y_i $(i=1,\ldots q)$ of experimental values. $Y=n+\epsilon_Y$, with n a vector containing the "true" integral and ϵ_Y a random vector normally distributed, representing the experimental errors. The covariance matrix Σ_Y is associated to ϵ_Y.

The p microscopic data m_j $(j=1,\ldots p)$ are the unknown parameters of the problem, described by $X=m+\epsilon_X$, where the random vector ϵ_X is also centered, normally distributed and associated to ND covariance matrix Σ_X.

The formal relationship between n and m is: n = f(m). The f function relates integral values to the microscopic nuclear data (Boltzmann and Bateman equations).

The mathematical expression can be written as :

$$Z = \eta(m) + \epsilon \qquad (1)$$

with:

$$Z = \begin{pmatrix} X \\ Y \end{pmatrix} \quad \eta(m) = \begin{pmatrix} m \\ n = f(m) \end{pmatrix} \quad \varepsilon = \begin{pmatrix} \varepsilon_X \\ \varepsilon_Y \end{pmatrix} \qquad (2)$$

This problem is viewed in the RDN method as a non-linear regression problem with known covariance matrix.

In order to maximize the Likelihood function, the Gauss-Newton method is used for the minimization of the non-linear square sum. The iterative technique needs a good m_0 initial estimation for parameter m. Each iteration consists in replacing the η function by the approximate formula near the m_k current estimated value:

$$\eta(m) = \eta(m_k) + D\eta(m_k)(m - m_k) \qquad (3)$$

$D\eta(m_k)$ is the Jacobian matrix, also called "generalized sensitivity matrix".

At every step k, the new estimation m_{k+1} is given by:

$$m_{k+1} = m_k + \left\{D\eta(m_k)^t \Sigma^{-1} D\eta(m_k)\right\}^{-1} D\eta(m_k)^t \Sigma^{-1}(Z - \eta(m_k)) \qquad (4)$$

and the posterior covariance matrix of the re-estimated m is:

$$\Sigma_{\hat{m}} = \left\{\Sigma_X^{-1} + Df(m_k)^t \Sigma_Y^{-1} Df(m_k)\right\}^{-1} \qquad (5)$$

V. SENSITIVITIES AND PRIOR COVARIANCES

K_{eff} sensitivity profiles to cross sections / multiplicities have been obtained with APOLLO2.8 using the Standard Perturbation Theory. These 281-group sensitivities were checked against TRIPOLI4/JEFF3.1.1 continuous-energy sensitivity profiles obtained by the IFP method [15]. Keff sensitivities to ^{235}U fission are compared for PWR-type regular cores in Figure 3. Sensitivities to ^{238}U capture for the four LCT007 cores are compared in Figure 4. Sensitivity coefficients on the JEF 15-group structure [16] were derived by integration.

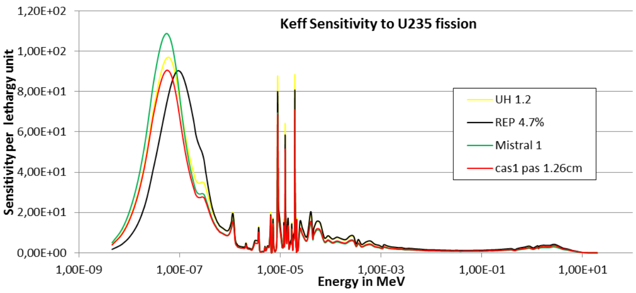

Fig. 3. Sensitivity profiles to ^{235}U(n,f) (UH1.2, PWR-4.7%, Mistral1, LCT007-case1)

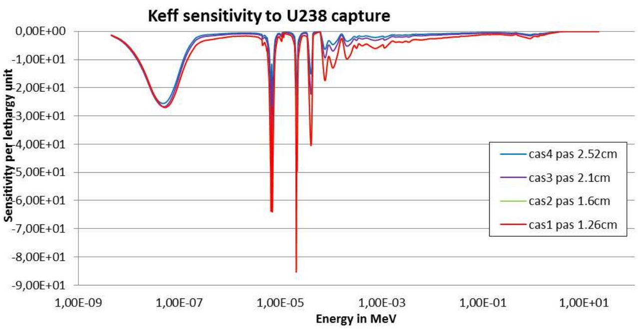

Fig. 4. Sensitivity profiles to ^{238}U(n,γ) for the 4 LCT7 cores

Concerning isotopic ratios, 15-group sensitivity coefficients at every burn-up were obtained by direct APOLLO2 calculations.

In the re-estimation process we paid attention to use realistic ND prior uncertainties to be associated with the JEFF3.1.1 evaluations. The ^{235}U prior covariance matrix was already available from a previous study [17] : Fission and Capture covariance matrices are plotted in Figures 5 and 6. The ^{238}U, ^{239}Pu, ^{240}Pu, ^{241}Am prior covariance matrices were extracted from the Cadarache COMAC-V0 file [18]. Covariance matrices of Pu, Am and Cm minor actinides, were derived from the validation work on previous JEF2.2 library [6].

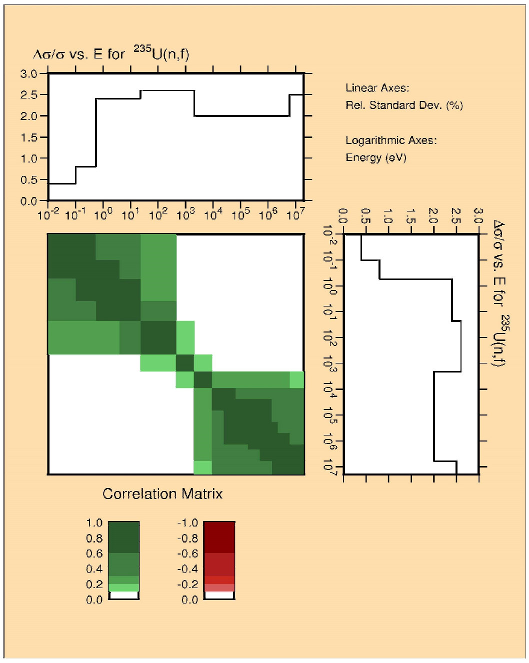

Fig. 5. Prior covariance for ^{235}U(n,f)

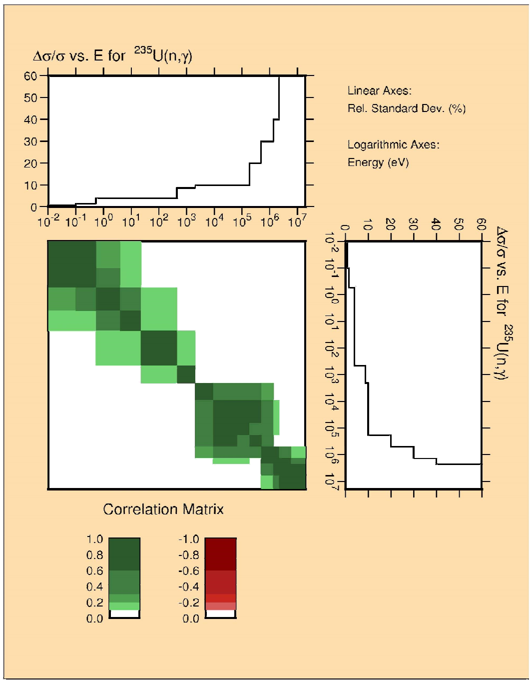

Fig. 6. Prior covariance for ^{235}U(n,γ)

VI. RE-ESTIMATION RESULT AND JEFF3 TRENDS

The JEFF3.1.1 re-estimation results in the relevant groups are summarized in Tables IV (uranium isotopes) and V (plutonium). In these tables, nuclear data modifications in bold are considered to be significant : the uncertainty after re-estimation is significantly reduced, and on the other hand lower (or similar magnitude) than the data modification.

Table IV points out that ^{235}U JEFF3.1.1 evaluation is particularly satisfactory for fission and multiplicity. Posterior standard deviation (3rd column) is the reliable uncertainty to be used in JEFF3.1.1 covariance matrix and propagation uncertainty in LWRs ; it is strongly reduced to ±0.2% for thermal neutrons, thanks to accurate LWR core benchmarks. The re-estimation suggests that ^{235}U capture resonance integral could be increased by about +2% ± 3%.

^{238}U capture cross-sections [19] are satisfactory, and the associated uncertainties are provided by RDN.

A trend to increase ^{236}U(n,γ) is obtained: +3.8%±3.5% on the E_R=5.4eV resonance.

Table IV. ND modification and uncertainty (%) for Uranium

Energy range	Modif. (in %)	Posterior Std. Dev.	Prior Std. Dev.
U235	**capture**		
12.0 keV – 454 eV	+1.5	7.2	8.0
454 eV – 22.6 eV	**+3.4**	**3.6**	**5.0**
22.6 eV – 4.0 eV	**+2.1**	**2.7**	**4.0**
4.0 eV – 0.54 eV	**+2.0**	**2.5**	**3.7**
0.54 eV – 0.1 eV	+0.9	1.1	1.6
< 0.1 eV	+0.3	0.7	1.0
U235	**fission**		
454 eV – 22.6 eV	-1.3	2.2	2.6
22.6 eV – 4.0 eV	-0.9	2.1	2.4
4.0 eV – 0.54 eV	-0.8	2.0	2.4
0.54 eV – 0.1 eV	+0.0	0.6	0.8
< 0.1 eV	+0.1	0.3	0.5
U235	$\boldsymbol{\nu_t}$		
22.6 eV – 4.0 eV	+0.26	0.47	0.7
4.0 eV – 0.54 eV	+0.27	0.34	0.6
0.54 eV – 0.1 eV	+0.22	0.27	0.5
< 0.1 eV	**+0.22**	**0.18**	**0.4**
U236	**capture**		
12.0 keV – 454 eV	+0.6	9.9	10.0
454 eV – 22.6 eV	+5.3	+8.1	10.0
22.6 eV – 4.0 eV	**+3.8**	**3.5**	**5.0**
U238	**capture**		
12.0 keV – 454 eV	-0.0	2.1	2.2
454 eV – 22.6 eV	-0.1	1.7	2.0
22.6 eV – 4.0 eV	+0.2	1.2	1.5
U238	**Fission**		
6.1 MeV – 2.2 MeV	-0.9	2.4	2.5
U238	**n,2n**		
19.6 MeV - 6.1 MeV	**+3.2**	**2.6**	**10.0**

Table V shows that JEFF3.1.1 evaluations of Pu isotopes are satisfactory, particularly ^{239}Pu [20] and ^{241}Pu [21].
It can be noted that ^{239}Pu(n,γ) post-uncertainty (3rd column) is lower than thermal/epithermal values in international covariance files.
The following improvements could be introduced in future evaluations:
- a slight increase of ^{239}Pu capture in the thermal/epithermal range : +1.8% ± 1.2% in the 0.3eV resonance ; on the contrary the resonance integral could be decreased by 2-3%,
- an increase by about +2% of the ^{240}Pu capture in the resolved resonance range, in agreement with the analysis of MOX sample reactivity worth in EOLE and DIMPLE [22],
- an increase by +3.9% ± 2% of the ^{242}Pu E_R=2.7eV resonance integral.

Table V. ND modification and uncertainty (%) for Pu

Energy range	Modif. (in %)	Posterior Std. Dev.	Prior Std. Dev.
Pu239	**Capture**		
12.0 keV – 454 eV	-2.3	3.9	5.1
454 eV – 22.6 eV	-3.1	4.7	6.3
22.6 eV – 4.0 eV	-3.1	4.8	6.4
4.0 eV – 0.54 eV	+0.8	1.0	2.6
0.54 eV – 0.1 eV	**+1.8**	**1.2**	**3.9**
< 0.1 eV	+1.7	1.6	3.9
Pu239	**Fission**		
454 eV – 22.6 eV	+0.2	2.9	3.1
22.6 eV – 4.0 eV	+0.2	2.6	2.8
4.0 eV – 0.54 eV	+0.1	1.1	1.2
0.54 eV – 0.1 eV	-0.1	1.4	1.7
< 0.1 eV	-0.0	0.5	0.6
Pu240	**Capture**		
454 eV – 22.6 eV	+2.3	3.9	4.1
22.6 eV – 4.0 eV	+2.1	3.4	3.6
4.0 eV – 0.54 eV	**+2.6**	**1.7**	**2.7**
0.54 eV – 0.1 eV	+1.7	1.8	2.1
< 0.1 eV	+2.2	1.2	2.0
Pu241	**Capture**		
454 eV – 22.6 eV	-1.6	9.8	10.0
22.6 eV – 4.0 eV	-3.2	6.6	8.0
4.0 eV – 0.54 eV	-0.2	7.0	7.0
0.54 eV – 0.1 eV	+0.1	2.8	5.0
< 0.1 eV	+0.0	1.4	1.5
Pu241	**Fission**		
454 eV – 22.6 eV	-0.5	3.9	4.0
22.6 eV – 4.0 eV	+0.3	2.9	3.0
4.0 eV – 0.54 eV	-0.1	3.0	3.0
0.54 eV – 0.1 eV	-0.4	1.8	2.0
< 0.1 eV	-0.1	1.0	1.0
Pu242	**Capture**		
4.0 eV – 0.54 eV	**+3.9**	**1.8**	**5.0**

Table VI summarizes the re-estimation results for minor actinides :
- due to the under-prediction of ^{238}Pu content in UOX irradiated fuels, an increase of the ^{237}Np epithermal capture is proposed; however, compared to the ±5% posterior uncertainty, this +3% modification is not a reliable trend,
- a trend to increase by +2.4% ± 2.4% the ^{241}Am epithermal capture is obtained, in agreement with measurement [23],
- an increase trend by +4.3% ± 2.8% on the ^{243}Am large resonance (E_R=1.4eV) is raised,
- ^{244}Cm evaluation in JEFF3.1.1 is very satisfactory, particularly for the E_R=7.7eV resonance integral,
- ^{245}Cm(n,γ) thermal value can be improved by a +8% ± 3.8% increase.

Table VI. ND trends and uncertainties (%) for Np, Am, Cm

Energy range	Modif. (in %)	Posterior Std. Dev.	Prior Std. Dev.
Np237	**Capture**		
4.0 eV – 0.54 eV	+3.2	6.4	7.0
0.54 eV – 0.1 eV	+2.5	4.4	5.0
< 0.1 eV	+0.4	2.9	3.0
Am241	**Capture**		
22.6 eV – 4.0 eV	+3.2	3.4	4.9
4.0 eV – 0.54 eV	**+2.5**	**2.4**	**3.7**
0.54 eV – 0.1 eV	**+2.3**	**2.4**	**3.7**
< 0.1 eV	+1.5	3.9	4.5
Am243	**Capture**		
22.6 eV – 4.0 eV	+1.6	6.9	7.0
4.0 eV – 0.54 eV	**+4.3**	**2.8**	**5.0**
Cm244	**Capture**		
454 eV – 22.6 eV	+0.3	8.9	9.0
22.6 eV – 4.0 eV	**+0.8**	**3.2**	**6.0**
Cm245	**Fission**		
< 0.1 eV	-0.1	2.6	2.7
Cm245	**Capture**		
< 0.1 eV	+8.1	3.8	5.0

Concerning H_2O cross-sections, small modifications were suggested : -0.7% ± 0.7% for the thermal scattering in the 0.5-4 eV energy range, and -0.16% ± 0.28% for the thermal capture.

Furthermore, uncertainty correlation between energy groups is strongly reduced, compared to prior correlation matrix, thanks to integral measurements in various spectra. Prior and posterior correlations are compared in Table VII for ^{235}U capture cross-section in the epithermal/thermal energy range: uncertainty correlations between groups are reduced by about a factor 2. Concerning ^{235}U fission cross-section, prior and posterior correlations are compared in Table VIII.

Table VII. Posterior and *Prior* correlations for ^{235}U(n,γ)

Energy	23–4eV	4–0.5eV	0.5–0.1eV	< 0.1 eV
23eV–4eV	1.	0.37 *0.70*	0.07 *0.50*	0.13 *0.50*
4eV–0.5eV	0.37 *0.70*	1.	0.45 *0.70*	0.10 *0.50*
0.5 – 0.1eV	0.07 *0.50*	0.45 *0.70*	1.	0.44 *0.70*
< 0.1 eV	0.13 *0.50*	0.10 *0.50*	0.44 *0.70*	1.

Table VIII. Posterior and *Prior* correlations for ^{235}U (n,f)

Energy	23–4eV	4–0.5eV	0.5–0.1eV	< 0.1 eV
23eV–4eV	1.	0.60 *0.70*	0.10 *0.30*	0.09 *0.25*
4eV–0.5eV	0.60 *0.70*	1.	0.65 *0.70*	0.09 *0.30*
0.5 – 0.1eV	0.10 *0.30*	0.65 *0.70*	1.	0.84 *0.90*
< 0.1 eV	0.09 *0.25*	0.09 *0.30*	0.84 *0.90*	1.

VII. CONCLUSION

This paper has described the ND re-estimation process based on targeted experiments. The use of integral measurements in the re-estimation of JEFF3.1.1 evaluations has provided some ND improvement trends. The uncoupling between capture, fission and multiplicity data was obtained through French Post-irradiation experiments and critical regular LWR cores in EOLE and Appareillage-B. Realistic covariance matrices were derived for the main isotopes, that allows reliable uncertainty propagation studies.

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