International Conference on Nuclear Data for Science and Technology 2007
DOI: 10.1051/ndata:07708

^{239}Pu nuclear data improvements in thermal and epithermal neutron ranges

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Abstract. The analysis of k_{eff} multiplicative factor of 100%-MOx cores and plutonium solutions showed a systematic discrepancy between calculation and experiment. The aim of this paper is to propose an improved version of the JEFF-3.1 ^{239}Pu evaluation in the low-energy neutron range. We propose here slight modifications of the evaluation file, consistent with the uncertainties of differential data measurements in sub-thermal/thermal and epithermal ranges, namely: modifications of the sub-thermal fission, capture and total cross sections, and a revised evaluation of ν_p from sub-thermal up to 20 eV using a phenomenological formalism. The proposed modifications are described and tested against integral measurements.

1 Introduction

^{239}Pu nuclear data accuracy is an important issue for reactor applications. k_{eff} analysis of Pu-fuelled systems showed systematic overestimation of the calculated core reactivity. The first part of this paper stresses the need for improvement. In the second, the modification of cross sections and the modification of the mean number of fission neutrons are described. Eventually, the integral experiment validation is presented.

2 Need for improvement

The good prediction of ^{239}Pu and ^{240}Pu contents in Post Irradiation Experiments Analysis mainly in PWR-MOx spent fuels [1] underlines the good accuracy of the JEFF-3.1 resonance absorption and capture integral [2]. However, a systematic overestimation of calculated k_{eff} values (using Monte Carlo codes) is observed in the analysis of critical experiments. The iso-thermal moderator temperature analysis shows discrepancies between calculation and experiment as well. Owing to sensitivity studies, such independent integral experiments point out the need for improvement of neutron induced ^{239}Pu sub-thermal data (σ_γ, σ_f, ν_p), thermal data (ν_p) and epithermal data (ν_p).

2.1 Analysis of multiplicative factor measurements

Two sources of critical experiments were considered in this study. First, the International Handbook of Evaluated Criticality Safety Benchmark Experiments [3] supplied various experiments with a wide range of neutron spectrum. The second was provided by reactor mock-up experiments performed in dedicated facilities such as EOLE at Cadarache.

The experimental validation of Monte Carlo codes through those integral experiments gives clear trends on ^{239}Pu thermal nuclear data within technological uncertainties ranging from ± 200 pcm to ± 500 pcm at 1σ. Such integral experiments permit us to constrain the well-posed inverse problem and to improve ^{239}Pu nuclear data.

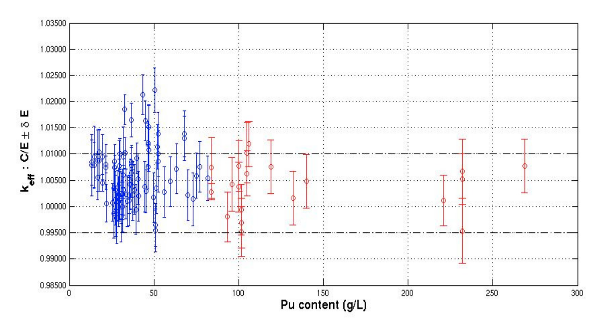

Fig. 1. MCNP analysis of ICSBEP/PST benchmarks versus ^{239}Pu amount with JEFF-3.1 nuclear data.

Plutonium-Solution-Thermal (PST) benchmarks of the ICSBEP database, performed in the 60's, consist in critical measurements, of (HNO_3+ ^{239}Pu) liquid spheres with various diameters and various plutonium amounts. The JEFF-3.1 MCNP [4] analysis [5] shows a systematic overestimation from +1200 pcm down to +200 pcm as a function of the ^{239}Pu concentration (see figure 1). Notice that ^{239}Pu concentration infers a strong neutron spectrum hardening. The averaged overestimation by about (+700 ± 200) pcm in the low ^{239}Pu concentration range ($C_{Pu} < 80$ g/l) shows that $\eta = \nu \cdot \sigma_f/(\sigma_\gamma + \sigma_f)$ value should be revised in the sub-thermal neutron range.

Critical LWR-MOx lattices in EOLE facility at Cadarache were performed during the MISTRAL [6] and BASALA [7,8] experimental programs. Besides the poorly predicted plutonium ageing, the JEFF-3.1 experimental validation using the TRIPOLI4 [9] Monte Carlo code shows a systematic overestimation of the whole fresh cores k_{eff} by about (+260 ± 200) pcm for various moderation ratios or void fractions (see table 1).

2.2 Moderator temperature coefficient measurements analysis

Measurements of the Isothermal Temperature Coefficient (mainly driven by Moderator Temperature Coefficient: MTC)

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Table 1. TRIPOLI4 analysis of EOLE critical lattices.

EOLE mock - up	Pu Ageing	Moderation Ratio	(C - E) ± (δ E) (pcm)
MH1.2 (PWR - MOx mixed core)	4 years	MR = 1.2	280 ± 250 (1σT4 = 20pcm)
MISTRAL - 2 (PWR - MOx)	8 years	MR = 1.7	630 ± 250 (1σT4 = 20pcm)
MISTRAL - 3 (PWR - MOx)	10 years	MR = 2.1	710 ± 250 (1σT4 = 20pcm)
BASALA - Hot (BWR - MOx)	12 years	42% void	610 ± 250 (1σT4 = 20pcm)
BASALA - Cold (BWR - MOx)	13 years	0% void	700 ± 250 (1σT4 = 20pcm)
FUBILA - Hot (BWR - MOx)	1 year	0% void	250 ± 250 (1σT4 = 20pcm)

Table 2. Reich-Moore parameters of the added (^{239}Pu+n)* bound level.

$E_0 = -20$meV	$J^{\pi} = 0^{+}$
$\Gamma_n = 10^{-7}$meV	$\Gamma_\gamma = 6$meV
$\Gamma_{f1} = -36$meV	$\Gamma_{f2} = 0$eV

of 100%-MOx cores were performed in the EOLE facility in cold (20 °C–80 °C) and hot operation conditions (150 °C–300 °C). Experimental validation of the APOLLO2 [10] deterministic code, using both JEF-2.2 and JEFF-3.1 based libraries, demonstrates [7,11]:

- a systematic underestimation of the MTC in cold conditions by about (–2.0 ± 0.3) pcm/°C,
- a well-assessed MTC in hot conditions (+1.0 ± 2.0) pcm/°C.

The analysis of physical phenomena [11] has shown that the negative error in the low-temperature range is linked to the thermal spectrum shift effect, which is strongly dependant on the sub-thermal and thermal shapes of the plutonium cross sections. The ^{239}Pu $\alpha = \sigma_\gamma/\sigma_\mathrm{f}$ proposed modification in the thermal range is shown in figure 2.

3 Revised ^{239}Pu neutron-nuclear data

Accurate independent integral experiments show the required enhancement of ^{239}Pu neutron-nuclear data, namely sub-thermal cross sections level and shape and the mean number of emitted neutrons per fission.

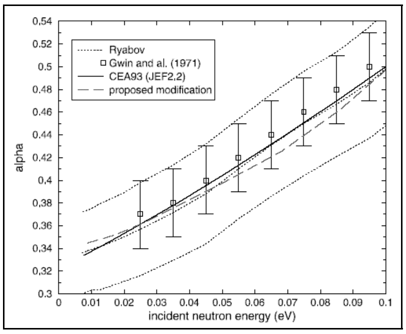

Fig. 2. Proposed modification of the ^{239}Pu α value compared to JEF-2.2 and experimental values.

3.1 Sub-thermal neutron capture and fission cross sections

Integral experiments have highly constrained the possible improvement of ^{239}Pu in the sub-thermal energy range. The a priori differential uncertainties in this energy range are about a few percent (see the dispersion of EXFOR measurements in figs. 3, 4 and 5 for total, capture and fission cross sections respectively). The given uncertainty of thermal values is 2 barns for capture and 1 barn for fission (1σ). The way to modify the cross section leaving unchanged the thermal and epithermal range is to adopt a new negative resonance close to the neutron separation energy. In JEFF-3.1, the ^{239}Pu evaluation of resonance parameters below 2.5 keV was performed by Derrien et al. [12] using the Reich-Moore formalism implemented in the SAMMY code [13]. Due to the interference between resonances in this formalism, the parameters of the added bound level have to be carefully chosen, namely the spin-parity values and the phase/amplitude of the two fission widths. The adopted parameters are summarized in table 2.

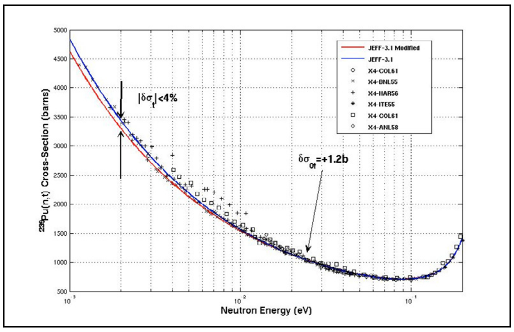

Fig. 3. Experimental, JEFF-3.1 and revised ^{239}Pu total cross section.

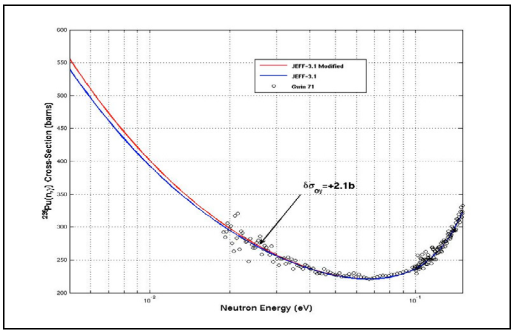

Fig. 4. Experimental, JEFF-3.1 and revised ^{239}Pu(n,γ) cross section.

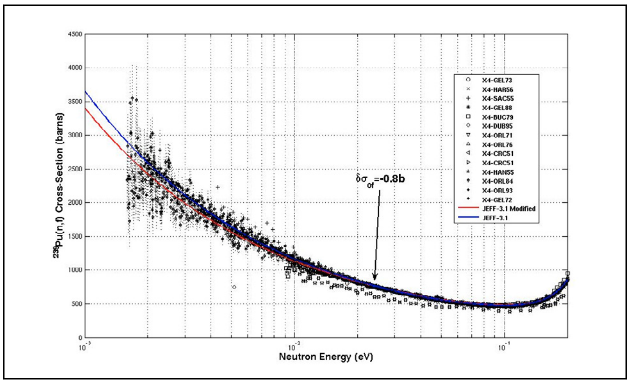

Fig. 5. Experimental, JEFF-3.1 and revised ^{239}Pu(n,f) cross section.

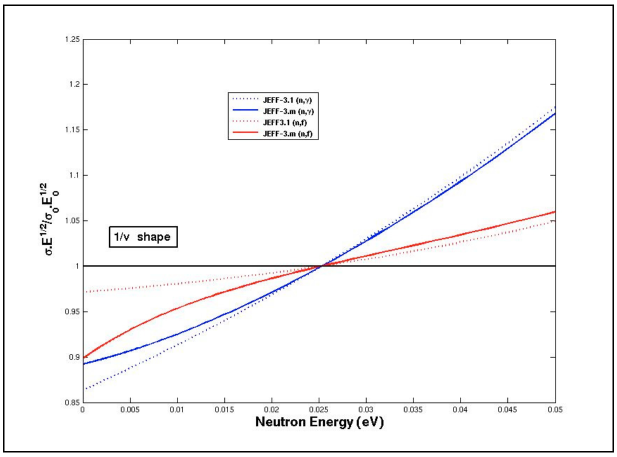

Fig. 6. Revised ^{239}Pu cross section shapes compared to “$1/v$” and JEFF-3.1 shapes.

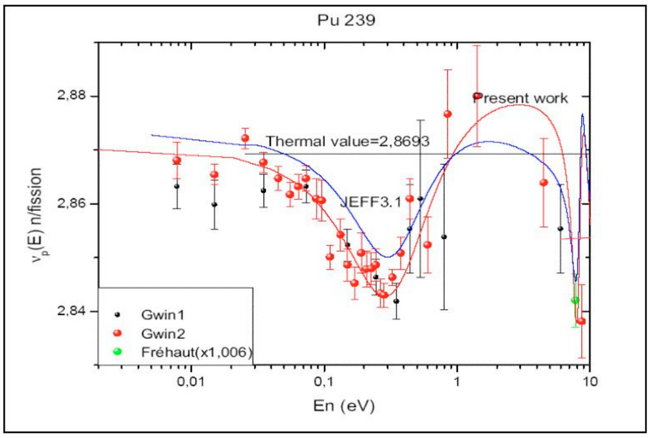

Fig. 7. Revised ^{239}Pu prompt neutron number per fission compared to JEFF-3.1 and experimental values.

The modified shape of sub-thermal ^{239}Pu cross sections are shown in figures 3, 4 and 5. Thermal capture and fission cross sections are modified by +2 barns and −1 barn respectively. The effective scattering radius was left unchanged; the scattering cross section is not affected.

As shown in figure 6, the product $\sigma(\mathrm{E})\cdot\sqrt{E}$ for the revised and for JEFF-3.1 original fission and capture, exhibits their “non $1/v$” behaviours.

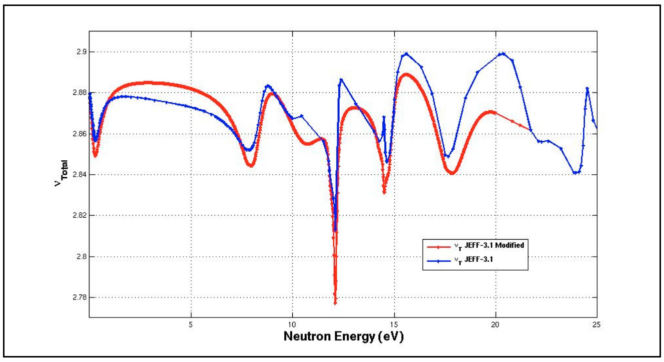

Fig. 8. Revised ^{239}Pu prompt neutron number per fission compared to JEFF-3.1 evaluation.

Table 3. Integral validation of ^{239}Pu revised nuclear data.

	JEFF-3.1evaluation	Present work
ICSBEP/PST sub-thermal systems	(+700±200)pcm	(+200±200)pcm
ICSBEP/PST epithermal systems	(+340±200)pcm	(0±200)pcm
Fresh 100%MOx-PWR type	(+260±200)pcm	(+130±200)pcm

This modification of the sub-thermal fission to capture ratio improves the MTC prediction. Revised cross sections are in accordance with the 1σ a priori differential experiments uncertainties.

3.2 Mean number of prompt neutrons per fission

The cross section modification is not sufficient enough to explain the $\mathrm{k_{eff}}$ overestimation in critical experiment analysis. Indeed, the ν_{p} needs to be revised as well.

A phenomenological formalism [14,15] is applied to ν_{p}. It consists first in breaking down the fission process into independent partial waves described with various reachable J^π values of the compound nucleus. Each J^π channel formation is associated to a probability expressed in term of fission channel cross sections. Hence, the total number of prompt neutrons is a linear combination of the number of prompt neutrons of each channel. Second, for a given channel, neutrons can be emitted by direct fission and after (n,γf) processes. The number of emitted neutrons after (n,γf) processes accounts for the reduced excitation energy after the electromagnetic transition. The decrease of prompt neutrons with the (n,γf) process is assumed to be proportional to the decrease of the compound-nucleus excitation energy. The formalism is summarized into the following equation:

$$\nu = \sum_{J^\pi} \left[\begin{array}{l} \nu_0^{J^\pi} \dfrac{\sigma_{n,f}^{J^\pi} - \sigma_{n,\gamma f}^{J^\pi}}{\sigma_f} \\ + \dfrac{\sigma_{n,\gamma f}^{J^\pi}}{\sigma_f} \left(\nu_0^{J'^{\pi'}} - E_\gamma^{J^\pi} \cdot \left(\dfrac{\partial \nu}{\partial E*} \right)^{J'^{\pi'}} \right) \end{array} \right] \tag{1}$$

- $\nu_0^{J^\pi}$: number of prompt neutrons emitted by direct fission of a compound nucleus J^π

- $\nu_0^{J'^{\pi'}}$: mean number of prompt neutrons emitted by fission of a compound nucleus in $J'^{\pi'}$ state after pre-fission γ emission. $\nu_0^{J'^{\pi'}} = \nu_0^{J^\pi}$ is assumed
- σ_f : total fission cross section including (n,γf) reactions
- $\sigma_{n,f}^{J^\pi}$: direct fission cross section of partial wave l, J
- $\sigma_{n,\gamma f}^{J^\pi}$: (n,$\gamma$f) cross section of partial wave l, J
- $E_{n,\gamma f}^{J^\pi}$: total energy of pre-fission γ transitions
- $(\partial\nu/\partial E * J')^{\pi'}$: mean number of prompt neutrons emitted by direct fission of a compound nucleus $J'^{\pi'}$ per unit of its excitation energy.

Only $l = 0$ neutron wave is considered here. Hence, $J^\pi = 0^+$ or 1^+ are reachable states for the $(^{239}\text{Pu+n})^*$ compound nucleus system. $\nu_0^{0^+} = 2.890$ and $\nu_0^{\Gamma} = 2.859$ are fitted on experimental ν. The work of Shackleton [14] enables to assign $\langle\partial\nu/\partial \text{E}\rangle = 0.131$ n/MeV whatever the J^π values.

This formalism is applied from sub-thermal neutrons up to $\text{E}_\text{n} = 20\,\text{eV}$ [16]. The modified, JEFF-3.1 original and available experimental values of prompt fission neutron number are shown in figures 7 and 8.

The modified mean number of prompt neutrons is less than in JEFF-3.1 evaluation by about −200 pcm in the thermal range and close to the resonance energy peaks. This leads to a clear improvement for the neutron multiplicative factor prediction.

4 Integral validation of ^{239}Pu revised nuclear data

The modified nuclear data namely sub-thermal cross sections and $\nu_\text{p}(\text{E}_\text{n} < 20\,\text{eV})$ values improve:

- the neutron multiplicative factor prediction by about 200 pcm for PWR type systems up to 500 pcm for sub-thermal spectrum systems,
- the cold operation MTC by about +0.3 pcm/°C.

Finally, averaged discrepancies of k_eff prediction are summarized in table 3.

k_eff discrepancies between calculation and experiment are cancelled using the ^{239}Pu revised nuclear data.

5 Conclusion

The aim of this paper was to describe the revised ^{239}Pu nuclear data taking into account independent integral experiments analysis. Sub-thermal level and shape of fission and capture cross sections are modified within 1σ differential uncertainties. A phenomenological formalism for prompt fission neutron number description is applied up to 20 eV. The modified ^{239}Pu evaluation in ENDF format is available at the OECD/NEA in the JEFF-3.2β file.

Integral validation emphasizes the cancellation of calculation/experiment discrepancies, namely for the k_eff of PWR-MOx whole fresh cores.

Further studies would be valuable, namely:
measuring the shape of α and η value in the sub-thermal range, understanding the fluctuations of prompt neutrons in the resonance range [17].

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