

## **Actinide Neutron Induced Cross-Sections; Analysis of the OSMOSE LWR-UO<sub>2</sub> Experiment in MINERVE.**

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### **Abstract**

This paper describes the interpretation of the first phase of the OSMOSE experimental program. The OSMOSE experiment began in 2005 in the MINERVE French facility and will continue until 2008. It consists in reactivity worth measurements of separated actinides by an oscillation technique. First results are obtained in a standard LWR neutron spectrum (UO<sub>2</sub> lattice).

The present study focuses on the following isotopes: <sup>234,236</sup>U, <sup>237</sup>Np, <sup>239,242</sup>Pu. The comparison between APOLLO2 accurate deterministic calculations and experiments shows the reliability of the latest JEFF-3.1 European nuclear data library for all oscillated isotopes, except <sup>237</sup>Np. The obtained (C/E-1)±(δE/E) values are the following:

$$\begin{array}{ll} {}^{234}\text{U}: & -5\% \pm 2\% & {}^{237}\text{Np}: & -11\% \pm 2\% \\ {}^{239}\text{Pu}: & +1\% \pm 2\% & {}^{242}\text{Pu}: & +2\% \pm 2\% \end{array}$$

An energetic decomposition of the reactivity worth is carried out using Standard Perturbation Theory that underlines the underestimation of the <sup>237</sup>Np(n,γ) thermal and resonant capture cross-section.

**KEYWORDS:** *Actinide, Neutron Cross-section Validation, APOLLO2, JEFF*

### **1. Introduction**

The need for accurate nuclear data on minor actinides has been stressed by various organizations throughout the world. The OSMOSE program [1] proposes to measure actinide energy-integral nuclear data. Then, the comparison to differential available data provides a direct assessment of cross-section deficiencies. This paper described the experimental validation of European nuclear data files [2] (JEF-2.2 and JEFF-3.1) through the interpretation of the OSMOSE experiment with the French APOLLO2 deterministic code.

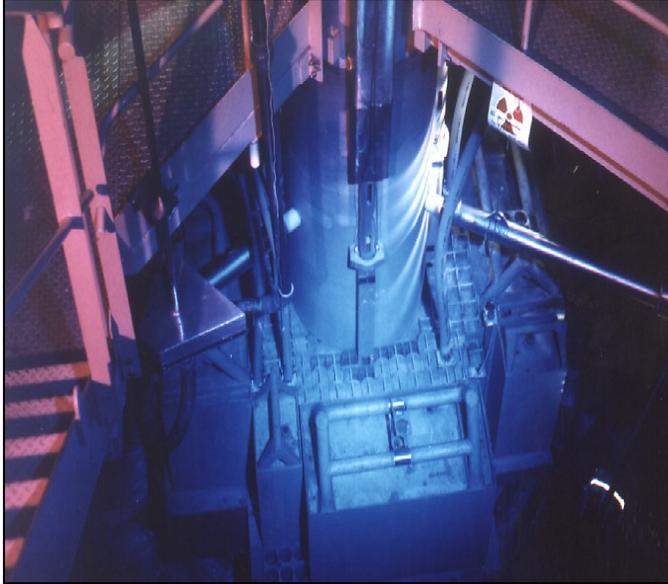
The first part of this paper is devoted to a brief description of reactivity worth measurement of separated actinides by the oscillation technique achieved in the MINERVE reactor. The second part summarizes the methodology to interpret the OSMOSE experiment, such as the calculation scheme used and the calibration factor between calculated and experimental results. The last part discusses the cross-section experimental validation and gives some trends on actinide data.

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## 2. Experimental Settings

### 2.1 The MINERVE Facility



The experimental reactor MINERVE is devoted to neutronics studies of different reactor types. MINERVE achieved its first criticality in 1959 at the center of Fontenay-Aux-Roses and was transferred to CEA-Cadarache in 1977. MINERVE is a zero-power (<100W) pool reactor. The MTR core is submerged under 3 meters of water and is used as a driver zone for the different experiments located in a central cavity. Several lattices corresponding to different neutron spectra can be loaded in this cavity, such as an overmoderated spectrum, two LWR spectra (UO<sub>2</sub> or MOX fuel), two intermediate spectra (MORGANE/S and R blocks for HCLWR studies) and a fast spectrum.

### 2.2 The Experimental Technique

The technique consists in oscillating samples (separated isotope in a UO<sub>2</sub> matrix) at the center of the experimental lattice in order to measure the associated reactivity variation. The reactivity worth is measured within 1% (1 $\sigma$ ) accuracy. The flux variation induced by the oscillation is detected in the driver zone by a boron chamber, which is servo-driven to the rotary automatic pilot rod (using the overlap of cadmium sectors). The experimental value corresponds to the angle of the rotor/stator reached by the pilot rod for the neutronics equilibrium and is linear versus the reactivity worth of the oscillation sample.

### 2.3 The OSMOSE Experimental Program

The OSMOSE experimental program consists in measuring the reactivity worth of separated actinides from <sup>232</sup>Th to <sup>244</sup>Cm by the oscillation technique in several lattices. At the moment, oscillations have been performed in the LWR UO<sub>2</sub> lattice (3%w/o <sup>235</sup>U; pitch: 1.26cm). Each oscillated sample is a double cladded column of 10 fuel pellets (<sup>nat</sup>UO<sub>2</sub> sintered ceramic matrix doped with the separated isotope) with standard PWR dimensions. High accuracy chemical and isotopic analysis are performed for each sample during every step of the fabrication process. Furthermore, for each sample, three control fuel pellets are fabricated in parallel to perform destructive radio-chemical analysis. Finally, 1% (1 $\sigma$ ) systematic error due to material balance knowledge can occur throughout the interpretation.

The OSMOSE oscillation program began in 2005. Current available results are the oscillation measurements in the LWR-UO<sub>2</sub> lattice of one <sup>234</sup>U sample, one URE (Enriched Reprocessed Uranium) sample, two <sup>237</sup>Np samples (two concentrations spanning High Burnup fuel up to Transmutation content), one <sup>239</sup>Pu sample and one <sup>242</sup>Pu sample.

### 3. Methodology of Interpretation using the APOLLO2 code

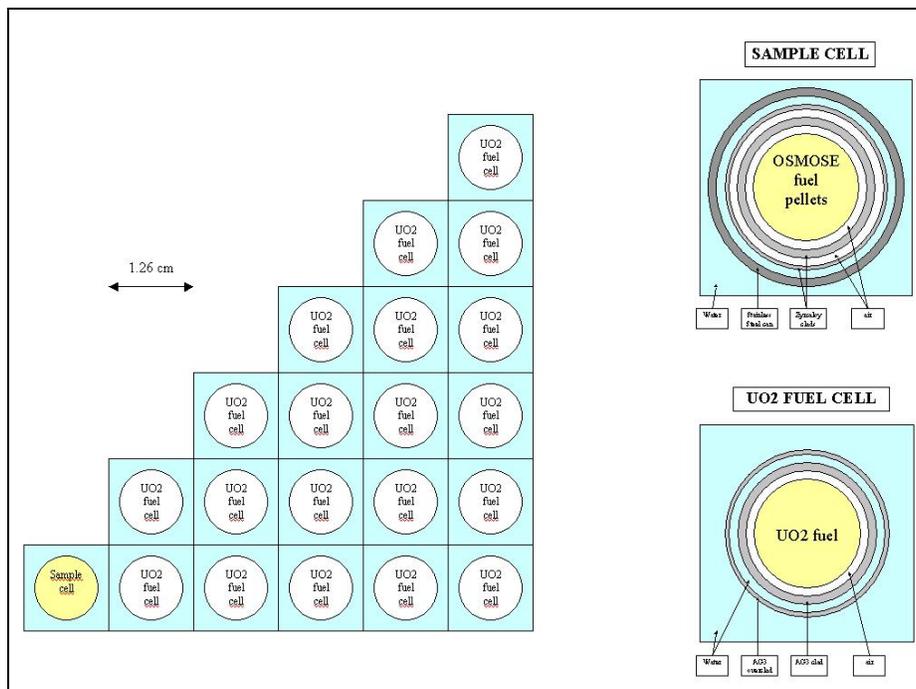
The interpretation is based on the French deterministic transport code APOLLO2 [3,4].

Reactivity worth is obtained using the exact perturbation theory : 
$$\delta\rho = -\frac{\langle \phi^*, \delta H \phi' \rangle}{\langle \phi^*, F' \phi' \rangle}$$

#### 3.1 Calculation Route

The APOLLO2 code is a modular code, which solves both the Boltzmann integral equation and the integro-differential equation. The recent APOLLO2 version uses two nuclear data libraries, which are processed from the European previous JEF-2.2 and new JEFF-3.1 files. Two multigroup structures are available : the XMAS-172g and the new SHEM-281g meshing [5] (which avoids mutual and self-shielding treatments below 22eV). The APOLLO2 reference scheme "CEA-97" was defined by selecting the code options that yield known and acceptable errors [6] (consistent with target accuracy). The analysis of the OSMOSE experiment allows a limited multicell geometry: a 2D 11x11 cells pattern is used (Figure 1). The pattern is a regular UO<sub>2</sub> lattice including the oscillation tube and sample in the central cell.

**Figure 1:** APOLLO2 Calculation Pattern for OSMOSE Experiment Analysis



The space-dependent self-shielding calculation (4 rings in every fuel pin) for resonant isotopes is performed using the Probability Table method. Modeling errors were assessed by comparison with the continuous-energy Monte-Carlo calculation TRIPOLI4 [7]. APOLLO2 calculation uses P<sub>ij</sub>-2D-UP1 model (interface current method with linear anisotropic angular fluxes).

In order to compare the calculated value ( $k$ -effective variation  $\Delta k_{\text{eff}}/k_{\text{eff}}$  in  $10^{-5}$ =pcm) to the experimental value (Pilot Units PU) of a sample oscillation, we have to calibrate the calculated

signal versus the experimental signal.

### 3.2 Calibration Factor between Calculation and Experimental Worth

The calibration factor  $\alpha$  links a calculated reactivity ( $\rho_c$ ) and a measured reactivity ( $\rho_m$ ) as follows:  $\rho_m = \alpha \times \rho_c + \beta$ . The determination of the calibration factor  $\alpha$  is achieved through the reactivity worth measurement of  $^{235}\text{U}$  and  $^{10}\text{B}$  (which assumes that nuclear data are accurately known for these two isotopes).

The calibration factor is carried out from the measured and calculated reactivity worth of the following oscillation samples:

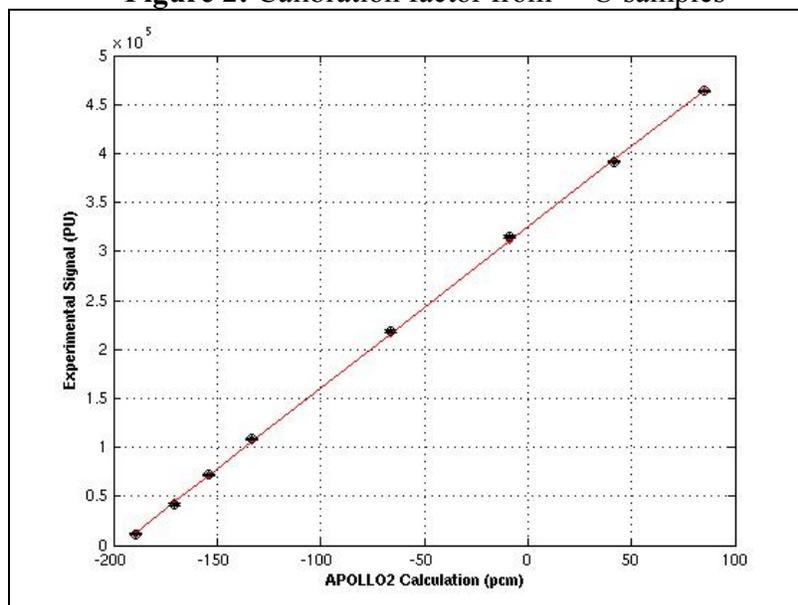
- 8  $\text{UO}_2$  samples with increasing enrichment (0.2% up to 5%) which allows experimental signal calibration through  $^{235}\text{U}$  nuclear data,
- 3  $\text{UO}_2$  samples (with a fixed  $^{235}\text{U}$  enrichment of 0.25%) with increasing boron content ( $^{10}\text{B}/\text{U}$  from 0 up to  $500 \times 10^{-6}$ ) that allows signal calibration through  $^{10}\text{B}(n,\alpha)$  capture,
- 2  $\text{UO}_2$  samples (with a fixed  $^{235}\text{U}$  enrichment of 0.53%) with increasing boron concentration ( $^{10}\text{B}/\text{U}$ : from 0 up to  $500 \times 10^{-6}$ ), which allows  $\alpha$  calibration through  $^{10}\text{B}$ .

Then, three calibration factors  $\alpha \pm \delta\alpha(1\sigma)$  are obtained using least-square linear fitting, one for  $^{235}\text{U}$  (positive or fissile signal) and two for  $^{10}\text{B}$  cross-section (negative or absorber signal). The overall uncertainty of each calibration factor is the quadratic combination of:

- the measured signal uncertainty (see ref. [8] for details),
- the least-square linear regression uncertainty,
- the material balance uncertainty on the  $^{235}\text{U}$  and  $^{10}\text{B}$  calibration samples,
- the uncertainty due to  $^{235}\text{U}$  and  $^{10}\text{B}$  nuclear data (assessed from JEFF-3.1/JEF-2.2 bias).

The calibration factor for JEFF3.1/SHEM calculation, derived from  $^{235}\text{U}$  samples by linear regression shown in Figure 2, is:  $\alpha_{235\text{U}} = 1675 \pm 28 \text{ PU}/\text{pcm}$ .

**Figure 2:** Calibration factor from  $^{235}\text{U}$  samples



### 3.3 Length Correction for Boron calibration samples

The APOLLO2 bi-dimensional calculation model does not take into account different axial geometries of the samples. The external length of all the samples is the same (aluminum container  $H = 103.5$  mm), however the fissile length  $L_s$  is sometimes higher (boron calibration samples) than a reference length value  $L_R=9.4$ cm which corresponds to the length of OSMOSE samples. Then, a correction called Length Correction ( $C_L$ ) is applied to the experimental value :

$$\rho'_m = \rho_m \times C_L$$

with

$$C_L = \frac{\int_{V_s} \Phi(\vec{r}, z)\Phi^+(\vec{r}, z)d\tau}{\int_{V_R} \Phi(\vec{r}, z)\Phi^+(\vec{r}, z)d\tau}$$

$$\text{and} \quad \int_V \Phi(\vec{r}, z)\Phi^+(\vec{r}, z)d\tau \propto \int_{-L/2}^{+L/2} \cos^2(B_z \cdot z)dz$$

$$\text{then} \quad C_L = \frac{[\cos(B_z \cdot L_s)\sin(B_z \cdot L_s) + B_z L_s]}{[\cos(B_z \cdot L_R)\sin(B_z \cdot L_R) + B_z L_R]}$$

using  $\phi, \phi^+$  respectively the nominal direct and nominal adjoint fluxes and  $B_z^2=0.193 \times 10^{-2} \text{cm}^{-2}$ . For boron calibration samples,  $C_L=1.06$ . The uncertainty due to this length correction is negligible.

Finally, the calibration factor averaged on  $^{235}\text{U}$  and Boron samples is:

$$\langle \alpha \rangle = 1688 \pm 22 \text{ PU/pcm.}$$

## 4 OSMOSE Qualification Results and Nuclear Data trends

The difference between an admixed sample signal ( $^{Nat}UO_2+Actinide$ ) and the  $^{Nat}UO_2$  sample signal corresponds to the actinide reactivity worth. The experimental validation of this difference permits us to provide trends on actinide nuclear data.

### 4.1 Qualification Results

Results of the experiment analysis are summarized in Table 1:

**Table 1:** OSMOSE Qualification Results.

Isotope	$(C/E-1) \pm (\delta E/E) (1\sigma)$ (in %)	
	JEF-2.2	JEFF-3.1
$^{234}U$ (0.3g)	$+0.7 \pm 2.0$	$-5.3 \pm 2.0$
Reprocessed Uranium	$+0.1 \pm 1.8$	$-1.5 \pm 1.8$
$^{237}Np$ (0.1g)	$-8.6 \pm 2.2$	$-13.1 \pm 2.2$
$^{237}Np$ (0.6g)	$-5.9 \pm 1.8$	$-10.9 \pm 1.8$
$^{239}Pu$ (0.6g)	$-0.5 \pm 1.8$	$+1.0 \pm 1.8$
$^{242}Pu$ (0.5g)	$+2.2 \pm 2.1$	$+1.9 \pm 2.1$

The  $(\delta E/E)$  uncertainty takes into account:

- the statistical measurement uncertainty of the signal difference,
- the statistical experimental material balance uncertainties (about 1% mainly due to actinide content),
- the systematic calibration uncertainty (1.3%).

### 4.2 Trends on Nuclear Data

Exact Perturbation Theory supplies the sensitive energy range of different neutron reactions of such reactivity worth:

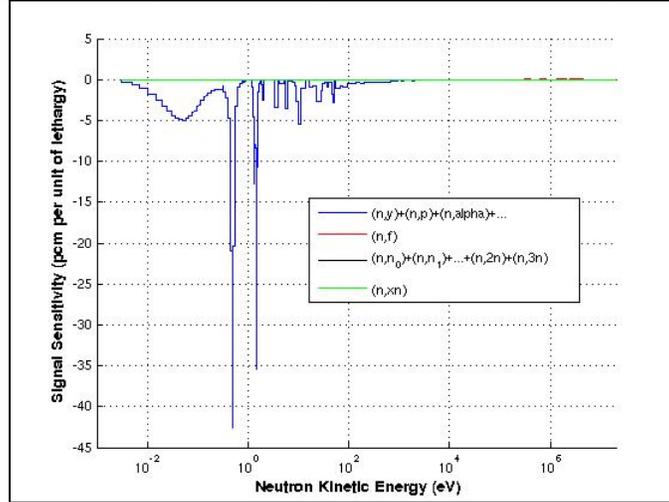
$$\Delta\rho^{TPE} = \frac{\langle \Phi', \Delta H \Phi' \rangle}{I_f} = \frac{\langle \Phi', (\Delta A - \lambda \Delta F) \Phi' \rangle}{I_f}$$

- $\Phi', \Phi^+$  respectively the perturbed direct and nominal adjoint fluxes,
- $H, A, F$  respectively the Boltzmann operator (and its associated eigenvalue  $\lambda$ ), the advection operator and the fission operator,
- $I_f$  the fission integral.

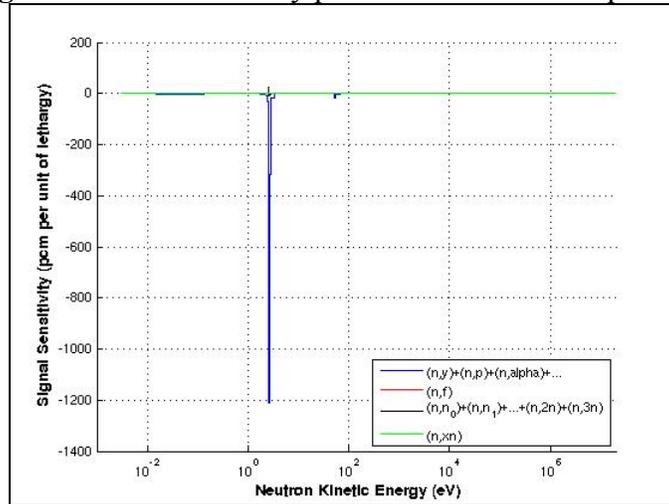
As shown in Figures 3 and 4, the sensitive energy range of  $^{237}\text{Np}$  is the thermal and epithermal range whereas the sensitive energy range of  $^{242}\text{Pu}$  focuses on the first  $E_0=2.7\text{eV}$  resonance.

Table 2 points out the neutron balance breakdown of each sample poisoning worth on isotopes, reactions and associated energy ranges.

**Figure 3:**  $^{237}\text{Np}$  sensitivity profile in OSMOSE experiment



**Figure 4:**  $^{242}\text{Pu}$  sensitivity profile in OSMOSE experiment



**Table 2:** Breakdown of the isotopic poisoning worth

Sample	Main Neutron induced reactions involved	Thermal range (<0.25eV)	Epithermal and fast range (>0.25eV)
$^{234}\text{U}$ (0.3g)	$(n, \gamma)$	40%	60% ( $E_0=5.2\text{eV}$ )
Reprocessed Uranium	no particular isotope	depend	depend
$^{237}\text{Np}$ (0.1g)	$(n, \gamma)$	40%	60% (30% for $E_0=0.55\text{eV}$ )
$^{237}\text{Np}$ (0.6g)	$(n, \gamma)$	40%	60% (30% for $E_0=0.55\text{eV}$ )
$^{239}\text{Pu}$ (0.6g)	$(n, f)$ & $(n, \gamma)$	depend	depend
$^{242}\text{Pu}$ (0.5g)	$(n, \gamma)$	10%	90% ( $E_0=2.7\text{eV}$ )

One can deduce the following trends on nuclear data in the resolved resonance range:

- the  $^{234}\text{U}$  neutron capture cross-section is underestimated in JEFF-3.1 evaluation; therefore the thermal cross-section and the large resonance  $E_0=5.16\text{eV}$  should be increased by 5%
- the calculation of Reprocessed Uranium is well predicted using both JEF2 and JEFF3 libraries; this result support the JEFF evaluation of the  $E_0=5.45\text{eV}$   $^{236}\text{U}$  resonance
- $^{237}\text{Np}$ : a clear underestimation of the thermal and resonant capture cross-section is observed with the two independent samples:  $-6\%\pm 2\%$  using JEF-2.2 and  $-11\%\pm 2\%$  using JEFF-3.1. This trend is consistent with recent thermal range measurements [9] performed at the ILL-MINI-INCA facility.
- $^{239}\text{Pu}$  worth is validated within 1.8% accuracy, in particular  $(\nu\sigma_f)-(\sigma_\gamma+\sigma_f) = \sigma_a (\eta-1)$
- $^{242}\text{Pu}(n,\gamma)$  is satisfactory within 2% accuracy, in particular the  $E_0=2.7\text{eV}$  resonance radiation width.

All these trends are consistent with Post Irradiated Experiment of LWR fuel pins analysis [10] and recent activation measurements [11].

## 5. Conclusion

This paper has presented the interpretation of the LWR-UO<sub>2</sub> phase of the OSMOSE experimental program.

The comparison between APOLLO2 deterministic calculations and experiments illustrates the reliability of the latest JEFF-3.1 European nuclear data file for oscillated actinides, except  $^{237}\text{Np}$ , which evaluation improvement is in progress.

(C/E-1) $\pm(\delta E/E)$  values, using JEFF-3.1 nuclear data evaluation, can be summarized as follow:

$$\begin{array}{ll} ^{234}\text{U}: & -5\% \pm 2\% & ^{237}\text{Np}: & -11\% \pm 2\% \\ ^{239}\text{Pu}: & +1\% \pm 2\% & ^{242}\text{Pu}: & +2\% \pm 2\% \end{array}$$

The  $^{234}\text{U}(n,\gamma)$  evaluation was more consistent with OSMOSE integral information in JEF-2.2 than in JEFF-3.1. A clear underestimation on  $^{237}\text{Np}$  capture cross-section is observed in JEFF-3.1 evaluation.  $^{239}\text{Pu}$  and  $^{242}\text{Pu}$  evaluations are satisfactory in both files.

In the next future, further measurements in the OSMOSE program will investigate the  $^{232}\text{Th}$ ,  $^{233}\text{U}$ ,  $^{238,240,241}\text{Pu}$ ,  $^{241,243}\text{Am}$ , Cm neutron induced cross-sections. In order to distinguish the thermal, resolved and unresolved range of the actinide cross-sections, all samples will be oscillated in various spectra from over-moderated spectrum (R2-UO<sub>2</sub> Phase) up to epithermal spectrum R1-MOX Phase) and intermediate spectrum (MORGANE Phase).

## Acknowledgements

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