

# RESONANCE EVALUATION OF $^{239}\text{Pu}$ IN THE RESONANCE REGION UP TO 4 KEV

Luiz Leal<sup>1</sup>, Nicolas Leclaire<sup>1</sup>, and Isabelle Duhamel<sup>1</sup>

<sup>1</sup> *Institut de Radioprotection et de Sûreté Nucléaire (IRSN), Fontenay-aux-Roses, 92262, France*

luiz.leal@irsn.fr, nicolas.leclaire@irsn.fr, isabelle.duhamel@irsn.fr

## ABSTRACT

Resolved resonance evaluation of the  $^{239}\text{Pu}$  cross section in the energy range up to 4 keV has been carried out with the SAMMY computer code. Existing resolved resonance evaluation such as ENDF/B-VIII.0 and JENDL4 data libraries for  $^{239}\text{Pu}$  is limited to 2.5 keV whereas above this energy the unresolved resonance methodology is used. High resolution transmission and fission data taken at the Oak Ridge Electron Linear Accelerator (ORELA) permitted extending the resonance region up to 4 keV. The thermal and average fission cross section values calculated with the resonance parameters derived in the evaluation fall close to that indicated in the suggested IAEA fission standards.

KEYWORDS: resonance, cross-sections, criticality, covariance

## 1. INTRODUCTION

An R-matrix resonance evaluation of the  $^{239}\text{Pu}$  cross section was carried out at the Institut de Radioprotection et de Sûreté Nucléaire (IRSN) in support of criticality safety applications. The reduced R-matrix formalism, also known as the Reich-Moore methodology, of the SAMMY code together with several experimental data was used in the evaluation. Recent capture data measurement performed at the Los Alamos Neutron Science Center (LANSCE) with the Detector for Advanced Neutron Capture Measurements (DANCE) was included in the evaluation. Transmission data from ORELA measured at the nitrogen liquid temperature and flight path of 80 meters and fission cross section measured at a flight path of 86 meters helped extending the energy resonance region up to 4 keV. Issues with the unresolved resonance representation of the  $^{239}\text{Pu}$  cross sections above 2.5 keV motivated the study of the feasibility of applying a resolved resonance representation from 2.5 keV to 4 keV.

## 2. $^{239}\text{Pu}$ RESONANCE EVALUATION IN THE ENERGY RANGE OF $10^{-5}$ EV TO 4 KEV

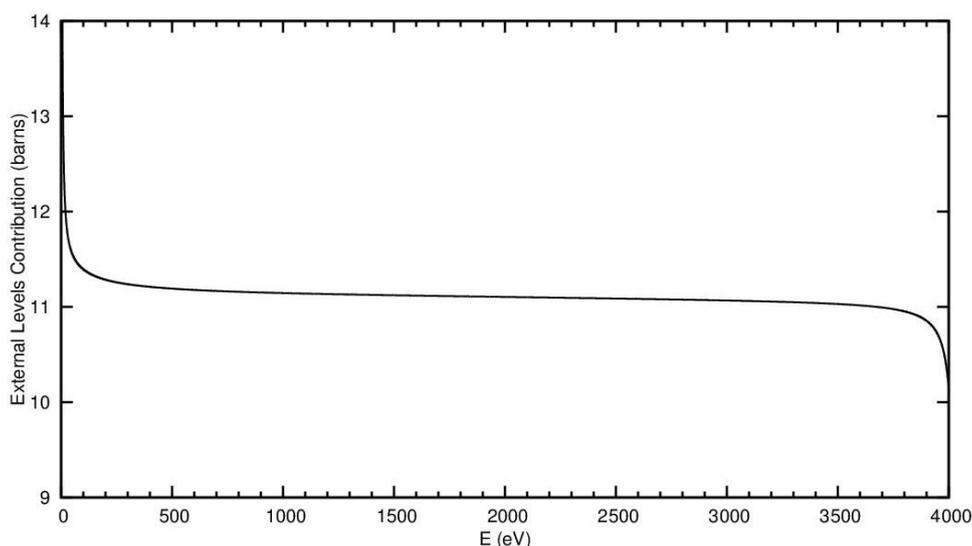
A resolved resonance range (RR) evaluation of the  $^{239}\text{Pu}$  cross section was carried using the multilevel SAMMY R-matrix code.[1] The resonance evaluation in most of the existing nuclear data library is limited to the energy range from thermal to 2.5 keV with an unresolved resonance range (URR) representation above 2.5 keV. An apparent issue with reproducing the average cross section in the URR exists in the energy range 2.2 keV to 4 keV. Hence, a study of the feasibility of applying a resolved resonance representation from 2.5 keV to 4 keV was carried out. The URR representation of  $^{239}\text{Pu}$  in existing data library above 2.5 keV is founded solely on statistical sampling on the basis of the average resonance parameters. While the URR evaluation were carefully done cluster of resonances in the energy range 2.5 keV to 4 keV precluded reproducing the average cross section based solely on the average resonance parameters.

## 2.1. External Energy Levels Determination

The long-range interferences in the R-matrix formalism play a major role in fitting fissile isotopes. Interference in the scattering and fission channels may lead to dubious interpretation of the resonance parameters. For fissile isotopes the latter is very pronounced while the former plays an important role as well. In the present evaluation the first step consisted of finding pseudo resonances below  $10^{-5}$  eV, also known as the energy bound levels, and above 4 keV which are the resonances that mock up the resonance above 4 keV. Five negative resonances and three energy levels above 4 keV were found that describe well the interference effect in the energy range 0 to 4 keV. The eight external energy levels are listed in Table I in which for each the resonance energy  $E_r$ , gamma width  $\Gamma_\gamma$ , neutron width  $\Gamma_n$ , two fission widths  $\Gamma_{f1}$  and  $\Gamma_{f2}$  and the spin and parity  $J^\pi$  are shown. Negative signs associated with the fission partial widths  $\Gamma_{f1}$  and  $\Gamma_{f2}$  reflect the sign of the reduced amplitude width  $\gamma_{f1}$  and  $\gamma_{f2}$ . It follows a convention established in the ENDF resonance parameters representation. The last column in Table I lists the resonance total angular momentum and parity. The ground state spin of the  $^{239}\text{Pu}$  is  $0^+$  which leads, for s-wave ( $l=0$ ), to two resonance total angular momentum, namely  $0^+$  and  $1^+$ . Penetrability for higher angular momentum ( $l>0$ ) shows negligible contribution to the cross section below 4 keV. The effect of the external level is shown in Fig. 1 which corresponds to the scattering cross section in the energy range  $10^{-5}$  eV to 4 keV without the presence of resonances in this energy range. In the plateau around the energy of 2 keV the scattering cross-section due to the external energy values is about 11.10 barns corresponding to a scattering radius of 9.40 fm. It is interesting to note that the analysis of high-resolution transmission data led to an effective scattering radius of 9.41 fm. The number of resonances that fitted the experimental data in the energy range 0 to 4 keV is 1572.

**Table I.** Resonance energies and parameters of the external levels.

$E_r$ (eV)	$\Gamma_\gamma$ (meV)	$\Gamma_n$ (meV)	$\Gamma_{f1}$ (meV)	$\Gamma_{f2}$ (meV)	$J^\pi$
<b>Energy bound Levels</b>					
-149.141	47.182	542.357	4226.105	0.0	$1^+$
-8.068	49.725	0.141	-1.499	0.0	$1^+$
-7.019	70.066	17.548	-117.345	223.288	$0^+$
-0.514	24.005	0.118	15.237	1189.353	$0^+$
-0.020	21.029	$6.597 \times 10^{-8}$	-4.880	0.0	$1^+$
<b>Energy levels above 4 keV</b>					
4006.706	39.000	19.901	48.847	0.0	$1^+$
4022.478	39.000	$4.963 \times 10^{-6}$	835.807	121.703	$0^+$
4035.401	39.000	2837.183	-181.877	0.0	$1^+$



**Figure. 1. External levels contribution on the scattering cross-section in the energy ranges  $10^{-5}$  eV to 4 keV.**

## 2.2. $^{239}\text{Pu}$ Resonance Parameter Evaluation

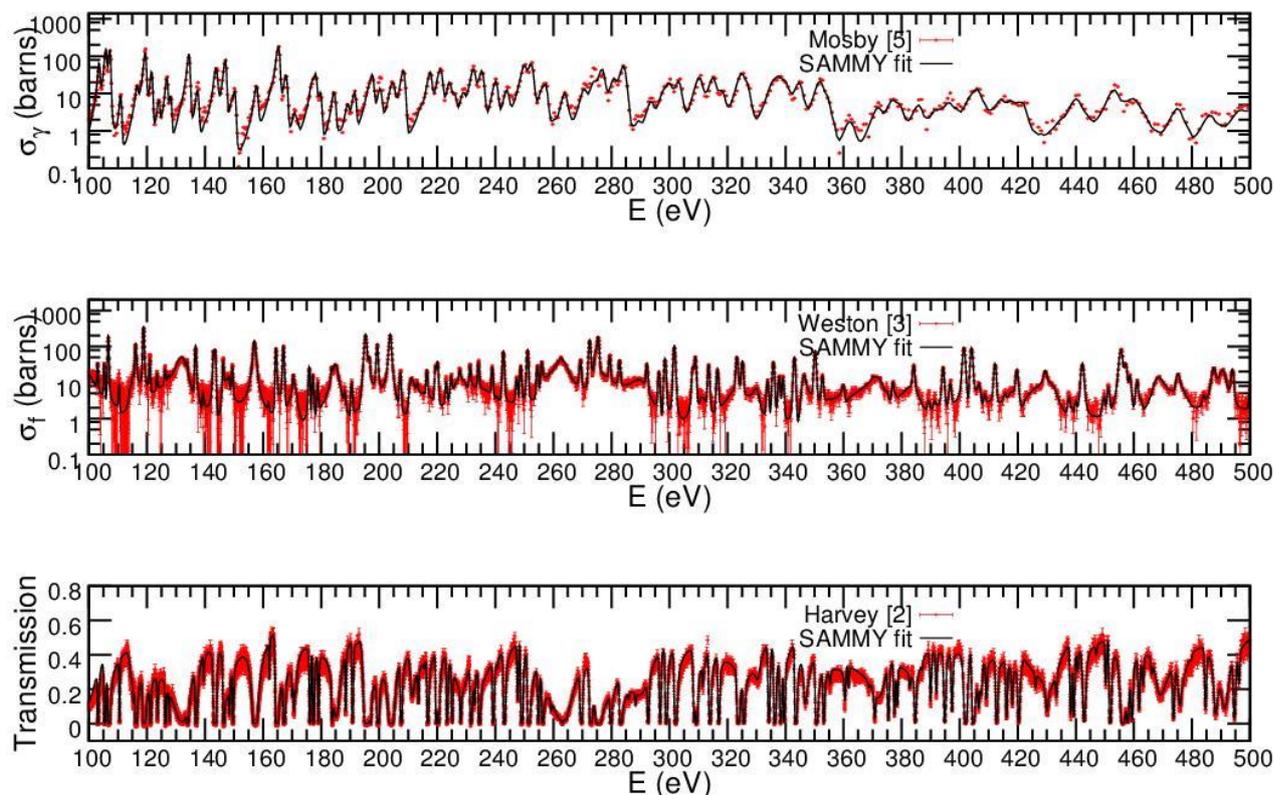
The experimental database used in the present evaluation is listed in Table II. The two sets of data that permitted extending the energy region up to 4 keV were the high-resolution transmission data of Harvey et al.[2] and the fission data of Weston et al.[3] Note that the only experimental total cross section data in the low energy range is that of Bollinger et al.[4] It should be noted that the most recent capture data of Mosby et al.[5] is listed in Table II and that there are no experimental capture data in the energy region above 1 keV suitable for resonance region analysis and evaluation. Sequential analysis of this data with the SAMMY code were performed until a reasonable representation of the data was achieved. The data of Gwin et al. indicated in Table II correspond to two sets of simultaneous capture and fission measurements [6,7] and one set of fission measurement.[8] These data covers the thermal energy region (0.0253 eV). Harvey et al. transmission data (total cross section) with different thicknesses were of paramount importance in the determination of effective scattering radius.

## 2.3. Results of the Fitting of the Experimental Data

The analysis and evaluation of the experimental data displayed in Table II were done up to 4 keV. As usual in the evaluation process, the first step consisted of verifying the consistency on the experimental data. Since the energy range of the experimental data is diverse, the SAMMY fitting of the data was performed by fitting the experimental data with common energy ranges. For instance, in the energy region 100 eV to 1000 eV the transmission data of Harvey (0.07471 atoms/barn) [2], the Weston fission data [3], and Mosby capture data [5] were used most of the time. It should be noted that aside from the Mosby data, there are no other capture data that could, reliably, be used in the energy range above 100 eV. An example of the SAMMY fit of the experimental data of Harvey transmission data [2], Weston fission cross section data [3], and Mosby capture cross section data [5] is displayed in Fig. 2.

**Table II:** Experimental Data.

Transmission (Total Cross Section)		
Data Set	Energy (eV)	Flight-Path (meter)
Bollinger et al.[4]	0.01 – 1.0	Fast Chopper
Harvey (0.00638 atoms/barn) [2]	0.3 – 40.0	18.0
Harvey (0.01803 atoms/barn) [2]	0.3 – 100.0	18.0
Harvey (0.07471 atoms/barn) [2] (77 K)	30.0 – 4000.0	80.4
Fission		
Gwin [6]	0.01 – 4.0	25.6
Gwin [8]	0.01 – 20.0	8.0
Weston [3]	100.0 – 4000.0	86.5
Weston [9]	0.02 – 40.0	18.9
Capture		
Gwin [6]	0.01 – 2.0	25.6
Gwin [7]	0.01 – 100.0	40.0
Mosby [5]	10.0 – 1000.0	25.6



**Figure 2.** SAMMY fit of the Harvey transmission data (bottom), Weston fission cross section data (middle) and Mosby capture cross section data (top).

It should be said that in this energy region the percentage difference between the average fission cross section of Weston [3] and that calculated with the present resonance evaluation is of  $\sim 2\%$ .

The thermal fission, capture and scattering cross sections (0.0253 eV) are displayed in Table III. Shown also in Table III are three values corresponding to ENDF/B-VIII.0 and the values listed at the Atlas of Neutron Resonances (ANR).[10] The standard fission recommended cross section is  $752.371 \pm 2.182$  barns.[11] The uncertainty in the results of this work were calculated based on data covariance generated along with the evaluation.

**Table III:** Thermal cross section values (in barns) calculated with SAMMY.

Quantity	ANR	ENDF/B-VIII.0	This work
$\sigma_\gamma$	$269.1 \pm 2.9$	270.6	$271.4 \pm 3.1$
$\sigma_f$	$748.1 \pm 2.0$	747.7	$750.3 \pm 2.8$
$\sigma_s$	$7.94 \pm 0.36$	8.0	$7.8 \pm 0.26$

### 3. BENCHMARK RESULTS (TEX EXPERIMENTS)

Benchmark calculations using the resolved resonance evaluation presented in this work were carried out to understand the impact of the new evaluation in benchmark calculations. The  $^{239}\text{Pu}$  ENDF/B-VIII.0 evaluation was used as a template. The only change to ENDF/B-VIII.0 is that the new resonance evaluation replaced that of the ENDF/B-VIII.0 in the energy region from  $10^{-5}$  eV to 4 keV. The benchmark calculations consisted of configurations of Zero Power Physics Reactor (ZPPR) Plutonium-Aluminum No-Nickel (PANN) plates covering five different fission energy regimes, with varying fractions of thermal, intermediate, and fast fissions. These correspond to experiments recently conducted within the Thermal/Epithermal eXperiments (TEX) program under the auspices of the Nuclear Criticality Safety Program. Five TEX baselines experiments (Experiments 1-5), meaning with no diluent, were designed.[12] For all five experiments, the ZPPR plates were arranged on the universal critical assembly machine, Planet, in layers of 24 plates (6 plates by 4 plates), resulting in approximately a 30 cm by 30 cm footprint. Multiple layers (0 to 17) were stacked together with varying thicknesses (0 to 1 inch) of interspersed polyethylene placed between the layers to tune the neutron spectrum of the assembly. The experiments were completed in multiple campaigns over 2017 and 2018 at the National Critical Experiments Research Center (NCERC) at the Nevada National Security Site (NNSS) and will be made available in the ICSBEP Handbook in 2020. [13]

The new evaluation of  $^{239}\text{Pu}$  cross sections was tested on these experiments. The  $k_{eff}$  results are reported in Table II—V. The experimental benchmark  $k_{eff}$  is listed in column four and uncertainties given in column five. The energy average lethargy of neutrons causing fission (EALF) is listed in column six. The following column lists the results of calculations using ENDF/B-VIII.0. The last column corresponds to the results using the  $^{239}\text{Pu}$  resonance evaluation developed in this work. Table V suggests that the  $k_{eff}$  results for EALF less than 4 keV have been improved in comparison to ENDF/B-VIII.0 results. This also indicates that extending the resonance range up to 4 keV, into the unresolved energy range, improves the benchmark  $k_{eff}$  estimation. All the benchmark calculations were done with MCNP with 10 pcm standard deviation that is  $\pm 0.0001$ .

**Table IV:** Benchmark  $k_{eff}$  results for TEX experiments.

Case	Thickness of CH <sub>2</sub> (inches)	Number of CH <sub>2</sub> moderating layers	Benchmark $k_{eff}$	Uncertainty	EALF (MeV)	$k_{eff}$	
						ENDF/B-VIII.0	This work
1	0	0	0.99991	0.00256	7.59E-02	1.00319	1.00441
2	1/16	17	1.00078	0.00228	5.37E-03	1.00075	1.00151
3	3/16	12	1.00081	0.00212	2.45E-04	1.01137	1.00708
4	7/16	8	1.00112	0.00266	3.35E-05	1.00352	0.99916
5	1	6	1.00006	0.00178	2.08E-06	1.00626	1.00211

### 4. CONCLUSIONS

Reassessment and evaluation of the  $^{239}\text{Pu}$  resonance parameters were done at IRSN with the SAMMY code in the energy range 0 to 4 keV. High-resolution transmission data were important in the resonance determination in the energy above 2.5 keV till 4 keV. Issues with the unresolved resonance representation of the  $^{239}\text{Pu}$  cross section motivated the evaluation. New capture measurements done at the LANCE were included in the evaluation. Benchmark calculations were done based on the recent TEX experiments and the results indicate that the new evaluation provides an improvement in the benchmark calculation, mainly for case 3 and 5. However, further investigation in the evaluation above 4 keV is needed.

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