^{239}Pu Resonance Evaluation for Thermal Benchamark System Calculations

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Analyses of thermal plutonium solution critical benchmark systems have indicated a deficiency in the ^{239}Pu resonance evaluation. To investigate possible solutions to this issue, the Organisation for Economic Co-operation and Development (OECD) Nuclear Energy Agency (NEA) Working Party for Evaluation Cooperation (WPEC) established Subgroup 34 to focus on the reevaluation of the ^{239}Pu resolved resonance parameters. In addition, the impacts of the prompt neutron multiplication (nubar) and the prompt neutron fission spectrum (PFNS) have been investigated. The objective of this paper is to present the results of the ^{239}Pu resolved resonance evaluation effort.

I. INTRODUCTION

In the 1980s and early 1990s, Derrien et al. [1] performed a ^{239}Pu evaluation in a collaborative work, including CEA and ORNL. At that time, due to computer limitations for data storage and processing, a decision was made to split the resonance region in three parts, namely, 10^{-5} eV to 1 keV, 1 keV to 2 keV, and 2 keV to 2.5 keV. The evaluation was accepted for inclusion in the ENDF and JEFF nuclear data libraries and is still included in the latest releases of ENDF, the ENDF/B-VII.1, and the JEFF-3.1 libraries. While the evaluation was performed based on high-resolution data, mainly transmission data [2] measurements taken at the Oak Ridge Electron Linear Accelerator (ORELA) at ORNL, no benchmark testing was done at the time the evaluation was released. Later, benchmark calculations indicated deficiencies in the ^{239}Pu evaluation in reproducing integral results. Additional issues with the previous evaluation can be attributed to the use of three distinct sets of resonance parameters. Specifically, the cross sections calculated at the energy boundary of two consecutive, disjoint resonance parameter sets could be different, leading to a discontinuity. Another concern relates to data uncertainty assessments using resonance parameter covariance data. For data uncertainty analyses, the use of a single resonance parameter set covering the entire energy region is preferable because the disjoint set of resonance parameters does not permit the determination of uncertainty correlations in the entire energy region. Hence, the decision was made to combine the three sets of resonance parameters and redo the evaluation. The task of generating a single resolved resonance region was achieved because computer resources have improved substantially since the previous ^{239}Pu evaluation effort. As a result, a resonance parameter evaluation was completed at ORNL in 2008 by Derrien, and this ^{239}Pu evaluation covers the energy range 10^{-5} eV to 2.5 keV [3]; however, the evaluation was unable to improve benchmark results and was not proposed for inclusion in either the ENDF or JEFF project. At about the same time as the work was being performed at ORNL, Bernard et al. [4] at CEA/Cadarache performed a reevaluation of the ^{239}Pu resonance parameters and nubar. Since the resonance evaluation for the whole energy region was not available, the work performed by Bernard was based on the JEFF-3.1 evaluation (i.e., with the three disjoint sets of resonance parameters). Bernard's ^{239}Pu evaluation improved the results of benchmark calculations; however, the evaluation did not provide resonance parameter covariance data. By building upon the previous ^{239}Pu evaluation work efforts, the WPEC Sub group 34 (SG34) entitled "Coordinated evaluation of ^{239}Pu in the resonance region" has been able to produce a new ^{239}Pu evaluation that provides improved benchmark performance for thermal plutonium solution systems.

II. ORNL/CEA NEW ^{239}PU EVALUATION IN THE ENERGY RANGE 10^{-5} EV TO 2.5 KEV

A. Resonance Parameter Evaluation Procedure

With the set of resonance parameters covering the energy region up to 2.5 keV a good set of external resonance parameters were determined. The technique used for deriving the external levels is that described in refer-

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ence [5]. Six resonance levels with negative energies and nine levels above 2.5 keV were enough to represent the external resonances interference effect in the energy region 10^{-5} eV to 2.5 keV. The first negative level (close to zero) has a neutron width very small. It does not contribute much to the interference effect in the resonance region. However, it is used to get a representation in the shape of eta (η) that bends down at very low energy. [6] Figure 1 shows the cross section shape in the resonance region due only to the external energy resonance levels. It should be noted that the cross section value converges to 11.13 barns, which represents the potential cross section for ^{239}Pu determined with an effective scattering radius of 9.41 fm. This feature indicates that the external levels contribution to the cross section in the energy range 10^{-5} eV to 2.5 keV is appropriate.

The experimental database used in the new evaluation is essentially the same as that listed in Reference [3]. However, information derived from the knowledge of benchmark calculation results was also included in the SAMMY analysis together with the fitting of the differential data. Two quantities were essential in determining the best set of resonance parameters that fitted the experimental differential data and improving the benchmark results. The two quantities are η and the effective K1. These quantities are defined as:

a) Eta (η)

$$\eta = \frac{\bar{\nu}\sigma_f}{\sigma_a} = \frac{\bar{\nu}}{1+\alpha} \tag{1}$$

where α

$$\alpha = \frac{\sigma_\gamma}{\sigma_f} \tag{2}$$

a) K1

$$K1 = \bar{\nu}\sigma_{0f}g_f - \sigma_{0a}g_a \tag{3}$$

The cross-sections σ_{0f} and σ_{0a} are, respectively, the fission and absorption cross-sections at the thermal energy (0.0253 eV), whereas g_f and g_a are the Westcotts g-factors. The value used in calculating the effective K1 from Eq. 3 is taken at thermal. It was noted that the benchmark results were very sensitive to η and K1. The benchmark results indicated that in some cases the sensitivity to K1 was more significant than on η. The K1 value for ^{239}Pu is higher than that of other major isotopes. For instance, for ^{235}U K1 value is around 722 barns whereas for ^{239}Pu it is 1160 barns. An example of the SAMMY fit of the experimental differential data is displayed in Fig.2 for the total cross-section of Bollinger, [7] fission cross-section of Wagemans, [8] and capture cross-section of Gwin [9] in the energy region from 0.01 eV to 3 eV.

Values of the cross-section at thermal (0.0253 eV), fission Westcott factors, thermal $\bar{\nu}$, resonance integrals and K1 value are shown in Table 1. The unit for cross sections, K1 and resonance integral is barns whereas the Westcott factor is dimensionless. Also, shown in Table 1 are the values listed in the Atlas of Neutron Resonance (ANR), [10] ENDF/B-VII.1, and the values calculated using Bernard's evaluation that is included in JEFF3.1.1. The thermal cross-section values listed in the ANR were used in the SAMMY evaluation. The values listed in Table I) were calculated with the SAMMY code.

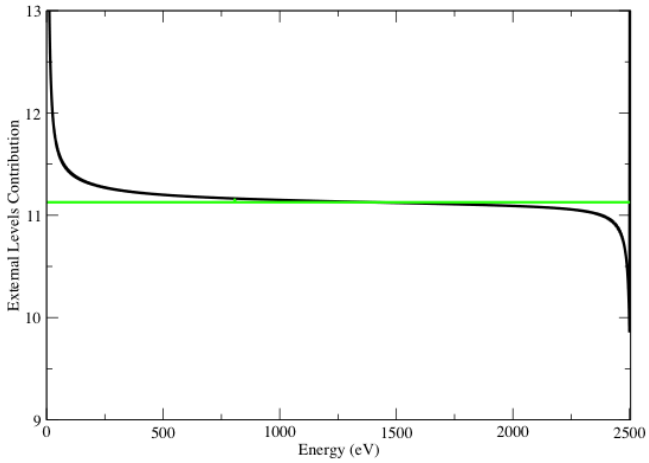

FIG. 1. Contribution of the External levels in the resonance region.

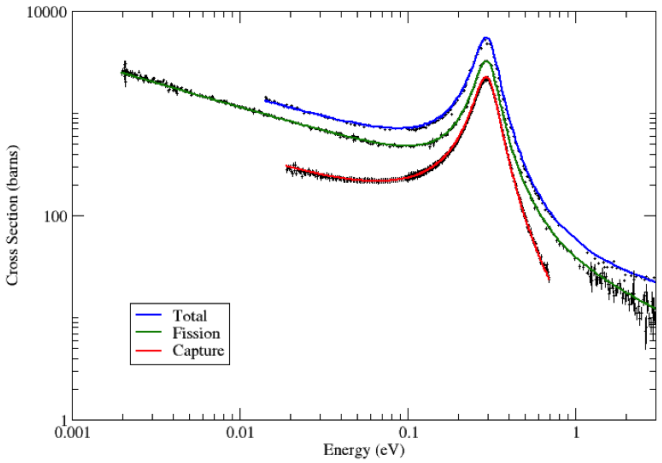

FIG. 2. Results of SAMMY fit of the total, fission and capture cross-section data.

TABLE I. Thermal values and integral quantities calculated with SAMMY.

Quantity	ANR	ENDF/B-VII.1	JEFF3.1.1	ORNL/CEA
σ_γ	269.3±2.9	270.64	272.72	270.06
σ_f	748.1 ± 2.0	747.65	747.08	747.19
g_f	1.0553 ± 0.0013	1.0544	1.0495	1.0516
g_a	1.0770 ± 0.0030	1.0784	1.0750	1.0771
$\bar{\nu}$	2.879 ± 0.006	2.873	2.873	2.873
I_γ	180 ± 20	181.44	181.50	180.09
I_f	303 ±10	302.60	303.58	309.09
$K1$	1177.25	1166.62	1156.35	1161.30

B. Benchmark Calculations

To verify the performance of the ^{239}Pu evaluation in benchmark calculations seven critical experiments were chosen from the International Criticality Safety Benchmark Evaluation Project (ICSBEP) in the International Handbook of Evaluated Criticality Safety Benchmark Experiments. [11] These benchmark experiments consist of light water reflected spheres of plutonium nitrate solutions. The benchmarks, listed in Table II), have the average of neutron lethargy causing fission (EALF) spanning the energy range of 0.04 eV to 3 eV. It should be noted that the uncertainty in these benchmarks are around 500 pcm.

Several resonance parameters were derived from the SAMMY fitting of the experimental differential data. Each time a resonance parameter was obtained with a satisfactory fitting of the differential data (a good χ^2) the SAMMY resonance parameter was converted in the ENDF format (MT2 MF151) and inserted into the JEFF3.1.1 by replacing the existing resonance parameter. The cross section library created was then processed for use in Monte Carlo calculation using the MCNP code. The MCNP libraries were generated with the NJOY/ACE code. All the cross section data for the remaining isotopes present in the benchmark experiments were taken from the ENDF/B-VII.0. The process from the SAMMY fitting of the experimental data to the MCNP calculation was automated, validated and tested. Various k_{eff} results were obtained for the seven benchmark listed in Table II). The impact of the cross section change in the k_{eff} values were analyzed and it was noted that a very minor change in the thermal cross section and in the first resonance around 0.2956 eV would significantly change the k_{eff} value of the thermal benchmark listed in Table II). In addition, results of sensitivity calculations using the TSUNAMI sequence of the SCALE code [12] indicated that to achieve a reasonable k_{eff} result a combined change on the nubar, on the fission and capture cross sections values were needed as opposed to a simple change in one of these quantities alone. The very first attempt made was to focus on η (or α) since it involves these three quantities as indicated in Eq. 1. However, further investigations indicated that the k_{eff} was also very sensitive to K1. No experimental measurement of K1 was found in the literature for ^{239}Pu. Nevertheless, integral experiments performed at the CEA/Cadarache MINERVE facility could be used to infer the value of K1 that provided the best results for reactivity changes. A K1 value of around 1161 barns indicated that a reasonable k_{eff} could be achieved for the seven benchmark listed in Table II. Hence, in addition to fitting the experimental differential data SAMMY also fitted K1. The benchmark results for the seven benchmark displayed in Table II are shown in Fig. 3. Despite the encoraging results presented in Table II more benchmark tests are needed. In fact, WPEC SG34 made available a ^{239}Pu library for further test.

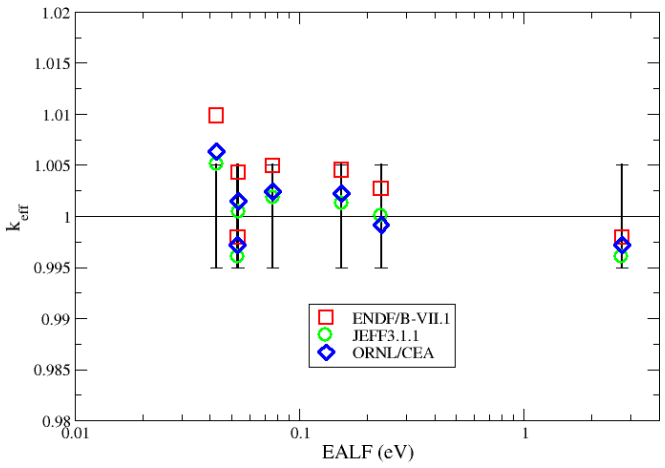

FIG. 3. MCNP results for seven ICSBE benchmarks listed in Table II.

III. CONCLUSIONS

The Reich-Moore reevaluation of the ^{239}Pu resonance parameters up to 2.5 keV reproduces the experimental cross section data very well. Integal quantities, mainly the K1 value was included in the resonance fitting with the SAMMY code. The ^{239}Pu benchmark results showed

TABLE II. ICSBEP ^{239}Pu thermal benchmark.

Benchmark	Experimental k_{eff}	EALF (eV)	Contents
PST12.13	1.0000±0.0050	0.0428	19.5 % ^{240}Pu
PST4.1	1.0000±0.0047	0.0531	0.5 % ^{240}Pu
PST12.10	1.0000±0.0033	0.0535	25 % ^{240}Pu
PST18.6	1.0000±0.0047	0.0761	43 % ^{240}Pu
PST1.4	1.0000±0.0047	0.154	5 % ^{240}Pu
PST34.4	1.0000±0.0047	0.231	116g Pu/L, 1.42 Gd/L
PST34.15	1.0000±0.0047	2.730	363g Pu/L, 20.25 Gd/L

a much larger dependence on K1 than η. In addition to the resonance evaluation, the SG34 also coordinates the evaluation of other quantities such as the prompt neutron fission spectrum and $\bar{\nu}$ which also impacts the benchmark results.

IV. ACKNOWLEDGMENTS

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