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^{239}Pu neutron resonance parameters revisited and covariance matrix in the neutron energy range from thermal to 2.5 keV

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Abstract. To obtain the resonance parameters in a single energy range up to 2.5 keV neutron energy and the corresponding covariance matrix, a reevaluation of ^{239}Pu was performed with the analysis code SAMMY. The most recent experimental data were analyzed in the energy range thermal to 2.5 keV. The experimental data were renormalized, aligned on a common energy scale, and corrected for residual background. Average neutron transmission and cross sections calculated with the new resonance parameters were compared to the corresponding experimental data and to ENDF/B-VI.

1 Introduction

The last evaluation of ^{239}Pu resonance parameters [1–4] was released in 1993 and was adopted in ENDF/B-VI, JEFF, and JENDL. The evaluation was performed in three uncorrelated energy ranges: 0 to 1 keV, 1 to 2 keV, and 2 to 2.5 keV. Because of the difficulties of handling large covariance matrices and of inadequate formats in the processing codes, the resonance parameter covariance file was not given at this time. Recently [5], advancements made at Oak Ridge National Laboratory (ORNL) have allowed the treatment of large covariance matrices by using faster and larger computers with the improved version of SAMMY [6]. The evaluation performed at ORNL between 1985 and 1990 [1–3] relied, for the fission cross section, on the 1984 measurement of Weston et al. [7] which was, on average 3% lower than the ENDF/B-VI standard cross sections [8] in the energy range 0.1 keV to 1.0 keV. New measurements were performed by Weston in 1991 [9] at ORNL and by Wagemans in 1993 [10] at Geel Linear Accelerator (GELINA), confirming the ENDF/B-VI standard values. The resonance parameters released in 1993 were slightly modified to take into account the need of a renormalization of earlier fission data, but a complete analysis of the new fission data was not undertaken. In the present work, the SAMMY analysis of an updated experimental data base is performed. A single set of resonance parameters and a complete resonance parameter covariance matrix is obtained in the neutron energy range thermal to 2.5 keV.

2 The experimental data base

Most of the experimental data used in the present evaluation were taken at the Oak Ridge Electron Linear Accelerator (ORELA) between 1971 and 1993 by Gwin et al. [11–13] for the fission and capture cross sections, by Weston et al. [7,9,14] for the fission cross sections and by Harvey et al. [15] for the neutron transmissions. These data cover the energy range 0.01 eV to 2.5 keV. The high resolution of Harvey transmission data [15] and of Weston 1988 [14] fission data allowed the resonance analysis to be performed in the energy range up to 2.5 keV. The fission cross section measurements performed by Wagemans et al. [16,17] at GELINA were used for normalization purposes. The only total cross sections available for the analysis in the thermal energy range were those from the measurement performed by Bollinger in 1956 [18]. The ORELA transmission data of Spencer et al. [19], which were used in earlier evaluations, were not available for the present evaluation.

The fission measurements, which include the thermal energy range, can be normalized on the standard cross sections at the neutron energy of 0.0253 eV. That is the case of Gwin 1971, 1976, and 1984 data; Wagemans 1988 and 1993 data; and Weston 1993 data. The other measurements, Weston 1984 and Weston 1988, should be normalized in an energy range overlapping with a measurement normalized at 0.0253 eV. The normalization performed by Weston in 1984 and 1988 gave average fission cross sections 3% to 4% smaller than the values recommended by the ENDF/B-VI standard committee in the energy range 100 to 1000 eV. The purpose of the fission measurement performed by Weston in 1993 was to accurately determine the shape of the fission cross section down to 0.0253 eV in order to obtain a more accurate normalization for previously reported ORELA fission cross section measurements. The results indicate that 3% higher normalization should have been used for the 1984 and 1988 data. At the same time, new measurements were also performed at GELINA by Wagemans et al. [17] to address the ORELA data discrepancy. Two different experiments were performed using different techniques. Wagemans obtained the value of 9277 b.eV for the fission integral in the energy range 100 eV to 1000 eV, which confirmed the renormalized results of Weston [9], at only 0.6% below the ENDF/B-VI standard.

After renormalization, adjustment of the neutron energy on the same energy scale, and some residual background corrections, a consistent experimental data base was obtained suitable for a sequential SAMMY analysis in the energy range 0.01 eV to 2.5 keV. The experimental parameters (Doppler and resolution broadening parameters, etc.) were taken from the earlier evaluation.

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Table 1. ^{239}Pu cross section at 0.0253 eV.

	Standard 1992	B-VI 1993	Mughabghab 2005	Present 2007
Total (b)	(1027.30)	1026.30	1025.3 ± 2.9	1027.30
Fission (b)	747.99 ± 0.25	746.66	748.1 ± 2.0	747.09
Capture (b)	271.43 ± 0.79	270.65	269.3 ± 2.9	271.40
Scattering (b)	7.88 ± 12.30	7.99	7.94 ± 0.4	8.81

3 Results of the analysis

The three independent sets of resonance parameters of ENDF/B-VI were merged in a single set by suppressing the external resonances in the inner boundaries of the sets, keeping only the external negative resonances and the external resonances above 2.5 keV. Because the three sets were obtained from uncorrelated evaluations, it was not expected that the single set could fit accurately the experimental data. A new set of external resonance parameters was evaluated from preliminary SAMMY fits of Harvey neutron transmission [15]. The effective scattering radius $R' = 9.41$ fm was kept from the previous evaluation. The final SAMMY analysis was performed in three stages in the energy range 0.01 to 7 eV, 7 to 100 eV, and 0.1 to 2.5 keV, respectively. In each stage the neutron cross sections and transmissions were calculated with the entire set of parameters. The prior values of the first stage were those of the single set deduced from ENDF/B-VI with new external resonance parameters as explained above. The experimental data base contained a total number of 110696 measured values, and the χ^2 value corresponding to the final set of resonance parameters was 1.31 per data point. The new set of resonance parameters contains 1030 s-wave resonances for the representation of the cross sections in the energy range 0 to 2.5 keV.

The cross sections at 0.0253 eV calculated with the resonance parameters are compared to other results in table 1. If needed, small variations of these cross sections can be obtained by small variations of the parameters of the resonance at −4.4 eV; a variation of 0.1% of the capture cross section is obtained by a variation of 0.9% of the capture width, with negligible effect on the fission cross section; a variation of 0.1% of the fission cross section is obtained with a variation of 0.37% of the neutron width, with very small effect on the capture cross section.

An example of SAMMY fit of the fissions cross section is displayed in figure 1. The average fission cross sections calculated with the resonance parameters in selected energy ranges are compared to ENDF/B-VI and to the experimental data in tables 2 and 3. As expected, the differences between the present evaluation and ENDF-VI are small. The new fission resonance integral is 302.53 b compared to the values of 302.56 b in ENDF/B-VI and 303 ± 20 b recommended by Mughabghab [20].

The average capture and absorption cross sections calculated with the resonance parameters are compared to ENDF/B-VI and to the experimental values in tables 4 and 5. In the thermal energy range, up to 0.7 eV, results of the Gwin measurements and the calculated cross sections are in very good agreement. Between 0.7 eV and 100 eV, the Gwin average cross sections are significantly larger, which is likely due to residual backgrounds in the experimental data. (see fig. 2, for example.) In the energy range 0.1 to 1.0 keV, the calculated capture cross section is, on average, 7% smaller than Gwin experimental value. That is mainly due to the difference of 3.4% between the absorption measured by Gwin and the value calculated by the resonance parameters. The calculated capture data agree within 1.3%, on average, with the Schomberg [21] measured values. In the energy range 1 to 2 keV the calculated capture cross section is between Schomberg and Gwin.

The dilute capture resonance integral calculated with the resonance parameters is 179.74 b after adding the contribution

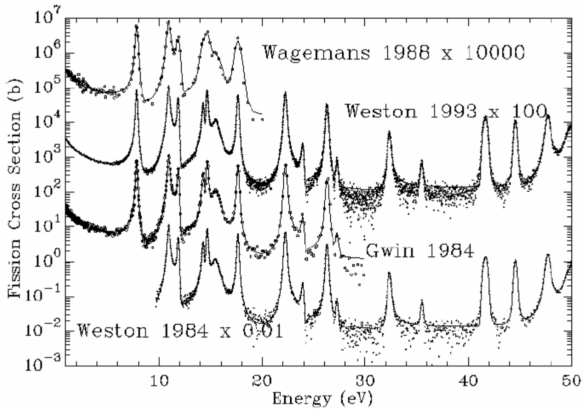

Fig. 1. Experimental ^{239}Pu fission cross section in the energy range 3 to 50 eV. The solid lines represent the cross sections calculated with the resonance parameters.

Table 2. ^{239}Pu average fission cross sections (barn) in the energy range 0.01 to 100.0 eV. The last column shows the percentage deviation between ENDF/B-VI and the present evaluation.

Energy Range (eV)	Weston 1984	Gwin 1984	Wagemans 1988	Weston 1993	B-6 1993	Present 2007	%
0.01–0.02		969.9	970.6	974.0	971.3	971.6	−0.3
0.02–0.03		752.5	754.4	756.0	755.5	755.0	−0.0
0.03–0.04		645.1	652.1	648.5	647.7	647.2	−0.1
0.04–0.05		578.4	584.9	581.7	582.4	582.4	0.0
0.05–0.10		497.8	498.7	497.8	499.3	501.7	+0.5
0.10–0.20		582.8	586.7	588.2	586.1	594.8	+1.5
0.20–0.30		1995	2030	2035	2015	2009	−0.3
0.30–0.40		1684	1708	1695	1689	1693	+0.2
0.40–0.50		349.4	346.1	347.0	348.0	341.0	−2.1
0.50–1.00		76.3	77.9	75.1	77.4	76.2	−1.6
1.00–6.00		12.1	12.1	11.5	11.6	11.7	+0.9
6.00–10.0		46.2	47.8	46.7	46.3	47.9	+3.5
0.01–10.0		79.93	81.31	80.34	80.10	80.73	+0.8
10.0–12.6	188.8	191.8	191.7	190.3	191.5	192.3	+0.4
12.6–20.0	74.0	74.7	74.0	73.7	72.6	73.9	+1.8
21.0–24.7	48.2	46.3		48.0	46.8	47.0	+0.4
24.7–30.0	18.9	18.6		18.7	18.2	18.0	−1.1
30.0–55.0	17.5			17.4	17.7	17.6	+1.1
55.0–80.0	71.9			72.3	72.0	71.0	−1.4
80.0–100	48.8			49.2	48.9	48.6	−0.6
0.01–100				53.9	53.7	53.6	−0.2
10.0–100	50.86			50.99	50.78	50.54	−0.5

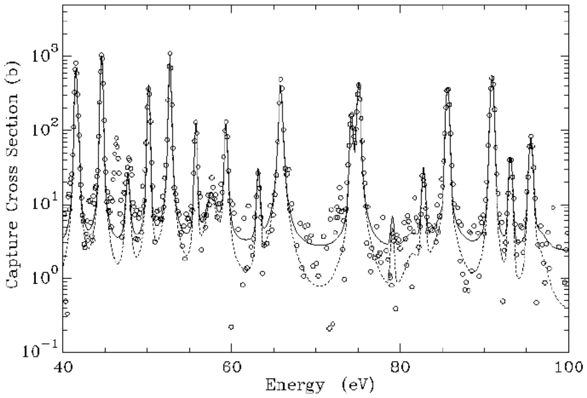

Fig. 2. Gwin 1971 experimental ^{239}Pu capture cross section in the energy range 40 to 100 eV. The dashed line represents the cross section calculated with the resonance parameters. The solid line takes into account a residual background correction.

Table 3. ^{239}Pu average fission cross sections (barn) in the energy range 0.1 to 2.5 keV. The last column shows the percentage deviation between ENDF/B-VI and the present evaluation.

Energy Range (keV)	Weston 1984	Weston 1988	Standard 1990	B-6 1993	Present 2007	%
0.10–0.20	18.50	19.00	18.67	18.69	18.41	−1.5
0.20–0.30	17.82	17.83	17.88	17.80	17.67	−0.7
0.30–0.40	8.313	8.203	8.430	8.301	8.268	−0.4
0.40–0.50	9.546	9.403	9.570	9.580	9.447	−1.4
0.50–0.60	15.51	15.31	15.56	15.40	15.36	−0.3
0.60–0.70	4.285	4.189	4.459	4.358	4.278	−1.9
0.70–0.80	5.507	5.502	5.630	5.509	5.525	+0.3
0.80–0.90	4.872	4.873	4.979	4.842	4.877	+0.7
0.90–1.00	8.361	8.437	8.297	8.436	8.424	−0.1
0.00–1.00	10.30	10.31	10.37	10.32	10.25	−0.7
1.00–2.00	4.419	4.555	4.466	4.432	4.41	−0.3
2.00–2.50	3.01	3.030		2.985	2.98	−0.6

of ENDF/B-VI above 2.5 keV. This is to be compared with the value of 181.54 b for ENDF/B-VI and the value of 180 ± 20 b recommended by Mughabghab [21].

4 The resonance parameter covariance file

The calculation of the resonance parameter covariance matrix was performed by a single SAMMY run in the energy range 0.01 eV to 2.5 keV, including sequentially all the experimental data, with variation of all the resonance parameters. The analysis used the PUP option of SAMMY that allows taking into account systematic errors in the experimental data (the important contributors being the experimental normalization and background). The calculation was time-consuming and needed large computer memories and storage, because of the handling of matrices of about 10^6 elements.

The resonance parameter covariance file is used to calculate the covariance matrix of the group cross sections. The calculation can be performed by SAMMY or by the processing codes PUFF-IV [22] and ERRORJ [23]. Actually, the uncertainties calculated for the group cross sections are still small, due to small uncertainties on the resonance parameters. An option of SAMMY allows to multiply, in chosen energy ranges, the uncertainties on the resonance parameters by a suitable coefficient in order to obtain more realistic uncertainties on the average cross sections. However, this method does not work for small cross sections between resonances, where the systematic uncertainties, mainly due to the uncertainties on the experimental background evaluation, are important compared to the calculated cross sections. An example of calculated group cross sections and uncertainties is given is table 6. In the energy range 1 to 7 eV the calculated uncertainties of 0.01 to 0.05 b are one order of magnitude too small compared to

Table 4. Average capture cross sections (barn) in the energy range 0.01 to 100 eV. The last column shows the percentage deviation between ENDF/BVI and the present evaluation.

Energy Range (eV)	Gwin 1971	Gwin 1976	Gwin EXFOR	ENDF/B-6 1996	Present 2007	%
0.02–0.03	273	282	267	272.9	273.7	+0.3
0.03–0.04	244	246	238	245.1	245.3	0.2
0.04–0.05	231	227	226	230.7	230.5	−0.1
0.05–0.06	226	226	222	223.5	223.0	−0.2
0.06–0.07	223	222	218	221.1	220.2	−0.4
0.07–0.08	226	213	221	222.1	221.0	−0.5
0.08–0.09	230	227	225	226.1	224.7	−0.6
0.09–0.10	236	236	231	232.7	231.1	−0.7
0.10–0.20	350	347	346	347.6	343.4	−1.2
0.20–0.30	1363	1359	1354	1362.6	1342.8	−1.5
0.30–0.40	1152	1139	1089	1090	1105.2	1.4
0.40–0.50	203	204	191	190	192.2	1.2
0.50–0.60	69	68	65	66.4	66.8	0.6
0.60–0.70	33	34	33	33.8	33.9	0.3
0.20–0.70	493.9	491	479.9	482.8	481.1	−0.4
0.70–7.30				3.12	2.91	−7.2
7.30–16.0	88.5 ± 15.			76.4	74.3	−3.0
16.0–37.5	22.0 ± 4.0			20.5	20.7	+1.0
37.5–50.0	62.4 ± 6.0			48.6	49.7	+2.3
50.0–100	37.3 ± 6.0			33.6	34.5	+2.7

Table 5. Average capture cross sections (barn) in the energy range 0.1 keV to 2.5 keV. The last column shows the percentage deviation between ENDF/B-VI and the present evaluation.

Energy Range (eV)	Gwin Abs.	Present Abs.	Shomberg Capt.	Gwin Capt.	B-6 Capt.	Present Capt.	%
0.05–0.10	92.84	92.35		35.88	33.60	34.00	+1.2
0.10–0.20	33.66	33.59	17.81	15.70	15.53	15.18	−2.3
0.20–0.30	34.69	32.70	14.86	16.79	15.74	15.02	−4.8
0.30–0.40	18.31	17.36	9.90	9.83	9.65	9.10	−6.0
0.40–0.50	13.56	13.30	4.29	4.16	3.94	3.85	−2.3
0.50–0.60	26.54	25.95	9.88	11.08	10.83	10.59	−2.3
0.60–0.70	11.57	10.85	6.91	7.02	6.50	6.57	+1.1
0.70–0.80	10.52	10.23	5.16	5.18	4.92	4.71	−4.5
0.80–0.90	9.30	8.56	2.85	4.20	3.63	3.69	+1.3
0.90–1.00	13.23	13.55	4.42	5.40	5.04	5.12	+1.6
0.10–1.00	19.04	18.42	8.31	8.81	8.43	8.20	−2.8
1.00–2.00	8.31	8.00	3.25	3.79	3.74	3.59	−4.2
2.00–2.50		6.13			3.46	3.16	−9.5

Table 6. Example of fission and capture group cross section and uncertainties calculated with the resonance parameter covariance file.

Energy Range (eV)	σ_f	$\Delta\sigma_f$	σ_c	$\Delta\sigma_c$
3000.000–550.000	4.48	0.04	3.74	0.13
550.000–100.000	14.04	0.17	10.68	0.14
100.000–30.000	45.52	0.68	33.99	0.54
30.000–10.000	68.16	0.36	46.22	0.33
10.000–8.100	6.59	0.15	4.83	0.05
8.100–6.000	85.26	0.90	62.51	0.91
6.000–4.750	7.14	0.18	1.24	0.01
4.750–3.000	8.30	0.22	1.33	0.01
3.000–1.770	12.98	0.31	2.26	0.02
1.770–1.000	24.57	0.49	5.34	0.05
1.000–0.625	56.00	0.80	18.52	0.19
0.625–0.400	225.25	2.02	118.91	1.54
0.400–0.375	734.33	7.16	451.80	6.24
0.375–0.350	1162.70	11.62	740.60	10.52
0.350–0.325	1936.90	19.88	1267.60	19.09
0.325–0.275	3070.00	33.60	2058.20	33.73
0.275–0.250	2332.10	25.94	1569.90	24.17
0.250–0.225	1506.90	16.56	1001.70	14.59
0.225–0.200	1020.80	10.66	661.26	9.31
0.200–0.150	679.55	6.42	413.56	5.55
0.150–0.100	504.47	5.14	269.12	3.40
0.100–0.070	484.93	5.75	225.16	2.90
0.070–0.050	525.90	6.41	221.29	3.09
0.050–0.040	582.17	6.96	230.17	3.48
0.040–0.030	647.22	7.54	245.05	3.94
0.030–0.025	717.91	8.19	263.27	4.45
0.025–0.010	908.11	10.13	317.82	5.80
0.010–0.007	1250.20	14.01	421.34	8.21
0.007–0.003	1645.80	18.64	545.65	10.94

the systematic uncertainties of about 0.5 b in the experimental data. Furthermore, because of the interference effects in the fission channels, the calculated cross sections in this energy range depend strongly on the parameters of the bound levels. Differences as large as 0.5 b are found in cross sections calculated with sets of bound levels fitting accurately the cross sections at 0.0253 eV. This region contributes about 4% to the resonance capture integral.

5 Conclusion

The goal of the present work was to create a full resonance parameter covariance matrix for ^{239}Pu in the resolved resonance region 0 to 2.5 keV, to be used in group cross section calculations for various applications. This work took advantage of advances made in SAMMY for the treatment of large covariance matrices and of the creation of new ENDF formats for the use of these matrices in nuclear data processing codes. Because the previous ENDF/B-VI resonance parameter evaluation was performed in three uncorrelated energy ranges not convenient for the creation of a resonance parameter covariance matrix in the entire resolved energy range, the evaluation needed to be revised. For this reevaluation, the experimental data base was updated according to the most recent work on the fission cross section standard. The cross sections calculated with the new set of resonance parameters are, as expected, very similar to those calculated with ENDF/B-VI.

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