Re-evaluation and validation of the ^{241}Pu resonance parameters in the energy range thermal to 20 eV.

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Abstract

This paper presents a new SAMMY analysis of the ^{241}Pu resonance parameters from thermal to 20 eV. This evaluation takes into account the trends given by integral experiments (Post-Irradiation Experiments performed in French PWR). Compared to the previous evaluations performed by Derrien et al. [1] and [3], the capture cross-section increases especially in the 0.26 eV resonance. It is shown that the new resonance parameters proposed in this work improve the prediction of the ^{242}Pu build-up in a PWR which was significantly underestimated with the previous evaluations.

1 Background

In 1988, the ^{241}Pu resonance parameters were evaluated in the neutron energy range from thermal to 300 eV by H. Derrien and G. de Saussure [1]. This evaluation was adopted in ENDF/B-VI and JEF2 libraries. After this work, a new fission cross-section measurement was performed by C. Wagemans et al. in 1991 [2] in order to check the shape of the fission cross-section in the thermal range. This measurement has demonstrated that the shape of the fission cross-section was compatible with the 1/v law, contrary to the previous differential experiments. In order to take this information into account, the resonance parameters were revised in 1993 [3] in the energy range from 0.002 to 3 eV and adopted in a new release of ENDF/B-VI and in the JENDL3.2 file. This evaluation was initially suggested for JEFF3; nevertheless, the experimental validation of the data through integral experiments (see [4] and [5]) has demonstrated that despite the reduction of C/E discrepancies given by the 1993 evaluation, the ^{241}Pu capture cross-section is still significantly underestimated. The purpose of the present work is to address this issue and describe the new revision.

2 Revision of the ^{241}Pu resonance parameters

The 1988 evaluation was mainly based on selected neutron transmission and fission cross section measurements. The only capture cross sections available were those obtained from simultaneous fission and absorption cross section measurements performed by Weston and Todd at ORNL [6]. In the 1993 evaluation, a renormalization coefficient of 0.952 for the fission and of 0.914 for the capture, was applied to the original Weston EXFOR data. In the present work, a new SAMMY [7] analysis of the Weston data was first performed in the energy range 0.03 eV to 20 eV; the neutron widths of the resonances were kept at the values of the previous evaluations in order to preserve the values of the calculated absorption cross section. The best consistent fit of the experimental data was obtained by allowing a fission renormalization of 0.955 and a capture renormalization of 0.970, which corresponds to a 5% increase of the average capture cross section compared to the 1993 evaluation.

A separate fit of the 0.26 eV resonance was then restarted in the energy range 0.01 eV to 1.0 eV in order to obtain the best values of the cross sections in the low energy range, using as input to SAMMY the total cross sections of Young and Smith [8], the 1991 fission cross sections of Wagemans [2] and the fission and capture data of Weston renormalized as above. The low energy part of the Weston data (below 0.03 eV), not consistent in shape with Wagemans data, was not considered. The new parameters of the 0.26 eV resonance are compared with those obtained in the previous evaluations in Table 2. The different values of the capture widths are due to a different normalization of the Weston capture data. The total widths are nearly the same. Two fission channels were used in the analysis; Γ_{f1} and Γ_{f2} are the corresponding partial fission widths. The sign is the sign of the fission width amplitude. The different sharing of the fission widths into the fission channels is due to different interpretation of the fission interference effects and to a small resonance added at 0.150 eV to improve the fit of the capture cross section.

The cross sections calculated with the new resonance parameters are compared to the experimental data in Fig 5 and 5. The cross sections at 0.0253 eV are also compared to the standard values of Axton in Table 3. The values of the integral cross sections obtained in 1988, 1993 and the present work are displayed in Table 4 ; the differences between the results are within the experimental errors of 2-3% on the fission and of 10-15% on the capture given by Weston. The Table of the resonance parameters in the energy range from thermal to 20 eV is given in Appendix 1.

3 Experimental validation with Post-Irradiation Experiment

The Post-Irradiation Experiments (PIE) on PWR spent fuel can provide accurate information on average cross-section in a PWR spectrum. The analysis of the measured isotopic ratios Pu241/U238 and Pu242/U238 is a good way to test the ^{241}Pu absorption and capture average cross-section, respectively. Furthermore, the ^{243}Am build-up is related to the ^{241}Pu capture cross-section (^{242}Pu formation) through the following process :

$$Pu241 \overset{n,\gamma}{\longrightarrow} Pu242 \overset{n,\gamma}{\longrightarrow} Pu243 \overset{\beta\ (4.9h)}{\longrightarrow} Am243 \quad (1)$$

To assess the effect of the new ^{241}Pu resonance parameters on build-up prediction, the irradiated fuel experiment performed in the Gravelines French PWR (UOX 17×17 assembly, 4.5% U235 enrichment) was used. Deterministic burn-up calculations were carried out with the French multigroup transport code APOLLO2 [10]. An accurate modelling for depletion calculations has been defined in previous studies [9]. The influence of sensitive physical parameters has been studied in detail, such as irradiation history, radial distribution of the fuel temperature within the pellet and its variation during irradiation, concentration of soluble boron, "stretch-out" operating mode. At every burn-up step, the APOLLO2 flux calculations were checked with the reference Monte-Carlo continuous-energy TRIPOLI4 [11] calculations. Furthermore, experimental uncertainties have been assessed from a detailed sensitivity study.

In a PWR spectrum, about 50% of the ^{241}Pu capture and fission rate occurs in the 0.26 eV resonance (from 0.1 eV to 1 eV). In the 0.01 eV - 0.1 eV energy range (left wing of the 0.26 eV resonance), about 35% of the reaction rate takes place. The rest of the reaction rate occurs above 1 eV and the contribution of the energy range below 0.01 eV is small.

Compared to the JEF2.2 evaluation, in the Gravelines spectrum at 10 GWd/t, the total ^{241}Pu capture rate increases by 5.1 % and the fission rate decreases by about 2.7 %. The absorption reaction rate is slightly lower by 0.7 %. Table 5 shows the C/E on ^{241}Pu, ^{242}Pu and ^{243}Am build up using JEF2.2 (1988 evaluation), JENDL3.2 (1993 evaluation) and the present revision (within the JENDL3.2 evaluation).

Table 5 shows that the decrease of the ^{241}Pu absorption cross-section leads to an improvement of the ^{241}Pu build-up prediction. The higher ^{241}Pu capture cross-section in the present work has significantly reduced the longstanding discrepancy on the ^{242}Pu formation. The correction of the ^{243}Am prediction shows that the previous discrepancy was partly linked to the underestimation of the ^{242}Pu build-up. Furthermore, the large discrepancy observed in the ^{244}Cm (-17% at 60 GWd/t) and ^{245}Cm (-18% at 60 GWd/t) build-up prediction [4] is slightly corrected with the new resonance parameters.

4 Conclusion

A new SAMMY analysis of the ^{241}Pu cross-sections was performed in the thermal range up to 20 eV to take account of the trends given by integral experiments (Post-Irradiation Experiment). This revision has led to a higher ^{241}Pu capture cross-section in the 0.26 eV resonance compatible with the differential measurements. The integral validation through the P.I.E in French PWR assemblies has shown that the new set of resonance parameters improves greatly the Pu242 formation as well as the prediction of ^{243}Am, ^{244}Cm and ^{245}Cm in a PWR. These revised resonance parameters were adopted for JEFF3.0 and proposed for the next release of ENDF/B-VII.

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Appendix 1 : ^{241}Pu resonance parameters from thermal to 20 eV

Energy eV	spin	Γ_n eV	Γ_γ eV	Γ_{f1} eV	Γ_{f2} eV
-5.953000E+1	2	5.961E-1	4.450E-2	4.153E-1	4.298E-2
-5.580000E+0	3	2.289E-3	3.644E-2	1.648E+0	1.576E-2
-1.405000E+0	3	1.513E-6	1.925E-2	-1.871E-2	3.694E-4
-1.030800E-1	3	1.263E-5	3.890E-2	2.026E-2	1.009E-3
1.500714E-1	2	3.839E-7	4.229E-2	2.000E-2	2.000E-2
2.640324E-1	3	4.367E-5	3.463E-2	-4.271E-2	3.206E-2
1.725295E+0	2	2.074E-6	4.030E-2	6.786E-2	2.823E-1
4.285520E+0	3	5.758E-4	3.471E-2	2.780E-2	0.000E+0
4.587276E+0	2	4.787E-4	3.677E-2	-1.922E-2	1.211E-1
5.813320E+0	2	2.774E-3	6.007E-2	-1.180E+0	1.805E-1
6.945672E+0	3	6.201E-4	3.546E-2	-1.075E-1	4.514E-4
8.622176E+0	3	7.797E-4	3.358E-2	8.195E-4	5.920E-2
9.649351E+0	2	5.924E-4	3.908E-2	2.037E-1	1.707E-3
9.938550E+0	2	1.894E-3	5.661E-2	8.886E-1	2.920E-4
1.281875E+1	2	8.637E-4	4.357E-2	-2.106E-1	4.218E-4
1.344322E+1	3	2.383E-3	3.690E-2	3.579E-7	2.420E-2
1.477338E+1	2	7.994E-3	3.573E-2	7.398E-2	2.628E-2
1.601637E+1	2	1.721E-3	3.889E-2	-4.227E-1	7.748E-3
1.669268E+1	3	1.074E-3	3.446E-2	2.079E-1	1.542E-3
1.786955E+1	3	2.968E-3	4.026E-2	5.960E-4	1.835E-2
1.828045E+1	3	1.301E-4	4.052E-2	-3.166E-2	1.510E-4
2.072000E+1	3	3.453E-4	3.975E-2	1.211E-2	4.409E-2

Table 1: ^{241}Pu resonance parameters from thermal to 20 eV

Tables

	Γ_γ meV	Γ_n meV	Γ_{f1} meV	Γ_{f2} meV
1988	32.555	0.04371	-78.04	-0.481
1993	33.350	0.04252	-50.42	25.36
this work	34.630	0.04367	-42.71	32.06

Table 2: Parameters of the 0.26 eV resonance from the different evaluations

	Standard (b)	present work (b)
fission	1012.68 ± 6.58	1012.2
capture	361.29 ± 4.95	363.0
scattering	12.17 ± 2.62	11.3
Total	1386.14 ± 8.64	1386.5

Table 3: ^{241}Pu values of cross-sections at 0.0253 eV

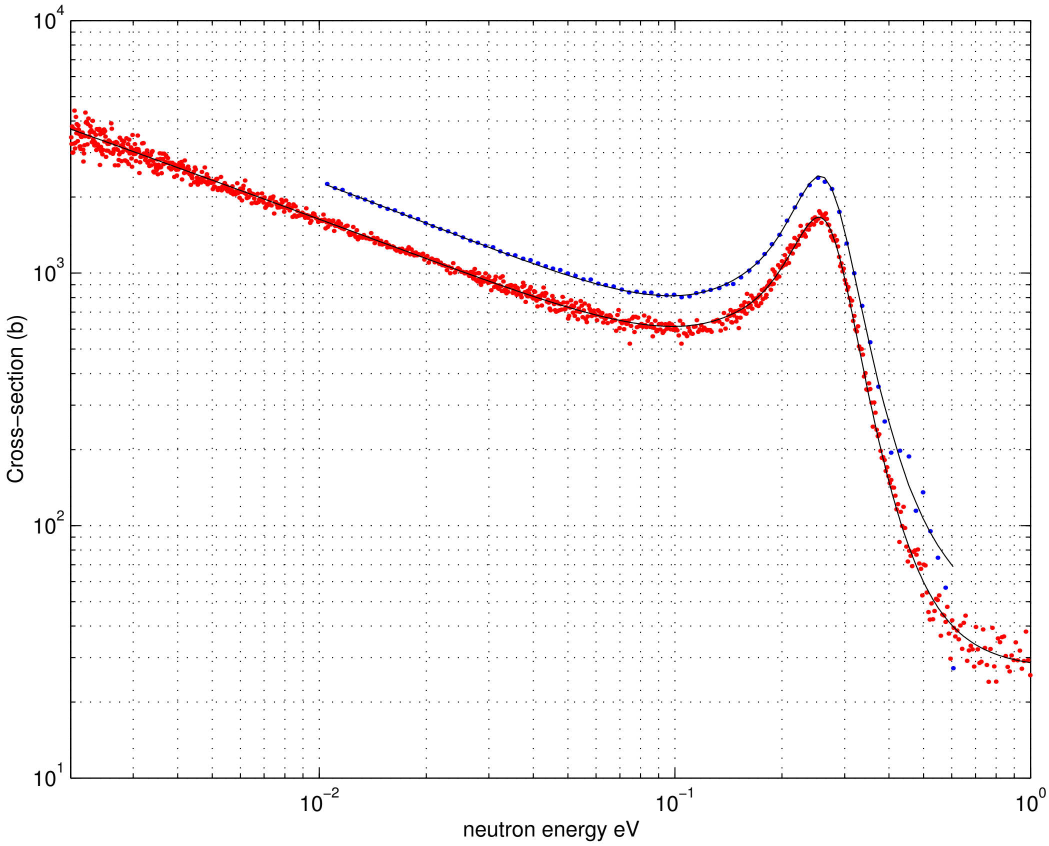
Energy Range eV	Integrated Capture cross-section b-eV				Integrated Fission cross-section b-eV			
	1988	1993	Present	diff	1988	1993	Present	diff
0.0021-0.020	12.28	12.25	12.51	1.8%	30.61	31.06	31.43	2.7%
0.0200-0.030	3.68	3.67	3.72	1.1%	10.22	10.24	10.36	1.4%
0.0300-0.100	15.39	15.28	15.59	1.3%	50.02	49.02	49.14	-1.8%
0.1000-0.500	109.47	110.58	117.40	7.2%	270.84	262.76	263.84	-2.7%
0.5000-1.000	5.87	5.90	6.03	2.7%	17.64	17.93	17.89	1.4%
1.0000-3.000	7.14	7.30	7.16	0.3%	55.62	54.88	52.89	-5.2%
3.0000-20.00	1138.52	1213.07	1233.78	8.4%	3066.37	3038.63	3025.61	-1.3%

Table 4: Integral values of the ^{241}Pu fission and capture cross-section, the percentage deviations between the present results and the 1988 results are given.

BU (GWd/t)	26.9	38.4	50.3	59.8
Pu241/U238 (JEF2.2)	-6.3 ± 2.3	-5.0 ± 1.8	-3.8 ± 1.6	-3.1 ± 1.6
Pu242/U238 (JEF2.2)	-10.5 ± 4.0	-9.7 ± 3.4	-8.8 ± 3.1	-8.6 ± 2.8
Am243/U238 (JEF2.2)	-18.0 ± 6.2	-11.6 ± 5.2		-8.8 ± 4.4
Pu241/U238 (JENDL3.2)	-5.4 ± 2.3	-3.8 ± 1.8	-2.5 ± 1.6	-1.6 ± 1.6
Pu242/U238 (JENDL3.2)	-9.0 ± 4.0	-7.9 ± 3.4	-6.8 ± 3.1	-6.4 ± 2.8
Am243/U238 (JENDL3.2)	-16.6 ± 6.2	-9.9 ± 5.2		-6.8 ± 4.4
Pu241/U238 (This work)	-5.9 ± 2.3	-4.4 ± 1.8	-3.1 ± 1.6	-2.4 ± 1.6
Pu242/U238 (This work)	-5.2 ± 4.0	-4.3 ± 3.4	-3.2 ± 3.1	-2.9 ± 2.8
Am243/U238 (This work)	-12.8 ± 6.2	-6.5 ± 5.2		-3.7 ± 4.4

Table 5: C/E-1 in % using JEF2.2, JENDL3.2 and the present ^{241}Pu resonance parameters for the Gravelines experiment.

^{241}Pu total cross section of Young and Smith and fission cross section of Wagemans in the energy range below 1 eV. The solid lines are the results of a SAMMY calculation using the new resonance parameters.



^{241}Pu Weston fission (upper curve) and capture cross sections (lower curve) in the energy range 0.03 eV to 1 eV. The solid lines are the results of a SAMMY calculation using the new resonance parameters.