Evaluation of ^{238}U Resonance Parameters from 0 to 20 keV

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Abstract. The neutron resonance parameters of ^{238}U were obtained in the energy range 0 to 20 keV from a sequential SAMMY [1] analysis of the most recent high-resolution neutron transmission and neutron capture cross-section measurements. Special care was taken in the analysis of the lowest s-wave resonances leading to resonance parameters slightly different from those of ENDF/B-VI (Moxon-Sowerby resonance parameters [2]). The resolved-resonance range was extended to 20 keV, taking advantage of the high-resolution neutron transmission data of Harvey [3] and neutron capture data of Macklin et al. [4]. Preliminary integral tests were performed with the new resonance parameters; thermal low-enriched benchmark calculations show an improvement of the k_{eff} prediction, mainly due to a 1.5% decrease of the capture cross section at 0.0253 eV and about a 0.4% decrease of the effective shielded resonance capture integral.

INTRODUCTION

Preliminary results of the evaluation of the ^{238}U resonance parameters in the energy range 0 to 20 keV were presented in [5] and [6]. In the present work, the results of the high-resolution neutron capture cross-section measurement of Macklin et al. [4] were added to the experimental data base with the aim of improving the accuracy of the parameters above 250 eV. Since the parameters of the first s-wave resonances play a major role in the determination of the resonance capture integral, the transmission data in the resonances at 6.67, 20.9, and 36.0 eV taken by Meister et al. [7] using metallic and oxide samples were also analyzed.

THE EXPERIMENTAL DATA BASE AND METHOD OF ANALYSIS

The experimental data used in the present evaluation are listed in Table 1. The SAMMY analysis was performed with the Reich-Moore formalism. The Doppler broadening used the Free Gas Model with an effective temperature accounting for chemical binding in samples [8]. As explained in the next section, the first ^{238}U resonance at 6.67 eV was studied with a more accurate model known as the Crystal Lattice Model [8]. The capture measurements were also corrected for self-shielding and multiple scattering effects. Experimental resolution functions were found in the original publications; however, some of the resolution function parameters are not well known, especially those defining the slowing down of neutrons in the moderator. Preliminary analyses of isolated resonances, or group of resonances, in various energy ranges, were performed to find the best values for the moderator parameters, in particular, the exponential tail of the resolution function used in SAMMY. Experimental corrections of transmission and capture measurements were systematically checked by allowing normalization and background parameters to vary in the SAMMY sequential fits.

The Harvey transmission data in the energy range 1 to 20 keV were first analyzed to assess the contribution of the resonances pertaining to the external region and the effective scattering radius R'. The value of 9.45 fm obtained for R' agrees with the value obtained by Olsen from the analysis of his transmission data.

THERMAL TO 1 KEV

The energy range below 1 keV is crucial for the calculation of thermal reactors and needs to be treated with great care. The thermal capture value was adjusted to $\sigma_0 = 2.683\ b$, following the recent recommendations of Trkov et al. [15]. The shape of the capture cross section in the thermal range was checked against capture measurements of Corvi et al. [10] performed at GELINA.

For the large resonances at 6.7, 20.8 and 36.6 eV, the seven transmission spectra of Olsen [13] were fitted using the four transmission measurements performed at room temperature at GELINA by Meister et al. [7] and the capture measurements of de Saussure et al. [12]. The low-temperature (23.7 K) data of Meister were not in-

TABLE 1. Experimental database used in the present SAMMY analysis

Energy Range	Reference	Measurement Type	Sample Thickness (at/b)	Flight path Length (m)
0.0253 eV	Poenitz et al. [9] ANL 1981	Activation		
Thermal Range	Corvi et al. [10] GELINA 1997	Capture	1 sample 0.0010	8.7
> 5 eV	Defilippo et al. [11] ORELA 1980	fission		40.
6 eV - 38 eV	Meister et al. [7] GELINA 1997	Transmission	4 samples U and UO_2	26.5
6 eV - 100 keV	de Saussure et al. [12] ORELA 1973	Capture	1 sample 0.0028	40.
0.5 eV - 4 keV	Olsen et al. [13] ORELA 1977	Transmission	7 samples 0.0002 to 0.175	42.
300 eV - 100 keV	Olsen et al. [14] ORELA 1979	Transmission	4 samples 0.0038 to 0.175	150.
250 eV - 130 keV .	Macklin et al. [4] ORELA 1988	Capture	2 samples 0.0031, 0.0124	150.
1 keV - 100 keV	Harvey et al. [3] ORELA 1988	Transmission	3 samples 0.0124 to 0.175	200.

cluded in the fit. Special attention was paid to the modeling of Doppler broadening to take chemical binding in metallic and oxide samples into account. As discussed in another paper [16], the Crystal Lattice Model (CLM) of SAMMY that explicitly accounts for phonon creation and absorption in the atomic lattice was used to describe the shape of the 6.7 eV resonance. Compared with the traditional approach using the Free Gas Model (FGM) and a fitted temperature, the influence of the CLM on neutron and radiation widths was found to be small. To get a better correction of errors in normalizations and backgrounds, fits were performed resonance by resonance up to 60 eV (see Fig. 1).

Below 500 eV, the normalizations of Olsen transmissions were found to be accurate within 1% except for the thickest sample which requires significant energy-dependent renormalization (from $\approx$ 1.05 at 6 eV to $\approx$ 1.03 above 500 eV). Background adjustments to correct negative transmission values in black s-wave resonances were also applied to the data from the three thickest samples data to improve the fits. Values for chi-square per degree of freedom were near unity except for the thickest sample (χ^2 between 2 and 3). As previously shown by Moxon and Sowerby [2], the de Saussure capture data, well normalized at 6.6 eV, needs to be significantly renormalized to be consistent with the Olsen measurements above the first resonance (about 1.08 around 100 eV). A residual constant background correction ($\approx$100 mb) was also found.

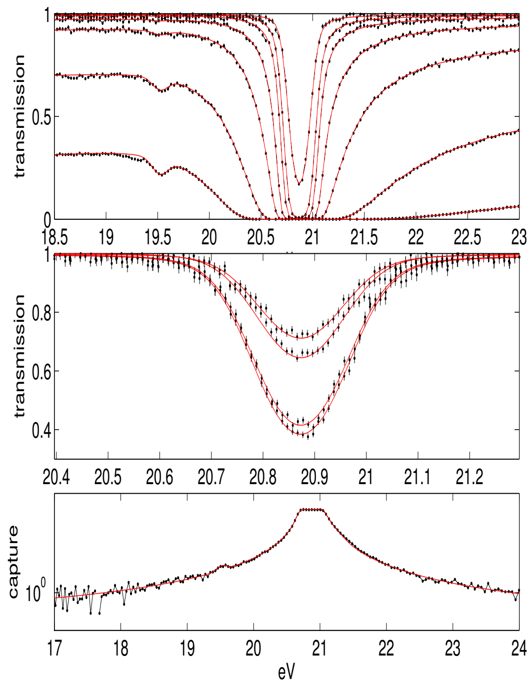

FIGURE 1. Sequential fits for the 20.8 eV resonance of the seven samples of Olsen et al. [13], the four samples of Meister et al. [7], and the capture measurements of de Saussure [12].

In the present work, the radiation widths of the lowest s-wave resonances, mainly determined by the thickest sample (0.17 at/b) data of Olsen, were fitted and are displayed in Table 2. For the important 6.67-eV resonance, the radiation width analyzed with the CLM is very close to the Moxon-Sowerby value while the neutron widths is smaller by 1.2%. For other s-wave resonances below 102 eV, the Γ_γ extracted are generally about 1-2% higher than those deduced by Moxon with the same data. This can be explained by the use of a different value of effective radius. The values of Γ_γ are still under investigation. The neutron widths of the lowest s-wave are generally smaller.

From 250 eV to 1 keV, the Macklin et al. [4] capture measurements were included in the analysis. The sequential fits led to a strong renormalization of the thin and thick samples by about 1.08 above 500 eV. A residual background correction ($\approx$80 mb for the thick sample and $\approx$140 mb for the thin sample) is also deduced from data between resonances. The capture data in the region 250-500 eV required a more complex energy-dependant renormalization.

The spins of several p-wave resonances were changed from the original ENDF/B-VI evaluation, as measured by Gunsing et al [18] using analysis of γ-rays spectra after capture. Reliable estimates of the p-wave neutron widths below 300 eV were obtained by Crawford et al. [19] from thick-sample transmission measurements to study parity violation in ^{238}U resonances and were used as prior parameters in the fit.

1 TO 20 KEV

Examples of SAMMY fits of the experimental data are given in Fig. 2 and 3. Figure 2 compares Olsen thick-sample transmission data, Macklin capture data, and de Saussure capture data with the SAMMY calculations using the present resonance parameters in the neutron energy range 1.5 to 1.75 keV. Figure 3 shows the results of the SAMMY fit in the energy range 17.25 to 17.50 keV for the Harvey thick-sample transmission and the Macklin capture data.

In general, the thick-sample transmissions calculated from the resonance parameters and averaged over 1-keV energy intervals agree within about 1% with the experimental values of the Harvey data and within 1.5% with the Olsen data, in agreement with the experimental errors quoted by the authors of the measurements.

As previously stated, the fit of the capture data could not be obtained without large normalization and background corrections; a background subtraction of 85 $\pm$ 30 mb for the thick sample and 140 $\pm$ 60 mb for the thin sample, followed by a renormalization of 1.13, were needed to fit the Macklin data. Macklin et al. normalized their data by using the Moxon technique in the resonances of small neutron widths in the energy range 600 to 1100 eV. The background correction for the de Saussure data was 40$\pm$20 mb and the renormalization about 0.91.

In the energy range 1 to 10 keV, the average values of the infinitely dilute capture and elastic cross section calculated from the present resonance parameters are respectively 2.4% and 1.3% larger than the ENDF/B-VI values. From 10 to 20 keV, the ENDF/B-VI cross sections were obtained from a statistical calculation performed by Froehner [17]; the present evaluation calculates capture cross sections that are smaller by 4.5% on average. This discrepancy might arise from p-wave resonances missed in the current analysis and is still under investigation.

STATISTICAL PROPERTIES OF THE RESONANCE PARAMETERS

The identification of the large s-wave resonances was straightforward from the asymmetry due to the potential-resonance interference effect. All the other resonances had to be distributed among three families: small s-wave resonances, $1/2^-$ p-wave resonances, and $3/2^-$ p-wave resonances, by trying to keep the 2J+1 dependence of the level density. However, because the area of the p-wave resonances in the capture cross-section depends on the spin, some spin assignments could be made by using this property in the Macklin capture data.

According to the 2J+1 law of the level-density spin dependence, the number of J = 1/2 p-wave resonances should be roughly the same as the number of the s-wave resonances, and the number of J = 3/2 p-wave resonances should be twice this number. The fits to the experimental data were obtained by using, in the energy range 0 eV to 20 keV, 898 s-wave resonances, as well as 849 and 1565 p-wave resonances of spin J = 1/2 and J = 3/2, respectively. In the low-energy part of the data, most of these resonances are seen in the experimental transmission or experimental capture (see, for example, Fig. 2). Due to the larger experimental resolution width and the increasing number of p-wave resonances, the number of multiplets becomes more and more important in the high energy part of the data (see, for example, Fig. 2). For instance, the number of resonances in each peak of the cross section could be three or four in the energy region around 19 keV neutron energy. Another type of resonance was added to the resonance set: those of very small neutron width values, which were not seen in the experimental transmission data or in the experimental capture data. As shown in Fig. 4, the statistical analysis of the resonance parameters shows a good agreement with the Wigner distribution for both s-wave and p-wave level spacings.

TABLE 2. Resonance parameters for ^{238}U s-wave when the radiation widths are fitted (left) and when kept to the ENDF/B-VI values (Moxon et al. [2]). The small uncertainty values quoted in this table takes into account only the statistical uncertainty of the measurements. The actual values accounting for systematical uncertainties are much larger.

Energy	Γ_γ meV present work	Γ_n meV present work	Γ_γ meV ENDF/B-VI	Γ_n meV ENDF/B-VI	Γ_n meV present work Γ_γ from ENDF/B-VI
	R' = 9.45 fm		R' = 9.42 fm		R' = 9.45 fm
6.674	23.01 ± 0.02	1.475 ± 0.001	23.00	1.493	1.476 ± 0.001
20.871	23.12 ± 0.03	10.04 ± 0.01	22.91	10.26	10.07 ± 0.01
36.682	23.41 ± 0.04	33.43 ± 0.02	22.89	34.13	33.55 ± 0.02
66.031	23.64 ± 0.10	24.17 ± 0.04	23.36	24.60	24.23 ± 0.03
80.747	23.31 ± 0.41	1.877 ± 0.01	23.00	1.865	1.877 ± 0.01
102.56	24.53 ± 0.14	70.62 ± 0.08	23.40	71.70	71.03 ± 0.08

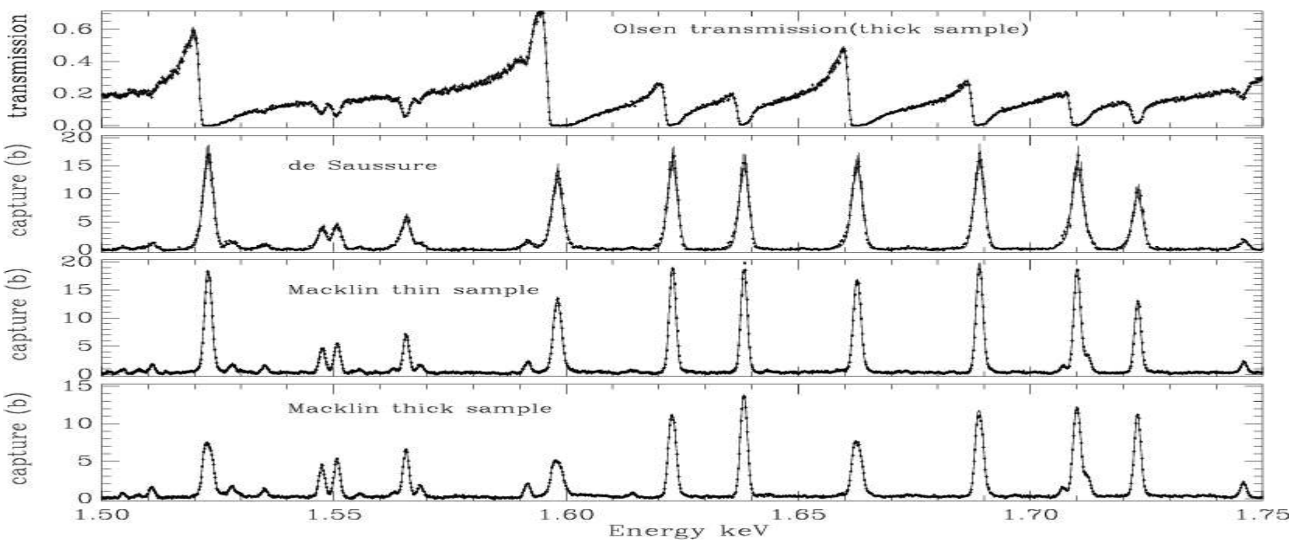

FIGURE 2. SAMMY fits of Olsen [14] transmission data with the capture data of de Saussure [12] and Macklin et al. [4].

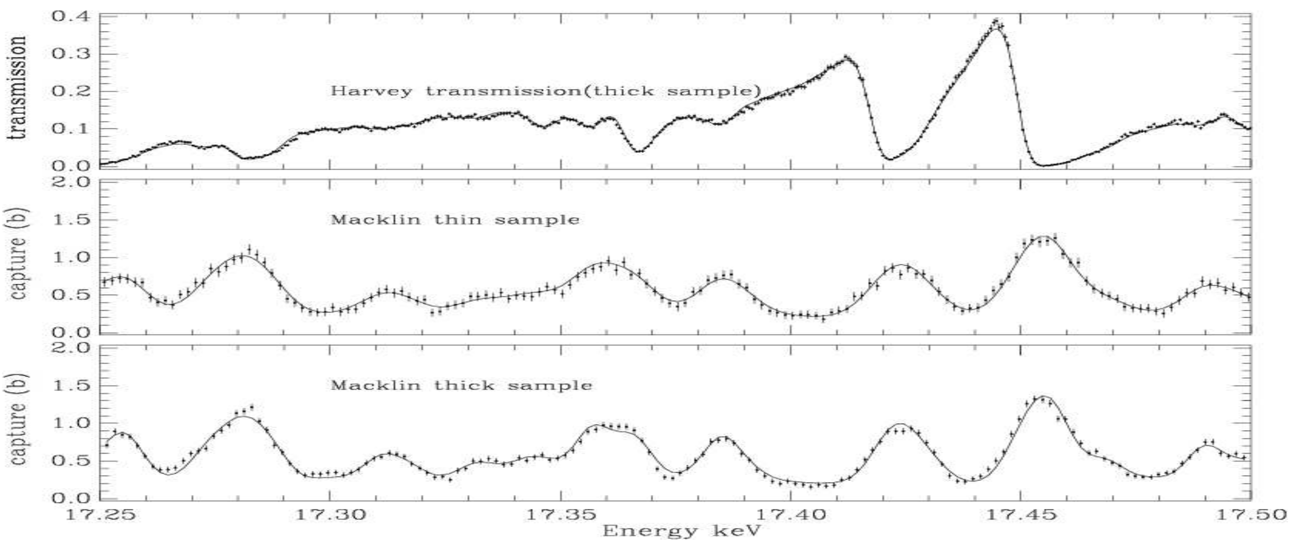

FIGURE 3. SAMMY fits of Harvey [3] transmission data with the capture data of Macklin et al. [4].

The agreement of the distribution of the reduced neutron widths with the Porter-Thomas distribution is also fairly good. The values of the neutron strength functions are given in Table 3.

TABLE 3. Neutron strength functions S_0, S_1 multiplied by 10^4.

Energy range	Present work s-wave S_0	Present work p-wave S_1
0 - 10 keV	0.980±0.064	1.596±0.067
10 - 20 keV	1.074±0.073	1.710±0.068
	ENDF/B-VI s-wave S_0	**ENDF/B-VI** p-wave S_1
0 - 10 keV	0.947±0.062	1.577±0.060

These values can be compared with the results of $S_0 = (1.077\pm0.016) \times 10^{-4}$ and $S_1 = (1.846\pm0.031) \times 10^{-4}$ from an unpublished statistical model fit of the average total cross sections in the energy range 10 keV to 100 keV.

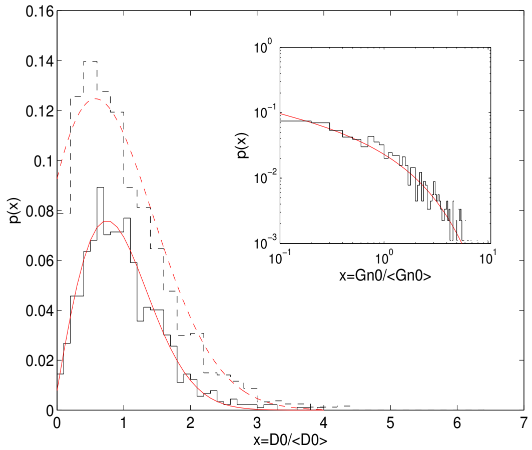

FIGURE 4. Distribution of s-wave (solid lines), p-wave average spacing (dashed line) and s-wave reduced neutron widths (smaller figure) compared with a Wigner distribution, a distribution of two uncorrellated population (p1/2 and p3/2), and Porter-Thomas distribution, respectively.

IMPACT ON INTEGRAL EXPERIMENTS

To assess the effect of the present resonance parameters (with Γ_γ from ENDF/B-VI below 102 eV) on integral experiments, the effective (or shielded) capture resonance integral was computed:

$$I_{eff} = \int_{E_{min}}^{E_{max}} \sigma(E)\varphi(E)\frac{dE}{E} = \int_{u_{min}}^{u_{max}} \sigma(u)\varphi(u)du \quad (1)$$

The lethargy variable u is equal to $\ln(E_0/E)$, and $\varphi(u)$ is the so-called fine-structure function per unit of lethargy. The main interest of the effective resonance integral concept is that it is more representative of the reaction rates in thermal reactor calculations than the usual infinitely-dilute resonance integral. $\varphi(u)$ and I_{eff} were calculated using the GROUPR module of NJOY [20], which solves the integral slowing-down equation in homogeneous material assuming isotropic scattering in the center-of-mass system. The resonance integral has been evaluated at 293 K from 1 eV to 10 keV for several dilution values (or background cross section).

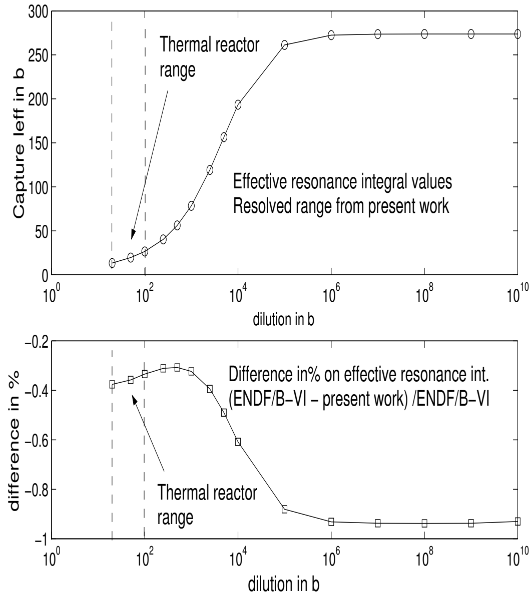

FIGURE 5. Effective resonance integral (1 eV - 10 keV) for various dilution values and comparison with ENDF/B-VI values.

Figure 5 shows that the reaction rate in PWR-like systems should be decreased by about 0.4% mainly due to smaller neutron widths of the lowest s-wave resonances. Consequently, the ^{239}Pu build up in burn up calculations should be slightly reduced as well.

For illustrative purposes, first tests on selected k_{eff} thermal benchmarks from the ICSBEP database were performed with the MCNP5 Monte Carlo transport code [21] with cross-sections processed by the NJOY99 code [20]. Present resonance parameters have been merged with the newest and still preliminary evaluation above the unresolved range by Los Alamos National Laboratory. Compared with the ENDF/B-VI.8 evaluation, the impact of the new resonance parameters is about 100 - 150 pcm on low-enriched thermal lattices, depending on the moderation ratios, and should have a negligible impact on the k_{eff} of low- and high-enriched solution systems (Leu-Sol-Therm and Heu-Sol-Therm) as well as fast systems.

As shown in Table 4, when combined with the preliminary LANL high-energy evaluation, the total effect on Leu-Comp-Therm compared with ENDF/B-VI is a

TABLE 4. Benchmarks k_{eff} calculated with ^{238}U from ENDF/B-VI.8, the preliminary LANL evaluation above the unresolved range, and the present ORNL resonance parameters set. The other isotopes nuclear data comes from the standard ENDF66 library of MCNP (ENDF/BVI.6). Statistical uncertainties of the Monte Carlo calculations are about 40 pcm (1σ).

ICSBEP	k_{eff}		
name	^{238}U ENDF/B-VI.8	^{238}U ENDF/B-VI.8 + LANL	^{238}U + LANL + ORNL
LCT6-1	0.99240	0.99634	0.99790
LCT6-4	0.99299	0.99593	0.99797
LCT6-9	0.99500	0.99747	0.99818
LCT6-14	0.99521	0.99778	0.99962

significant increase in k_{eff} that greatly improves the long-standing Leu-Comp-Therm reactivity underestimation (see [22] for further details). Similar improvement is also observed with the new high-energy evaluation from Bruyeres-Le-Chatel [22].

CONCLUSION

A new evaluation of ^{238}U was undertaken at the Oak Ridge National Laboratory. This work aimed at investigating the current underprediction of k_{eff} observed with the modern libraries (JEFF3.0, JENDL3.3, and ENDF/B-VI.8). This evaluation is a follow-up of an important previous work on ^{238}U resonances (NEANDC task force on ^{238}U [2]) up to 10 keV. The present analysis includes, for the first time, the most recent high-resolution transmission of Harvey et al. [3] and a complete analysis of capture cross-section data of Macklin et al. [4], allowing the resolved range to be extended to 20 keV. This new evaluation, proposing a slight decrease of the effective capture resonance integral, improved the prediction of integral thermal benchmarks from $\approx$ 70 to 200 pcm. Work is still on-going to improve this evaluation.

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REFERENCES

1. N. M. Larson, *Updated Users' Guide for SAMMY: Multilevel R-Matrix Fits to Neutron Data Using Bayes' Equations,* ORNL/TM-9179/R6 and ENDF-364, 2003.
2. M. Moxon and M. Sowerby, *Summary of the Work of the NEANDC Task Force on ^{238}U, Nuclear Energy Agency OECD report, 1994.*
3. J. A. Harvey et al., "High Resolution Neutron Transmission Measurements on ^{235}U, ^{239}Pu and ^{238}U," *International Conference on Nuclear Data for Science and Technology, Mito, Japan (1988).*
4. R. L. Macklin et al., "High Energy Resolution Measurement of the ^{238}U Neutron Capture Yield in the Energy Region Between 1 and 100 keV" *International Conference on Nuclear Data for Science and Technology, Mito, Japan (1988).*
5. H. Derrien, L.C. Leal, and N. M. Larson "New Evaluation of ^{238}U Neutron Resonance Parameters" *International Conference on Nuclear Criticality Safety, Tokai, Japan, Oct. 20 - 24 (2003)*
6. H. Derrien, L.C. Leal, and N. M. Larson, "Status of a New Evaluation of the Neutron Resonance Parameters of ^{238}U at ORNL," *PHYSOR-2004, International Topical Advances in Reactor Physics and Mathematics and Computation*, Chicago, USA, April 25-29 (2004).
7. A. Meister et al., "Experimental Study of the Doppler Broadening of Neutron Resonances at GELINA," *International Conference on Nuclear Data,* Long Island, October 5 - 8, 1998.
8. W. E. Lamb, "Capture of Neutrons by Atoms in a Crystal," *Phys. Rev.* **52**, 295 (1937).
9. W. P. Poenitz et al., *Nucl. Sci. Eng.* **78**, 239-247 (1981).
10. F. Corvi and G. Fioni, "Shape of the ^{238}U Neutron Capture Cross-Section in the Range 0.002-0.1 eV," *International Conference on Nuclear Data for Science and Technology*, Mito, Japan, (1988)
11. F. C. Difilippo et al. *Phys. Rev.* **21**, (4), (1980).
12. G. de Saussure et al., *Nucl. Sci. Eng.* **5**, 385 (1973).
13. D. K. Olsen et al., *Nucl. Sci. Eng.,* **62**, 479 (1977).
14. D. K. Olsen et al., "Measurement and Resonance Analysis of Neutron Transmission Through Uranium-238," *Nucl. Sci. Eng.* **69**, 202-222 (1979).
15. A. Trkov et al., "Revisiting the ^{238}U Thermal Capture Cross-Section and Gamma Emission Probabilities from Np239 Decay," *Submitted to Nucl. Sci. Eng.*
16. A. Courcelle, G. Noguere, and N. Larson, "Experimental tests of the Crystal Lattice Model of SAMMY," *International Conference on Nuclear Data for Science and Technology,* Santa Fe, N.M., USA, 2004
17. F. H. Frohner, *Nucl. Sci. and Eng.* **103**, 119-128 (1988).
18. F. Gunsing, K. Athanassopulos, and F. Corvi, *Phys. Rev. C* **56**, (3), (1997).
19. B. E. Crawford et al., *Phys. Rev. C* **58**, (2), (1998).
20. R. E. MacFarlane and D.W. Muir, *The NJOY Nuclear Data Processing System, LA-12740-M.*
21. F. B. Brown et al. *MCNP: A General Monte Carlo N-Particle Transport Code, Version 5, LA-UR-1987, 2003.*
22. A. Courcelle et al., "First Conclusions of the WPEC/Subgroup-22" *International Conference on Nuclear Data for Science and Technology,* Santa Fe, N.M., USA, 2004.