*PHYSOR 2004 -The Physics of Fuel Cycles and Advanced Nuclear Systems: Global Developments*
Chicago, Illinois, April 25-29, 2004, on CD-ROM, American Nuclear Society, Lagrange Park, IL. (2004)

# An Unresolved Resonance Evaluation for $^{235}U$

L. C. LEAL*, H. DERRIEN, and N. M. LARSON
*Oak Ridge National Laboratory*
*P. O. Box 2008*
*Oak Ridge, TN 37831 USA*

This paper discusses the resonance evaluation of the $^{235}U$ cross sections in the unresolved resonance region from 2.25 keV up to 25 keV and its use in benchmark calculations for criticality safety applications.

A criticality safety calculation for a nuclear system with an energy spectrum that peaks in the intermediate energy region requires accurate neutron cross sections in both the resolved and the unresolved resonance regions. A resolved resonance region evaluation of the $^{235}U$ cross section was performed in the 1990s, and the evaluation has greatly improved results of benchmark calculations.[1)] Average values of the Reich-Moore resonance parameters obtained in the resolved region were used to initiate a new resonance evaluation of the $^{235}U$ cross sections in the unresolved resonance region. The experimental data used in the analysis consisted of high-resolution transmission data, plus fission and capture cross-section data measured at the Oak Ridge Electron Linear Accelerator (ORELA). The evaluation was performed using the computer code SAMMY, which incorporates a methodology for data evaluation in the unresolved resonance region.

***KEYWORDS*: unresolved energy region, neutron cross section, data evaluation**

## 1. Introduction

In the resolved resonance range, the experimental resolution is smaller than the width of the resonances; consequently, resonances can be "seen" and resonance parameters can be extracted via cross-section fitting using methodology such as the R-matrix formalism and generalized-least-squares techniques in the code SAMMY.[2)] In the unresolved resonance region, however, the fluctuations in the measured cross sections are smaller than those in the resolved range but are still important for correct calculation of the energy self-shielding of the cross sections. These fluctuations are due to unresolved multiplets of resonances for which it is not possible to determine parameters of individual resonances as in the resolved region. The mechanism utilized for cross-section treatment in the unresolved region is based on average values of physics quantities obtained in the resolved range. The knowledge of average values for level spacing, strength functions, widths and other relevant parameters is used to infer calculations in the unresolved energy region. Therefore, for consistency, it is desirable that a complete evaluation for both the resolved and unresolved resonance region use a computer code which contains formalisms for both energy regions. To accomplish that, an unresolved resonance formalism was added to the computer code SAMMY.

*Corresponding author, Tel. 865-574-5281, Fax. 865-574-8727, E-mail: leallc@ornl.gov

The unresolved resonance formalism included in SAMMY is based on the methodology used in the computer code FITACS.[3] FITACS is a statistical model code developed by F. Froehner that uses the Hauser-Feshbach theory for the calculation of average total and partial cross sections. The calculations are done in FITACS for the first four angular momenta, *l*, that is, the s-, p-, d-, and f-wave. However, the SAMMY calculations have been extended to as many angular momenta as the user requires. The energy dependence of the parameters are obtained using the Bethe theory for level density, the Hill-Wheeler fission barrier penetration for the fission widths, and the giant dipole model for the capture widths.

SAMMY generates average resonance parameters based on a statistical model analysis of the experimental average cross sections. These parameters are then converted into the ENDF/B format for use in a Single-Level Breit-Wigner cross-section calculation. The primary use of the average resonance parameters is to reproduce the fluctuations in the cross sections for the purposes of energy self-shielding calculations.

## 2. Cross-Section Formalism

From the Hauser-Feshbach theory the partial cross sections can be written as

$$\left\langle \sigma_{ab} \right\rangle = \frac{2\pi g_a}{k_a^2} \frac{T_a T_b}{T} \int_0^\infty dt\, e^{-tT_r/T} \prod_{c \neq \gamma} \left( 1 + \frac{2}{\nu_c} \frac{T_c}{T} t \right)^{-\nu_c/2 - \delta_{ac} - \delta_{bc}} . \qquad \textbf{(1)}$$

Here *a* is the incident channel (neutron) and *b* is the exit channel (gamma or fission); $\nu_c$ and $T_c$ represent the degrees of freedom and the transmission coefficient, respectively, for channel *c*. The transmission coefficients are defined as the sum over all channels as

$$T = \sum_c T_c \; . \qquad \textbf{(2)}$$

The integral allows the calculation of the width fluctuation corrections.

The transmission coefficients for the neutron channels can be written in terms of the average of the scattering matrix $S_{cc}$ as

$$T_c = 1 - \left| \left\langle S_{cc} \right\rangle \right|^2 . \qquad \textbf{(3)}$$

For photon and fission channels, the transmission coefficients for a resonance of spin *J* are, respectively, related to the average width $\left\langle \Gamma \right\rangle$ as

$$T_\gamma = 2\pi \left\langle \Gamma_\gamma \right\rangle / D_J \tag{4}$$

and

$$T_f = 2\pi \left\langle \Gamma_f \right\rangle / D_J \ , \tag{5}$$

where $D_J$ is the average energy level spacing for two consecutive resonances of spin *J*.

In the single-level Breit-Wigner formalism the partial cross section at energy *E* is given as

$$\sigma_{ab} = \frac{\pi g_a}{k_a^2} \sum_\lambda \frac{\Gamma_{\lambda a} \Gamma_{\lambda b}}{\left(E - E_\lambda\right)^2 + \left(\Gamma_\lambda / 2\right)^2} \ . \tag{6}$$

The energy average of the previous equation gives

$$\left\langle \sigma_{ab} \right\rangle = \frac{\pi g_a}{k_a^2} 2\pi\rho_a \left\langle \frac{\Gamma_a \Gamma_b}{\Gamma} \right\rangle . \tag{7}$$

The average indicated in the brackets can be written in terms of individual averages as proposed by Dresner; the average single-level Breit-Wigner partial cross section becomes

$$\left\langle \sigma_{ab} \right\rangle = \frac{\pi g_a}{k_a^2} 2\pi\rho_a \frac{\langle \Gamma_a \rangle \langle \Gamma_b \rangle}{\langle \Gamma \rangle} \times \int_0^\infty dt \prod_{c \neq \gamma} \left( 1 + \frac{2}{\nu_c} \frac{\langle \Gamma_c \rangle}{\langle \Gamma \rangle} t \right)^{-\nu_c/2 - \delta_{ac} - \delta_{bc}} \ . \tag{8}$$

The average partial cross section calculated with the single-level Breit-Wigner formalism is equivalent to the average partial cross section calculated using Eq. 1, given that

$$T_c = 2\pi\rho_c \left\langle \Gamma_c \right\rangle . \tag{9}$$

## 3. Method of Evaluation

Calculation of the average partial cross sections in the unresolved resonance region from the Evaluated Nuclear Data Files (ENDF) is performed with processing codes such as NJOY, AMPX using Eq. (8), for which the average width values are required. In the SAMMY evaluation the fit of the data is performed using Eq. (1)—that is, via the transmission coefficients. Subsequently, the set of parameters that best reproduces the cross section is converted into average widths.

Three sets of experimental data were used in the evaluation from 2.25 keV to 25keV.

1. The effective average total cross section of Harvey et al.[4] obtained from the experimental transmission was analyzed. The Harvey data is a time-of-flight transmission measurement performed at a 80.4-m flight path for two sample thicknesses of 0.0328 and 0.00236 atm/barn. The samples were cooled to liquid-nitrogen temperature to reduce the Doppler

broadening of the resonances. The average cross sections were derived by Derrien et al.[5] and corrected for the self-shielding effect.

2. The fission cross section of Weston and Todd[6] carried out at a 86.5 m flight path was analyzed.
3. The capture data used in the evaluation were obtained from the capture-to-fission ratio, α, of Weston.[7]

The data input for SAMMY was selected for two angular momenta, $l=0$ and $l=1$, that is s-wave and p-wave. The input is shown in Table 1. The parameters for $l=0$ are those from the statistical study of the resonance parameters in the resolved energy region 0 eV to 2.25 keV. The energies of the $^{235}$U low-lying levels were obtained from *Nuclear Data Sheets*[8], whereas the fission barrier values were obtained from Bjornholm and Lynn.[9]

Several iterations are needed to determine the best set of average parameters and background cross sections to describe the experimental data. The parameters obtained from the fit of the experimental data are shown in Table 2. The s-wave strength function shown in Table 2 is 2.8 % larger than the value of $(0.88 \pm 0.09)10^{-4}$ calculated in the resolved resonance region 0 eV to 110 eV. Before they were put into the ENDF format, SAMMY converted the parameters given in Table 2 into the ENDF average resonance parameters for use in calculation with the single-level Breit Wigner formalism. Table 3 displays the average resonance parameters for six reference energies for the total angular momentum *J*=3 with, respectively, two degrees of freedom for the neutron width distribution and three degrees of freedom for the fission width distribution.

**Table 1** Initial values for the parameters used in SAMMY calculations

| Input Parameters of s-wave | |
|---|---|
| Average Level Spacing | D = 0.446 ± 0.031 eV |
| Neutron Strength Function | $S_0 = (1.049 \pm 0.024)\ 10^{-4}$ |
| Capture Width | $\Gamma_\gamma = 38.14 \pm 1.70$ meV |
| Effective Scattering Radius | $R' = 9.602 \pm 0.050$ fm |
| Distant Level Parameter | $R^\infty = -0.155 \pm 0.006$ |
| $3^-$ channel Fission Width | $\Gamma_f = 213.0 \pm 20$ meV |
| $4^-$ channel Fission Width | $\Gamma_f = 146.5 \pm 15$ meV |
| **Input Parameters of p-wave** | |
| Neutron Strength Function | $S_1 = (1.76 \pm 0.25)\ 10^{-4}$ |
| Distant Level Parameter | $R^\infty = 0.12 \pm 0.050$ |
| Capture Width | $\Gamma_\gamma = 38.14 \pm 1.70$ meV |
| Fission Width (Channels $2^+$ to $5^+$) | $\Gamma_f = 200 \pm 100$ meV |

Comparisons of the average cross section calculated with SAMMY with the experimental data in the energy region 2.25 keV to 25 keV are given in Figure 1. The bottom curve is the total cross section, the middle curve is the capture cross section and the top curve represents the fission cross section.

**Table 2** Parameters of the SAMMY fit of the experimental data in the energy region 2.25 keV to 25 keV

| Angular Momentum | s-wave | | | p-wave | | |
|---|---|---|---|---|---|---|
| Neutron Strength Function $10^4 S_l$ | 0.905 ± 0.005 | | | 1.812 ± 0.021 | | |
| Average Capture Width meV | 36.06 ± 1.91 | | | 14.09 ± 2.11 | | |
| Distant Level Parameter $R^\infty$ | -0.153 ± 0.002 | | | 0.104 ± 0.004 | | |
| Effective Scattering Radius $R'$ fm | 9.680 ± 0.020 | | | 7.517 ± 0.211 | | |
| Fission Channel | $3^-$ | $4^-$ | $2^+$ | $3^+$ | $4^+$ | $5^+$ |
| Fission Width meV | 345 ± 15 | 208 ± 10 | 331± 17 | 78 ± 10 | 268 ± 21 | 229 ± 19 |

**Table 3** Average resonance parameters for *J*=3 (units are eV).

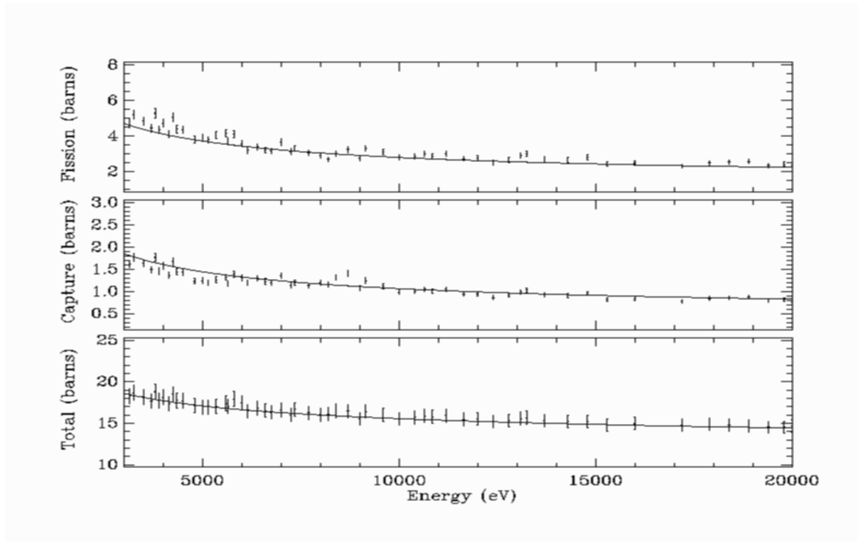
| $E_r$ | <*D*> | $<\Gamma_{n0}>\times 10^{-5}$ | $<\Gamma_\gamma>\times 10^{-2}$ | $<\Gamma_f>$ |
|---|---|---|---|---|
| 2250 | 0.98367 | 8.800 | 3.341 | 0.2338 |
| 2500 | 0.98317 | 8.789 | 3.341 | 0.2338 |
| 3500 | 0.98120 | 8.748 | 3.342 | 0.2338 |
| 4500 | 0.97924 | 8.710 | 3.344 | 0.2338 |
| 5500 | 0.97727 | 8.675 | 3.345 | 0.2338 |
| 6500 | 0.97531 | 8.641 | 3.347 | 0.2338 |

Table 3 presents the average resonance parameters, where $E_r$ is the reference energy, <*D*> is the average level spacing, $<\Gamma_{n0}>$ is the average reduced neutron width, $<\Gamma_\gamma>$ is the average radiation width, and $<\Gamma_f>$ is the average fission width. These quantities are given in units of electron volts.

## 4. Criticality Safety Application

The effect of the unresolved $^{235}U$ resonance evaluation on benchmark calculations was verified by performing calculations of benchmark systems with neutron spectra in the intermediate energy region. In particular, the benchmark systems $UH_3$, Zeus, and HISS/HUG, were used. Six experiments exist for the $UH_3$ system and three for the Zeus. The calculations were done for the $UH_3$ system (1) and for the three Zeus benchmarks, namely, Zeus (1), Zeus (2), and Zeus (3).

**Fig. 1.** Comparisons of average cross sections calculated with SAMMY with experimental data. Bottom curve is the total cross section, middle and upper curves are, respectively, capture and fission cross sections.

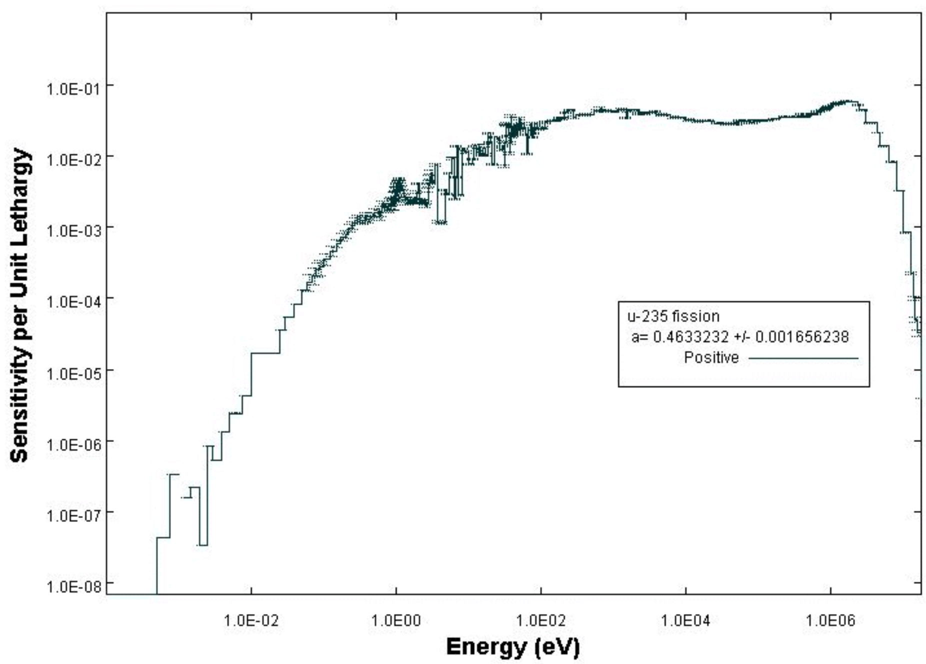



The change of the multiplication factor, $k_{eff}$, is well understood through the sensitivity coefficient which is defined as

$$S_x = \frac{dk_{eff} / k_{eff}}{d\Sigma_x / \Sigma_x} . \qquad \textbf{(10)}$$

Sensitivity calculations were done for the Zeus (1) and HISS/HUG benchmark systems. Figures 2 and 3 show the sensitivity coefficients $S_x$ of the fission cross section of $^{235}$U as a function of energy. The sensitivity calculations were done using the computer code TSUNAMI[10)] with the 238

**Fig. 2.** Sensitivity of $k_{eff}$ to the $^{235}$U fission cross section for the Zeus (1) benchmark.



neutron group structure of the SCALE system.[11)] Figures 2 and 3 show that a component of the $k_{eff}$ is due to the unresolved energy region.

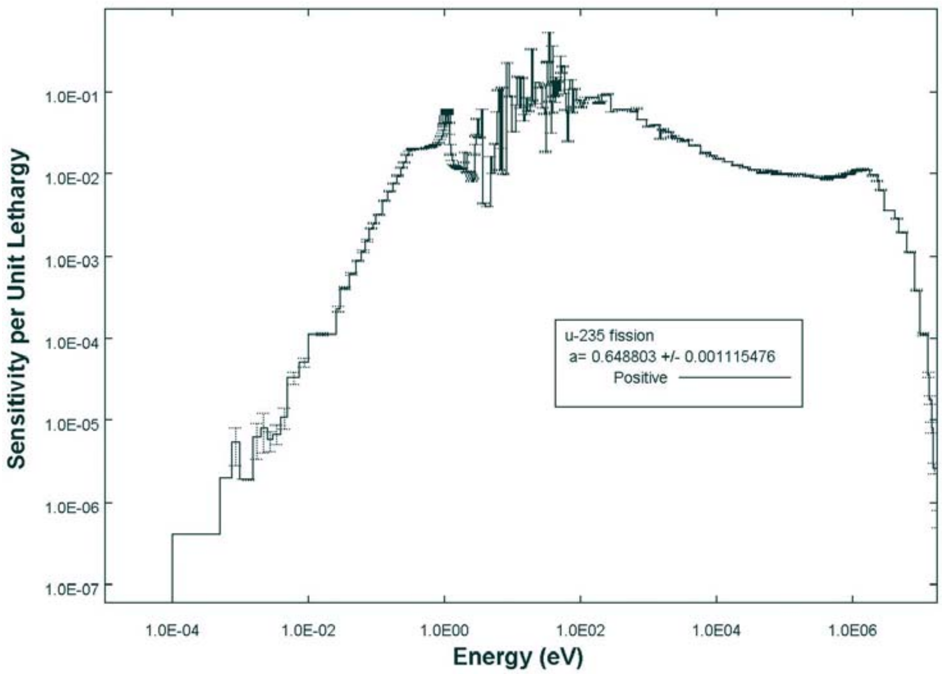



**Fig. 3** Sensitivity of $k_{eff}$ to the $^{235}$U fission cross section for the HISS/HUG benchmark.

For testing the unresolved $^{235}$U evaluation, benchmark calculations were done with MCNP4C with the recently issued nuclear data library ENDF66.[12] The $^{235}$U evaluation was processed with the ACER module of the NJOY code in a format suitable for use in the MCNP calculations and added to the ENDF66 library. In addition to the intermediate spectra benchmark, calculations were also done for one thermal and one fast benchmark. The results of the calculation with $^{235}$U in the ENDF66 library and the new evaluation are shown in Table 4.

**Table 4** Comparisons of $k_{eff}$ calculations using the unresolved $^{235}U$ evaluation.

| Benchmark | Experimental $k_{eff}$ | MCNP ENDF66 | MCNP ENDF66 with $^{235}U$ ORNL Evaluation |
|---|---|---|---|
| ORNL10 | 1.0015 ± 0.0010 | 0.9987 ± 0.0004 | 0.9991 ± 0.0004 |
| HISS/HUG | 1.0000 ± 0.0040 | 1.0099 ± 0.0005 | 1.0092 ± 0.0005 |
| $UH_3$ (1) | 1.0000 ± 0.0047 | 1.0040 ± 0.0050 | 1.0020 ± 0.0005 |
| Zeus (1) | 0.9976 ± 0.0008 | 0.9918 ± 0.0003 | 0.9899 ± 0.0003 |
| Zeus (2) | 0.9997 ± 0.0008 | 0.9945 ± 0.0003 | 0.9927 ± 0.0003 |
| Zeus (3) | 1.0010 ± 0.0009 | 0.9990 ± 0.0003 | 0.9965 ± 0.0003 |
| Godiva | 1.0000 ± 0.0010 | 0.9966 ± 0.0001 | 0.9964 ± 0.0001 |

## 5. Conclusion

Comparisons of the results between calculations done with MCNP using the $^{235}U$ evaluation in the ENDF66 and the ORNL evaluation do not display significant changes. MacFarlane *et al*[13)] have used these results as part of a new $^{235}U$ evaluation and find improved values for the effective multiplication factors. As mentioned above, in both evaluations the average resonance parameters are used for calculation of the cross section and self-shielding based on the use of single-level Breit-Wigner formalism. To improve self-shielding calculations in the unresolved energy region for fissile materials, it appears that one should use a cross-section formalism that can correctly represent the level-level interferences in the fission cross section. Work is under way to implement such a innovative cross-section formalism for the unresolved energy region.[14)]


## Acknowledgments

This work was sponsored by the Office of Environmental Management, U.S. Department of Energy, under contract DE-AC05-00OR22725 with UT-Battelle, LLC.


## References


1) L. C. Leal, H. Derrien, N. M. Larson, and R. Q. Wright, *Nucl. Sci. Eng.*, **131**, 230 (1999).

2) N. M. Larson, *Updated Users' Guide for SAMMY: Multilevel R-Matrix Fits to Neutron Data Using Bayes' Equations*, ORNL/TM-9179/R6 (July 2003).

3) F. H. Froehner,"Evaluation of the Unresolved Resonance Range of $^{235}U$," *Nucl. Sci.Eng.* **103**, 119, 128 (1989).

4) J. A. Harvey, N. W. Hill, F. G. Perey, G. L. Tweed, and L. C. Leal, *Proc. Int. Conf. on Nuclear Data for Science and Technology*, Mito, Japan (May 30-June 3, 1988).

5) H. Derrien, J. A. Harvey, N. M. Larson, L. C. Leal, and R. Q. Wright, *Neutron Total Cross Sections of $^{235}$U from Transmission Measurements in the Energy Range 2 keV to 300 keV and Statistical Model Analysis of the Data*, ORNL/TM-2000/129 (May 2000).

6) L. W. Weston, and J. H. Todd, *Nucl. Sci. Eng.,* **88**, 567 (1984).

7) L. W. Weston, G. de Saussure, and R. G. Gwin, *Nucl. Sci. Eng.,* **20**, 80 (1964).

8) *Nuclear Data Sheets*, **36**, 3 (1982).

9) S. Bjornholm and J. E. Lynn, *Rev. Mod. Phys.*, **52**, 4, 275 (1980).

10) B. L. Broadhead, C. M. Hopper, R. L. Childs, and C. V. Parks, *Sensitivity and Uncertainty Analyses Applied to Criticality Safety Validation*, NUREG/CR-6655, Vols.1 and 2 (ORNL/TM-13692/V1 and V2), U.S. Nuclear Regulatory Commission/Oak Ridge National Laboratory (1999). See also B. T. Rearden, "Perturbation Theory Eigenvalue Sensitivity Analysis With Monte Carlo Techniques," submitted to *Nuclear Science and Engineering*, and B. L. Broadhead, B. T. Rearden, C. M. Hopper, J. J. Wagschal, and C.V. Parks, "Sensitivity- and Uncertainty-Based Criticality Safety Validation Techniques," submitted to *Nuclear Science and Engineering*.

11) *SCALE: A Modular Code System for Performing Standardized Computer Analysis for Licensing Evaluatio*n, NUREG/CR-0200, Rev. 6 (ORNL/NUREG/ CSD-2/R6), Vols. I–III, (May 2000), available from the Radiation Safety Information Computational Center at Oak Ridge National Laboratory as CCC-545.

12) J. M. Campbell, S. C. Frankle, and R. C. Little, "ENDF66: A Continuous-Energy Neutron Data Library for MCNP4C,"p. 19 in *Proc. 12th Biennial Topl. Mtg. Radiation Protection and Shielding Div*., Santa Fe, New Mexico, April 2002, American Nuclear Society (2002).

13) R. MacFarlane, personal communication, January 2004.

14) A. Lukyanov, N. Koyumdjieva, N. Janeva, K. Volev, A. Brusegan, P. Schillebeeckx, G. Lobo, and F. Corvi, "Neutron Capture of $^{232}$Th in the Unresolved Resonance Region - Data Corrections and Analysis," *Nuclear Mathematical and Computational Sciences: A Century in Review—A Century Anew,* Gatlinburg, Tennessee, April 6-11, 2003, on CD-ROM, American Nuclear Society, LaGrange Park, IL (2003).