BNL-81625-2008-CP

Neutron Cross Section Uncertainties in the Thermal and Resonance Regions

S.F. Mughabghab and P. Oblozinsky

National Nuclear Data Center, Brookhaven National Laboratory, Upton, NY 11973-5000
(Dated: September 2, 2008)

Presented at the Covariance Workshop
Port Jefferson, New York,
June 24-27, 2008

National Nuclear Data Center
Energy Sciences & Technology Department
Brookhaven National Laboratory
P.O. Box 5000
Upton, NY 11973-5000
www.bnl.gov

Notice: This manuscript has been co-authored by employees of Brookhaven Science Associates, LLC under Contract No. DE-AC02-98CH10886 with the U.S. Department of Energy. The publisher by accepting the manuscript for publication acknowledges that the United States Government retains a non-exclusive, paid-up, irrevocable, world-wide license to publish or reproduce the published form of this manuscript, or allow others to do so, for United States Government purposes.

DISCLAIMER

This report was prepared as an account of work sponsored by an agency of the United States Government. Neither the United States Government nor any agency thereof, nor any of their employees, nor any of their contractors, subcontractors, or their employees, makes any warranty, express or implied, or assumes any legal liability or responsibility for the accuracy, completeness, or any third party's use or the results of such use of any information, apparatus, product, or process disclosed, or represents that its use would not infringe privately owned rights. Reference herein to any specific commercial product, process, or service by trade name, trademark, manufacturer, or otherwise, does not necessarily constitute or imply its endorsement, recommendation, or favoring by the United States Government or any agency thereof or its contractors or subcontractors. The views and opinions of authors expressed herein do not necessarily state or reflect those of the United States Government or any agency thereof.

Neutron Cross Section Uncertainties in the Thermal and Resonance Regions

S.F. Mughabghab* and P. Obložinský
National Nuclear Data Center, Brookhaven National Laboratory, Upton, NY 11973-5000
(Dated: September 2, 2008)

In the *Atlas of Neutron Resonances*, special care was expended to ensure that the resonance parameter information reproduces the various measured thermal cross sections, as well as the infinite dilute resonance integrals for Z = 1-100. In contrast, the uncertainties of the recommended quantities do not match those generated from the uncertainties of the resonance parameters. To address this problem, the present study was initiated to achieve consistency for 15 actinides and 21 structural and coolant moderator materials. This is realized by assigning uncertainties to the parameters of the negative-energy resonances and changing, if necessary, significantly the uncertainties of the low-lying positive-energy resonances. The influence of correlations between parameters on the derived uncertainties is examined and discussed.

I. INTRODUCTION

The National Nuclear Data Center produced a set of preliminary neutron covariance estimates for the international project, Nuclear Data Needs for Reactor Systems. The project is sponsored by the OECD Nuclear Energy Agency (NEA), Paris, under the Subgroup 26 of the International Working Party on Evaluation Cooperation (WPEC), chaired by M. Salvatores, ANL and CEA Cadarache. These preliminary covariances are described in the recent BNL report [1]. The project is interested in 53 materials (isotopes) which include 19 actinides and 34 structural, coolant and moderator materials. Out of them, the NNDC produced covariance estimates for 36 materials.

In the low-energy region, the NNDC used the method developed by BNL and LANL [2] that combines the recent information in the *Atlas of Neutron Resonances* [3] and the Bayesian code Kalman by Kawano and Shibata [4]. The idea of this approach is to use uncertainties of various parameters given in Ref. [3] and propagate them to cross section covariances. This can be done in two ways. First, one starts with cross sections and produces covariances straight in ENDF-6 file MF33 [1]. Second, as proposed in Ref. [5], one works purely with resonance parameters, produces covariances in MF32 and leaves up to processing codes the generation of cross section covariances.

As emphasized in [3], the absolute values of various recommended quantities, such as thermal capture, fission and scattering cross sections, as well as capture and fission resonance integrals, exhibit internal consistency with values calculated from the resonance parameters. However, no attempt was made to achieve consistencies between the various uncertainties of these quantities. Such inconsistencies would then propagate into the resulting cross section covariances rendering them less reliable.

In this study, consistencies between uncertainties of thermal cross sections and resonance parameters for capture and fission, but not scattering, cross sections were achieved for the following 36 nuclei: ^{19}F, ^{23}Na, ^{27}Al, ^{28}Si, ^{52}Cr, ^{55}Mn, ^{56,57}Fe, ^{58}Ni, ^{90,91,92,94}Zr, ^{166,167,168,170}Er, ^{206,207,208}Pb, ^{209}Bi, ^{233,234,236}U, ^{237}Np, ^{238,240,241,242}Pu, ^{241,242m,243}Am and ^{242,243,244,245}Cm.

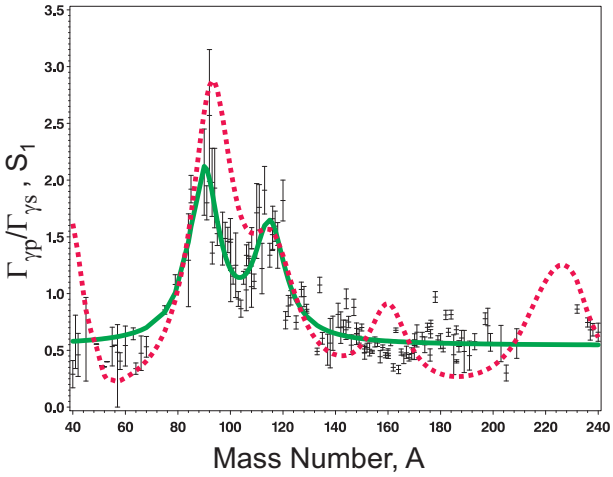

FIG. 1: Novel systematics for the average capture widths [6]. The ratio of the p-wave and s-wave radiative widths, displayed as a function of mass number, is designated by a solid green line. The p-wave strength function S_1, multiplied by 0.4×10^4, is shown by a broken red line.

II. METHODOLOGY

In this section, a brief account of the methodology and procedure considered in the analysis of covariances is presented.

- The starting point of the uncertainty analysis is an examination of the electronic file of the resonance parameters [3] in question to search for missing information.
- Where entries of the individual resonance parameters, such as radiative or scattering widths, are absent due to lack of measurements, then data based on the systematics described in [3] or other sources are supplied or estimated.
- In addition to the average resonance parameters and the systematics reported in [3], a recent detailed study [6]

*Corresponding author: mugabgab@bnl.gov

revealed for the first time a novel relationship between the average p- and s-wave radiative widths, as illustrated in Fig. 1. The observed structures for the ratio of the p-wave to s-wave radiative widths, $\Gamma_{\gamma p}/\Gamma_{\gamma s}$, at mass numbers 90 and 114 are associated with the splitting of the 3p single-particle state into its $3p_{1/2}$ and $3p_{3/2}$ components due to the nuclear spin-orbit interaction. Since these peaks are correlated with the p-wave strength function (Fig. 1), the p-wave radiative widths in the mass region 80-120 exhibit non-statistical effects in the neutron radiative capture mechanism. The background term for the ratio is interpreted in terms of the contribution of the giant dipole resonance to the capture process [6]. This significant information can be utilized in determining either the s- or p-wave radiative widths, when one is known with better accuracy than the other. Furthermore, for certain nuclei where information is lacking, scattering widths can be estimated from the reported measured capture kernels.

- The Atlas file is then converted into an ENDF-6 format with the aid of the computer program PTANAL [7]. In this procedure, average s- and p-wave radiative widths are supplied when such data is missing, and spin assignments of resonances of undetermined values are randomly made with the condition that the (2J+1) law for level density is followed.

- The utility code RECENT and a PSY-325 program are applied to determine the various contributions to the thermal cross sections and resonance integrals from individual resonances.

- The reported uncertainties of the thermal cross sections are attributed to the uncertainties of the bound levels and/or few low-energy resonances.

- Adjustments of the resonance parameter uncertainties are carried out iteratively until the calculated uncertainties of the thermal capture or fission cross sections converge to the measured ones.

The total uncertainty of the capture (fission) cross section in terms of the uncertainties of the individual resonance parameters, E_0, Γ_n, Γ_γ, and Γ_f, is expressed by Eq.(1) and Eq.(2), when correlations do not exist between the parameters; otherwise, additional terms, such as Eq.(3) and Eq.(4), have to be considered.

Since a large number of resonance uncertainties contribute to the scattering uncertainty, only the capture and fission uncertainties were treated in this study; for an explanation, refer to Eq.(1.9) and Eq.(1.14) in [3].

At the start, the correlations between the parameters were not considered; their effects are studied later on for some nuclei. Also in this study, the Breit-Wigner relations for capture and fission are utilized for the determination of the partial derivatives in Eqs. (1-4).

$$(\Delta\sigma_\gamma)^2 = \left(\frac{\partial\sigma_\gamma}{\partial E_0}\Delta E_0\right)^2 + \left(\frac{\partial\sigma_\gamma}{\partial\Gamma_n}\Delta\Gamma_n\right)^2 + \left(\frac{\partial\sigma_\gamma}{\partial\Gamma_\gamma}\Delta\Gamma_\gamma\right)^2 + \left(\frac{\partial\sigma_\gamma}{\partial\Gamma_f}\Delta\Gamma_f\right)^2 \quad (1)$$

$$(\Delta\sigma_f)^2 = \left(\frac{\partial\sigma_f}{\partial E_0}\Delta E_0\right)^2 + \left(\frac{\partial\sigma_f}{\partial\Gamma_n}\Delta\Gamma_n\right)^2 + \left(\frac{\partial\sigma_f}{\partial\Gamma_\gamma}\Delta\Gamma_\gamma\right)^2 + \left(\frac{\partial\sigma_f}{\partial\Gamma_f}\Delta\Gamma_f\right)^2 . \quad (2)$$

The calculated uncertainties for the thermal capture and fission cross sections due to all the individual s-wave resonances, reported in [3], are determined by these relations and then combined in quadratures to obtain the total uncertainty.

When correlations do exist between the various resonance parameters, such as between Γ_γ and Γ_n or Γ_f and Γ_γ, then the following additional terms, Eqs. (3)-(4) have to be added to Eqs. (1-2)

$$(\Gamma_\gamma,\Gamma_n) = 2\left(\frac{\partial\sigma_\gamma}{\partial\Gamma_n}\Delta\Gamma_n\right)\left(\frac{\partial\sigma_\gamma}{\partial\Gamma_\gamma}\Delta\Gamma_\gamma\right)\rho(\Gamma_\gamma,\Gamma_n)\,, \quad (3)$$

$$(\Gamma_f,\Gamma_\gamma) = 2\left(\frac{\partial\sigma_f}{\partial\Gamma_f}\Delta\Gamma_f\right)\left(\frac{\partial\sigma_f}{\partial\Gamma_\gamma}\Delta\Gamma_\gamma\right)\rho(\Gamma_f,\Gamma_\gamma)\,, \quad (4)$$

where $\rho(\Gamma_\gamma,\Gamma_n)$ and $\rho(\Gamma_f,\Gamma_\gamma)$ are the correlation coefficients between Γ_γ and Γ_n or between Γ_f and Γ_γ, respectively. The origin of these types of correlations is discussed briefly in the following section.

III. SOME SOURCES OF CORRELATIONS

In this section, a brief discussion of the sources of correlations between the various parameters, emanating from the analysis of the measurements, is presented.

- In capture measurements when the experimental resolution function is larger than the natural width of a resonance, area analysis yields a capture kernel, defined by $A_\gamma = g\Gamma_n\Gamma_\gamma/\Gamma$, where g is the statistical spin factor. If $A_\gamma < g\Gamma_\gamma$, then a scattering width can be evaluated from this relation by assuming an average capture width, derived from the systematics [3, 6], and a

spin value for the considered resonance. In this case, $\rho(\Gamma_n, \Gamma_\gamma)$ is negative.

- In combined capture and fission measurements, capture and fission kernels are determined, in which case Γ_f/Γ_γ is found. The fission widths are subsequently derived on the basis of an assumed average capture width, as, for example, in the case of the isotopic curium measurements of [8]. In this case $\rho(\Gamma_f, \Gamma_\gamma)$ is positive.

- Neutron sensitivity corrections, applied to capture measurements, are generally made with the help of the relation

$$\Gamma_\gamma^{\rm exp} = \Gamma_\gamma + k\Gamma_n \,,$$

where k is obtained experimentally. If k is not determined correctly, then $\rho(\Gamma_n, \Gamma_\gamma)$ can be positive. A Γ_γ-Γ_n correlation analysis, as well as theoretical calculations [9], has to be performed to determine as to whether the correlation exists. If affirmed, a determination has to be made as to whether it is due to neutron sensitivity or valence capture [9].

- In determining the parameters of bound levels, for nuclei where the positive-energy resonances also contribute significantly to the thermal capture cross section, then a negative correlation exists between the parameters of the positive-energy and bound resonances. Relations similar to those of Eqs. (3-4) can be employed to take into account their effects on the uncertainties.

IV. RESULTS

Our experience in the adjustment procedure is presented and the outstanding issues resulting from the present analysis are discussed for the following nuclei: ^{55}Mn, ^{56}Fe, and ^{167}Er.

A. ^{55}Mn

Adopted as a standard in neutron activation analysis, the thermal capture cross section of ^{55}Mn, σ_γ^0 = 13.36 $\pm$ 0.05 b (0.37%), is measured with high accuracy. As noted in a footnote in the Atlas [3], the adoption of the capture widths measured at ORNL [10], which were the only available data at the time, leads to a calculated resonance capture integral of 11.7 b. This value is discrepant with a measured value of 13.4$\pm$0.5 b [3]. To resolve this discrepancy, adjustments in the capture widths of [10] were made; the results are shown in column 5 of Table I.

In addition, two bound s-wave levels with spins 2 and 3 are invoked in order to account for the coherent and incoherent scattering amplitudes of ^{55}Mn. The contributions to the thermal capture cross section due to the bound levels, as well as the 3 positive-energy resonances, are presented in the last column of Table I. With these parameters and those in [3] above 3 keV, the calculated capture cross section in the energy region from 0.01 eV to 20 keV is displayed in Fig. 2 and is compared with the measurements of [11] below 300 eV.

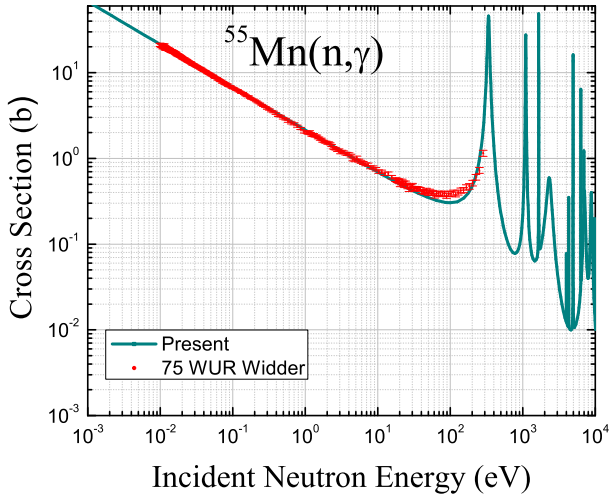

FIG. 2: The measured capture cross section of ^{55}Mn in the energy region 0.01 to 300 eV. The solid line represents the present calculations based on the resonance parameters of Table I and in Ref. [3] above 2.4 keV.

TABLE I: ^{55}Mn resonances and their contribution to the thermal capture cross section. In the present notation, a = data from the Atlas [3], and b = quantities evaluated in the present study. The energy uncertainty of the 337.3 eV resonance is altered from 1.0 eV [3] to 0.7 eV.

E_0 (eV)^a	$2g\Gamma_n$ (eV)^a	$2g\Gamma_n$ (eV)^b	Γ_γ(eV)^a	Γ_γ (eV)^b	σ_γ (b)^b
-16150	6255	6255	.75 $\pm$.0%	.79$\pm$.0	0.32
-202	1.15	1.15	.75 $\pm$.0%	.79$\pm$.0	3.38
337.3 $\pm$0.7	18.3 $\pm$2.2%	18.3$\pm$.2%	.31 $\pm$6.5%	.40$\pm$.0	7.44
1099 $\pm$ 2.0	18.0 $\pm$4.0%	18.0 $\pm$.4%	.435 $\pm$23.0%	.40$\pm$.0	1.50
2327 $\pm$ 5.0	460 $\pm$5.2%	460 $\pm$.2%	.34 $\pm$38.2%	.40$\pm$.0	0.17

To account for a 0.37% uncertainty in the thermal capture cross section, the uncertainties of the resonance parameters have to be drastically reduced. It is significant to note that consistency between the two uncertainties can be achieved by changing the energy uncertainty of the resonance at 337.1 eV from 1.0 eV to 0.7 eV.

This procedure presents an outstanding issue. To overcome this problem, one can invoke an anti-correlation between the parameters of the positive-energy and bound levels, which can alleviate this problem. Such a procedure entailed imposing large uncertainties on the parameters of the bound parameters while retaining, with minor adjustments, the uncertainties of the positive energy resonances as reported in [3]. The latter procedure is justifiable on physical grounds. For details, refer to [12].

B. ^{56}Fe

In this simple case, the thermal capture cross section section of ^{56}Fe, σ_γ^0 = 2.59±0.14 b, is dominated by a bound level, specified at an energy of -6.52 keV. The positive-energy resonance contributions due to all resonances up to an energy of 850 keV is only 0.09 b [3]. The pointwise capture cross section in the energy region 0.007 eV to 1.41 keV was measured by [13]. The computed and measured cross sections are depicted in Fig. 3. Note that the resonance at 1.147 keV has a p-wave assignment, and hence does not have a contribution to the thermal capture cross section of ^{56}Fe.

The 5.4% uncertainty of the thermal capture cross section can be propagated by assigning a 3.82% uncertainty to both the scattering and capture widths of the bound level only. The positive-energy resonances then maintain their original uncertainties [3]. In this case, it is not necessary to invoke an anti-correlation between the bound and positive energy resonances.

At this point, we point out the similarity of the capture cross sections of ^{52}Cr and ^{56}Fe in that both are dominated by bound levels; refer to the discussion in section V.

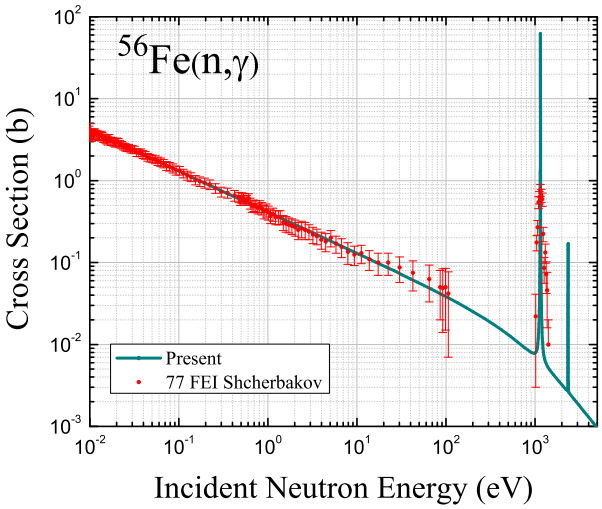

FIG. 3: The measured capture cross of ^{56}Fe in the energy region 0.01-3000 eV [13]. The solid line represents the calculations on the basis of the resonance parameters of the Atlas [3].

C. ^{167}Er

The thermal capture cross section of ^{167}Er, σ_γ^0 = 649±8 b(1.2%) is dominated by two positive-energy resonances at 0.460 eV and 0.584 eV. The scattering and capture widths of these two resonances are determined recently [14] by applying shape fit analysis to the transmission data, obtained for a natural Er sample in the energy region from 0.03-20 eV. To propagate a 1.2% uncertainty for σ_γ^0, the uncertainties of the capture widths of the bound level and the resonance at 0.460 eV were changed as shown in column 3 of Table II. The uncertainties of the scattering widths were maintained at the Atlas values.

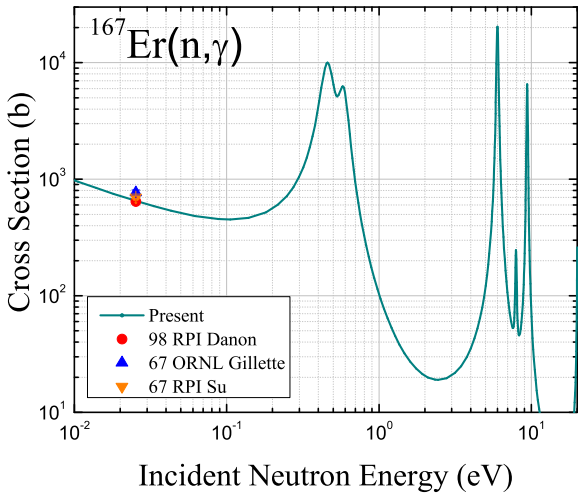

FIG. 4: The measured capture cross of ^{167}Er in the energy region from 0.001 to 10 eV. The solid line represents the calculations on the basis of the resonance parameters of the Atlas [3].

TABLE II: ^{167}Er resonances and their contribution to the thermal capture cross section. The two quantities in parentheses in columns 2 and 3 are the % uncertainties in the Atlas and the present study, respectively.

E_0 (eV)	$2g\Gamma_n$ (meV)	Γ_γ(meV)	σ_γ (b)
-23.6	117 (0.0, 10.0)%	88.0 (0.0, 11.0)%	7.9
0.460 ±0.002	0.3031 (0.33, 0.33)%	87.12 (0.2, 0.4)%	423
0.584 ±0.001	0.2163 (0.46, 0.46)%	86.20 (0.4, 0.4)%	161

V. CORRELATION EFFECTS ON CALCULATED UNCERTAINTIES

A correlation analysis between capture and scattering widths for the following nuclei ^{52}Cr, ^{55}Mn, ^{56}Fe, ^{167}Er and ^{237}Np was carried out. The correlation coefficients are, respectively, 0.63±0.09 (8 s-wave resonances), 0.700±0.001 (49), 0.20±0.58 (10), 0.35±0.01 (54), 0.12±0.12 (147). Note that the significance levels for ^{52}Cr and ^{56}Fe are high and null, respectively. For this reason, ^{56}Fe is replaced by ^{52}Cr in this analysis. In addition, we believe that the high significance level of the correlation coefficient for ^{55}Mn is due to neutron sensitivity correction and not arising from valence capture.

The effects of the correlation between the capture and scat-

TABLE III: Effects of correlations between Γ_γ and Γ_n on the thermal capture cross section uncertainties of ^{52}Cr, ^{55}Mn, and ^{237}Np for three values of the correlation coefficients, 0.0, -1.0, and 1.0.

nucleus	σ_γ (b)	$\Delta\sigma_\gamma$ (b)	$\Delta\sigma_\gamma$ (b)	$\Delta\sigma_\gamma$ (b)
$\rho(\Gamma_\gamma, \Gamma_n)$		0.0	-1.0	+1.0
^{52}Cr	0.86±0.02	0.020	0.018	0.023
^{55}Mn	13.36±0.05	0.047	0.047	0.049
^{237}Np	178.7±3.0	3.0	1.6	4.0

tering widths of the same resonance on the capture uncertainties for three nuclei, ^{52}Cr, ^{55}Mn, and ^{237}Np, were studied and summarized in Table III. The case for $\rho(\Gamma_\gamma, \Gamma_n) = 0$ corresponds to the results of the main study with no correlations, where the calculated uncertainties reproduce those of the recommendations [3]. In columns 4 and 5 of Table III are the results for $\rho(\Gamma_\gamma, \Gamma_n)$= -1 or +1. As shown, the correlations produce negligible effects for ^{52}Cr, ^{55}Mn. This is a result of the fact that the final adjusted uncertainties for the capture widths are set to zero; refer to Eq.(3). On the other hand, a non-negligible change occurs in the uncertainties of ^{237}Np.

VI. CONCLUSIONS

Internal consistency between the uncertainties of thermal capture and fission cross sections and the uncertainties of resonance parameters is achieved for 15 actinides and 21 coolant and structural materials. This was realized by re-assigning uncertainties to the parameters of bound levels and low-energy resonances. If the major contribution to the thermal capture cross section is due to the positive-energy resonances, then their uncertainties are significantly changed from values reported in [3]; this pauses certain challenge as to what extent these changes are physically justifiable. Such a situation can be resolved by invoking an anti-correlation between the bound and positive energy resonances, as determined for ^{55}Mn [12].

In other cases, where the thermal capture cross section is dominated by bound levels, as in ^{52}Cr and ^{56}Fe, the uncertainties of the positive-energy resonances are unaffected from those reported in [3]. The correlations between parameters of the same resonance are studied and their effect on the adjusted uncertainties is determined for some cases

Acknowledgments

The support of B. Pritychenko with LaTex is gratefully acknowledged. Work at Brookhaven National Laboratory was sponsored by the Office of Nuclear Physics, Office of Science of the U.S. Department of Energy under Contract No. DE-AC02-98CH10886 with Brookhaven Science Associates, LLC.

[1] D. Rochman, M. Herman, P. Obložinský and S. F. Mughabghab, *Preliminary Cross Sections and Nubar Covariances for WPEC Subgroup 26*, Report BNL-77407- 2007-IR, January 2007.

[2] M. B. Chadwick, P. Obložinský, M. Herman et al., *ENDF/B-VII.0: Next Generation of Evaluated Nuclear Data Library for Nuclear Science and Applications*, NUCLEAR DATA SHEETS, **107**, 2931-3060 (2006).

[3] S. F. Mughabghab, ATLAS OF NEUTRON RESONANCES: RESONANCE PARAMETERS AND THERMAL CROSS SECTIONS Z=1-100, Elsevier, Amsterdam 2006.

[4] T. Kawano and K. Shibata , *Covariance Evaluation System* , in Japanese, JAERI Report JAERI- Data/Code 97-037(1997).

[5] M. Herman, S. F. Mughabghab, P. Obložinský, M. T. Pigni and D. Rochman, *Neutron Cross Section Covariances in the Resolved Resonance Region*, Report BNL-80173-2008, April 2008.

[6] S. F. Mughabghab, *Spin-Orbit Splitting of the 3P Single Particle State: Non-statistical Effects in Radiative Neutron Capture*, Report BNL-80311-2008-JA, July 2008; submitted for publication.

[7] S. Oh, J. Chang and S. F. Mughabghab, *Neutron Cross Section Evaluations of Fission Products Below the Fast Energy Region*, Report BNL-NCS-67469, April 2000.

[8] M. Moore and J. Keyworth, *Analysis of the Fission and Capture Cross Sections of the Curium Isotopes*, Phys. Rev. C 3, 1656 (1971).

[9] A. M. Lane and S. F. Mughabghab, *Relation between the Optical Model and the Valence Model of E1 Neutron Capture at and between Resonances*, Phys. Rev. C 10, 412 (1974).

[10] R. L. Macklin, *Resonance Neutron Capture by Manganese below 2.5 keV*, Nucl. Sci. Eng. 89, 362 (1985).

[11] F. Widder, *Neutron Capture Cross-Section Measurements from 0.01 to 10 Electron Volts*, Second Neutron Capture Gamma-Ray Spectroscopy and Related Topics, p. 757 Petten, The Netherlands, Sept 2-6, 1974.

[12] M. Herman, M.T. Pigni, P. Obložinský et al., *Development of Covariance Capabilities in EMPIRE Code*, this issue of Nuclear Data Sheets.

[13] A. Scherbakov, and A. N. Glukhovec, *Slow Neutron Total and Radiative Capture Cross Section of* ^{56}Fe, Report YFI-24, 3 (1977).

[14] Y. Danon et al., *Neutron Total Cross Section Measurements and Resonance Analysis of Holmium, Thulium and Erbium from 0.001 to 20 eV*, Nucl. Sci. Eng. 128, 61 (1998).