RESONANCE REGION CROSS-SECTION DATA ADVANCEMENTS FOR NUCLEAR CRITICALITY SAFETY APPLICATIONS AND ENDF/B-VII.1[1]

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ABSTRACT

ORNL has completed new resonance region cross-section evaluations with covariance data to support nuclear criticality safety applications. This paper summarizes some of the most recent cross-section evaluations that have been completed and will be made available to process into nuclear data libraries for use with radiation transport codes. Specifically, the paper summarizes the work that has been completed for ^{35}Cl, ^{37}Cl , ^{50}Cr, ^{52}Cr, ^{53}Cr, ^{54}Cr, ^{39}K, ^{41}K, ^{19}F, ^{55}Mn, ^{58}Ni, ^{60}Ni, ^{235}U, ^{238}U, and ^{239}Pu. Many of the new cross-section evaluations are based on new differential data measurements in the resonance region. Furthermore, corresponding covariance data analyses have been performed in conjunction with the resonance evaluation effort thereby providing a consistent set of covariance data to complement the resonance region cross-section evaluations. As a result, the new cross-section evaluations with covariance data can be used to support sensitivity/uncertainty analyses for criticality safety applications.

Key Words: Resonance Region, Neutron Cross-Section Data, Covariance Data, Nuclear Data

1 INTRODUCTION

For more than 30 years, the Oak Ridge National Laboratory (ORNL) has performed research and development to provide accurate nuclear cross-section data in the resonance region. Since the mid 1990s, many of the resonance region advances have been driven by nuclear criticality safety needs in the intermediate energy range, and the resonance region nuclear data work at ORNL has been performed in support of the U.S. Nuclear Criticality Safety Program (NCSP) [1]. At ORNL, the nuclear data research and development effort provides support for the NCSP in four complementary areas of research: (1) differential cross-section measurements, (2) resonance analysis methods development with the SAMMY [2] R-matrix analysis software, (3) cross-section evaluation development, and (4) cross-section processing methods development with the AMPX [3] software system. Overall, the nuclear data work effort is tightly coupled with nuclear criticality safety analysis efforts and SCALE [4] radiation transport methods development efforts at ORNL. As a result, resonance region measurements and evaluations are performed in concert with nuclear criticality safety application needs and requirements. For example, assessments of previous nuclear data measurements and evaluations have revealed deficiencies in nuclear data (e.g., missing resonances, high neutron sensitivity, etc.) that are important for criticality safety applications. As a result, new measurements and evaluations have been performed to address key nuclear data deficiencies, and in the past five years, ORNL has submitted more than 20 new cross-section and/or covariance evaluations (e.g., ^{235}U, ^{238}U, ^{239}Pu, ^{55}Mn, ^{58}Ni, ^{60}Ni, etc.) for distribution as part of the

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[2]Retired.

[3]Deceased.

U.S. Evaluated Nuclear Data File (ENDF/B) database [5]. Many of the new ORNL cross-section evaluations will be distributed with ENDF/B-VII.1, which is scheduled for release from the U.S. National Nuclear Data Center (NNDC) in December 2011. The objective of this paper is to document the new resonance region cross-section evaluations with covariance data that have been developed to support criticality safety applications.

2 U.S. NUCLEAR CRITICALITY SAFETY PROGRAM

Within the U.S. NCSP, the nuclear data evaluation work effort is performed to support the end user and provide improved nuclear cross-section data to address criticality safety application needs. Moreover, the Nuclear Data Advisory Group (NDAG) provides technical support to the NCSP in the area of nuclear data. The NDAG reviews the nuclear data needs that have been identified by the end-user for criticality safety applications within the U.S. Department of Energy (DOE) Complex. Based on the NDAG assessment of the data need, the NDAG determines if new cross-section measurements and/or evaluations are needed to address the nuclear data need. If additional work is required, the NDAG prioritizes and schedules the nuclear data measurements and evaluation effort that is needed to address the nuclear data need. As noted previously, ORNL has submitted more than 20 new cross-section and/or covariance evaluations for release as new ENDF/B evaluations, and each of these nuclear data evaluations was performed in accordance with the NCSP Nuclear Data work plan. ORNL has extensive experience performing nuclear data measurements and evaluations in the resonance region. Likewise, the Los Alamos National Laboratory (LANL) has extensive experience performing cross-section measurements and evaluations at energies above the resonance region (i.e., “high-energy” region). Based on the complementary capabilities, ORNL and LANL work together within the NCSP to complete full-energy-range cross-section evaluations with covariance data for supporting nuclear criticality safety applications. The work presented in this paper is focused on the resonance region nuclear data measurement and evaluation efforts that have been performed at ORNL in support of the NCSP.

3 ORNL NUCLEAR DATA EVALUATIONS

In the following sections, additional details are provided for the select, new cross-section evaluations that have been submitted for release with ENDF/B-VII.1. It should be noted that ORNL has also completed new cross-section measurements and a corresponding nuclear data evaluation for ^{48}Ti, and the details for the ^{48}Ti evaluation work are presented in a companion paper [6] to this paper at the conference. As a result, the details concerning the ^{48}Ti evaluation work are not presented in this paper.

3.1 ^{35}Cl and ^{37}Cl Evaluations

Chlorine is a key neutron absorber in materials that are encountered in various industrial applications (e.g., PVC piping/containers, chemical processes, etc.). Therefore, accurate chlorine cross-section data are needed to support radiation transport analyses for nuclear criticality safety. As part of the work for the NCSP, neutron resonance evaluations were performed using the SAMMY R-matrix analysis software for ^{35}Cl and ^{37}Cl in the energy region from the thermal region up to 1.2 MeV [7]. In conjunction with the evaluation effort, ORNL developed a new resonance parameter format that expands upon the existing Reich-Moore resonance format currently in the ENDF/B files. The new format, known as the R-Matrix Limited Format, includes the charged particle exit channels that are needed for the ^{35}Cl evaluation. For the ^{37}Cl evaluation, the conventional three-channel Reich-Moore formalism was used. In addition, the new evaluations for both ^{35}Cl and ^{37}Cl also include resonance parameter covariance matrices that correspond to the new resonance parameter evaluations. The Cl evaluations were based on fits of many data sets with the SAMMY software. A detailed discussion of the analysis methods used to determine parameter values and uncertainties is given in [7]. For example, for capture and neutron width uncertainties,

for each resonance, several SAMMY calculations with different width values were overlaid with the data. Both the overlay plots and χ^2 changes with width variation were used to determine final uncertainties that were, in most cases, significantly larger than the SAMMY values. At the time of the Cl evaluation, SAMMY did not incorporate the now available "Prior Uncertainty Parameter," or PUP procedure. Thus, some normalization and background uncertainties were not propagated properly through the sequential analysis of multiple data sets. Although uncertainties in resonance energies and widths are judged to be realistic, the uncertainties in computed cross sections in valleys between resonances were underestimated. Furthermore, the resonance parameter format at the time of the evaluation was limited to resonance parameter uncertainties. As a result, the evaluations do not include correlations and uncertainties in nuclear radii. The "normalization/background/radius" effects were represented approximately by adjusting the resonance parameter uncertainties for the external resonance parameters and for select resonances in the energy range of the evaluation. Since the resonance parameter representation does not include the Direct Capture (DC) part of the capture cross-section data, the DC component was included as a "background" 1/v cross-section in the continuous-energy part of the ENDF/B evaluation. At thermal energy (E = 0.0253 eV), the ^{35}Cl DC cross section is 0.16 b, which is a small fraction of the overall capture cross-section value of 43.60 b. For ^{37}Cl, the DC cross section is 0.31 b, which is a large fraction of the overall capture cross-section value of 0.433 b. The DC cross section was computed below 10 (100) keV for ^{35}Cl (^{37}Cl). The 1/v cross section was extended to 1.0 MeV to ensure continuity in the evaluation range. In summary, the new resonance parameter evaluations for ^{35}Cl and ^{37}Cl are based on state-of-the-art resonance analysis methods and include consistent covariance data that are needed to support criticality safety analyses.

3.2 ^{50}Cr, ^{52}Cr, ^{53}Cr, and ^{54}Cr Evaluations

Resolved resonance parameter evaluations for chromium isotopes [8], namely ^{50}Cr, ^{52}Cr, ^{53}Cr, and ^{54}Cr, were performed with SAMMY. The determination of the resolved resonance parameters for the chromium isotopes below 100 keV relied mainly on the recent ORELA measurements. The ORELA capture cross-section data were essential to supporting a new evaluation in the resonance region because capture cross-section data measurements were not available in the literature prior to this work. In addition to the new ORELA measurements, existing high-resolution transmission data for enriched nuclides were also included in the evaluation. Thermal cross-section data available in the EXFOR data system were also included in the evaluation. The resonance regions for each chromium isotope were extended by taking advantage of the high resolution in the new ORELA data and existing transmission data. Comparisons of the SAMMY R-matrix fits to the data of Guber [9] for natural chromium up to 100 keV are shown in Fig. 1. The top curve represents the capture cross-section data whereas the two other curves are the total cross sections corresponding to transmission with sample thickness of 0.026269 at/b (thin) and 0.053103 at/b (thick), respectively. Below 100 keV the evaluation relied on the transmission and capture data measurements done by Guber et al. at ORELA [9]. As part of the SAMMY analysis, resonance parameter covariance matrices (RPCMs) were generated for each chromium isotope. In the SAMMY fit of the experimental data systematic uncertainties such as data normalization, background, etc. were propagated into the final RPCM. As a result, a consistent set of covariance matrices were generated with each new chromium isotopic resonance parameter evaluation.

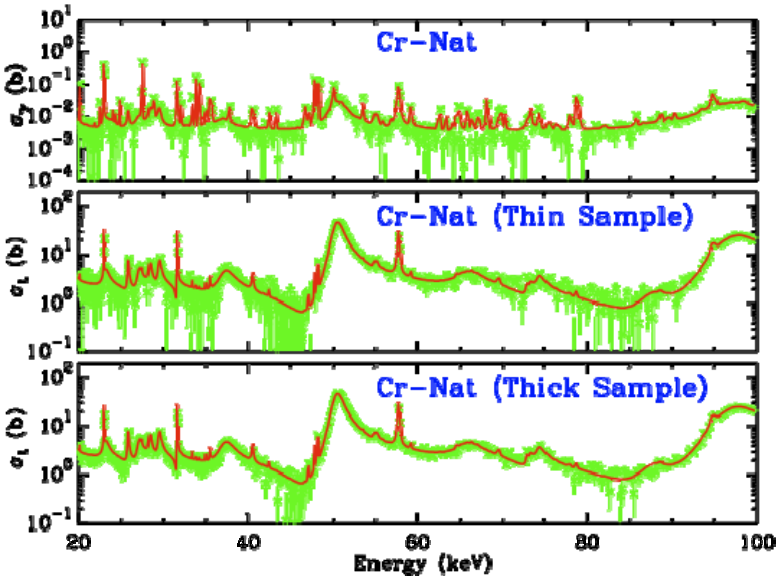

Figure 1. Comparison of SAMMY calculations obtained with the resonance parameters (solid line) with the measured cross-section data for total and capture cross sections between 20 keV and 100 keV.

3.3 ^{39}K and ^{41}K Evaluations

ORNL has completed new evaluations for ^{39}K and ^{41}K neutron cross section data in the resolved resonance region using the Reich-Moore R-matrix formalism in SAMMY. The evaluation incorporates recent high-resolution capture and transmission measurements made at ORELA to extend the resolved resonance energy range up to 1.0 MeV with much more accurate representation of the data than previous evaluations available in ENDF/B-VI.8. The data include transmission measurements by Guber et al. [10] and Harvey et al. [11] on the 80-m flight path at ORELA, total cross section data of Cierjacks et al. [12] on a 57-m flight path performed at the Karlsruhe Isochronous Cyclotron, and measurements of Singh et al. [13] done at the 200-m flight path at the Columbia synchrocyclotron. Also included in the evaluation were the high-resolution capture cross section of Guber et al. [9] measured in the energy range of 0.1 keV to 600 keV, and an older low-resolution capture data of Joki et al. [14] in the energy region from 0.02 eV to 10 eV. The new evaluations include the resonance parameters along with the corresponding RPCMs that were obtained during the SAMMY analysis of the measured data.

3.4 ^{19}F Evaluation

The resonance parameters for ^{19}F were obtained from a sequential SAMMY analysis of the experimental neutron transmission data, capture cross sections, and inelastic cross-section data in the resolved energy region up to 1 MeV [15]. This evaluation is the first in the resolved resonance region that includes the inelastic cross-section channels that open in the resonance region and makes use of the R-Matrix Limited formalism. Three transmission data sets, one capture cross-section data set, and three inelastic cross-section data sets were used in the ^{19}F analysis. The evaluation was performed up to 1 MeV with 2 s-wave, 5 p-wave, 17 d-wave, and 7 f-wave resonances. In the energy range up to 1 MeV, ^{19}F has two inelastic channels starting at the energies 109.9 keV with spin 1/2- and 197.2 keV with spin 5/2+. These two inelastic channels were included in the SAMMY evaluation. The set of experimental data used in the evaluation consisted of three transmission measurements of Larson [16] with thicknesses of 0.13093 at/b, 0.016886 at/b, and 0.024184 at/b, respectively, measured on the 80-m flight path at ORELA from 0.2 to 20 MeV. In addition, the evaluation included one capture cross-section measurement by Guber [17] at ORELA up to 700 keV and three inelastic cross-section measurements by Broder [18] up to 1 MeV. For the thermal cross section, ORNL used the total cross-section data of Rainwater [19] in the energy region from

thermal up to 5 eV. Rainwater total cross-section data were in agreement with the effective total cross section derived from the ORNL thick sample transmission data. No data normalization was needed for the Rainwater total cross section. With exception to the capture cross-section measurement of Raman et al. [20] at thermal energy, there is no other thermal experimental capture cross-section data available in the literature. To fit the capture cross-section data at low energy, the ENDF/B-VII ^{19}F capture cross-section evaluation, normalized to the Raman thermal data, was used in the latest evaluation effort. A comparison of the transmission data of Larson with the results of the SAMMY calculations up to 200 keV, using the resonance parameters obtained in the analysis, is shown in Fig. 2. In addition to the new resonance parameter evaluation, covariance data for the resonance parameters were generated in the resolved energy region as part of the resonance evaluation. The experimental data uncertainties from the cross-section measurements were propagated with SAMMY to generate the RPCM.

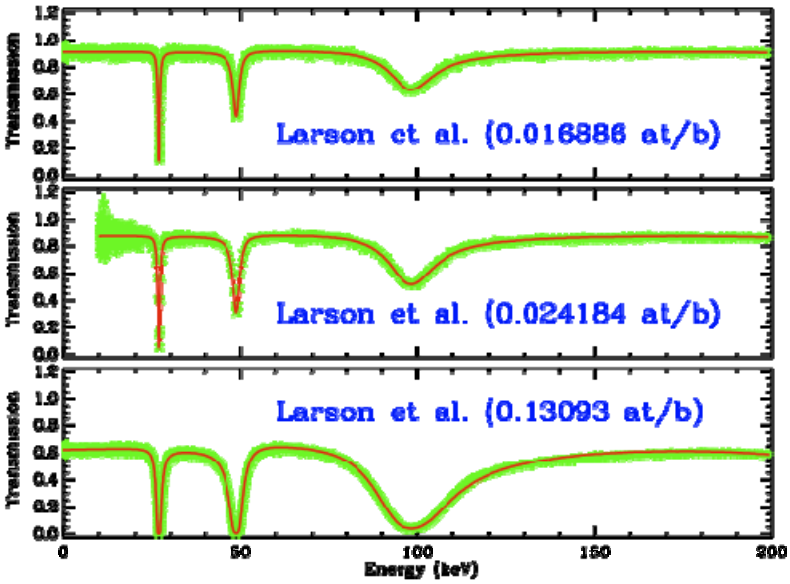

Figure 2. SAMMY fit to ^{19}F transmission data in the energy region from 10 eV to 200 keV.

3.5 ^{55}Mn Evaluation

Accurate neutron capture cross sections of ^{55}Mn are important for criticality safety applications because of its use in structural materials. Resonance parameters for ^{55}Mn in the previous evaluated data files are mainly based on the work performed by Garg et al. [21] and by Macklin [22]. The target accuracy of the previous work by Macklin was 10% in the lowest energy resonances and 20% accuracy up to 100 keV. Measurements done by Perrot et al. [23] using a lead slowing-down spectrometer and simulation have shown that the ^{55}Mn-evaluated capture cross sections are inadequate in the energy range 50 eV to 30 keV. Within the NCSP, a new ^{55}Mn evaluation was needed to improve the performance of the capture cross-section evaluation in systems that are sensitive to ^{55}Mn as a structural material. As a result, a new resonance evaluation of ^{55}Mn was performed using a SAMMY analysis of recent experimental transmission and capture data measured at the ORELA and GELINA facilities. The ^{55}Mn evaluation was performed in attempt to obtain more accurate resonance parameters in the thermal energy range to 125 keV, below the first inelastic channel. The experimental database includes the 1988 neutron transmission of Harvey et al. [24], the 2005 GELINA capture cross-section measurement of Schillebeeckx et al. [25], and the ORELA capture measurement of Guber [26]. The experimental resolution of the capture data allowed a good separation of the resonances up to ~120 keV neutron energy. For the evaluation of the cross sections in the thermal energy range, earlier measurements were added to the experiment data base: total cross section from Cote et al. [27] and Rainwater et al. [28] and capture cross

section from Widder et al. [29]. The results of the SAMMY analysis between 80 keV and 100 keV are presented in Fig. 3 for the cross-section data obtained from the experimental transmission and capture cross-section measurements. The ^{55}Mn resonance parameters and the corresponding covariance matrix derived in the evaluation have been used to prepare a new cross-section evaluation for release with ENDF/B-VII.1.

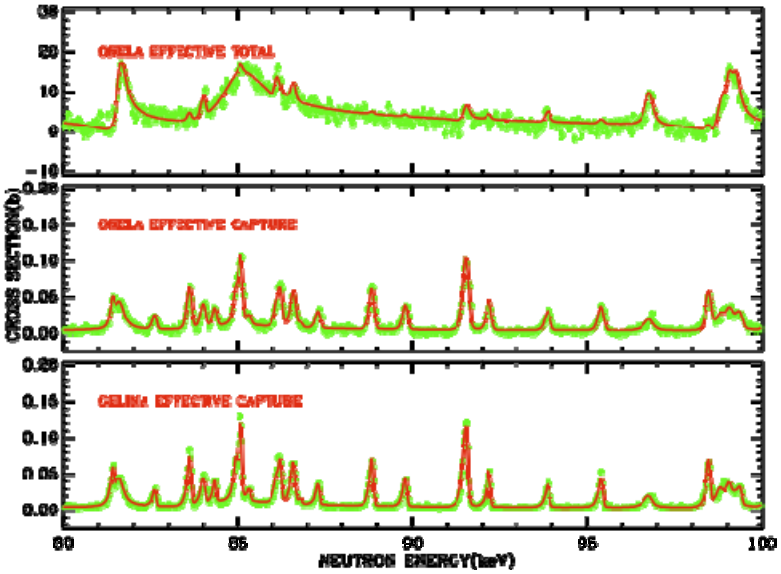

Figure 3. SAMMY fit of the measured ^{55}Mn total and capture cross-section data between 80 keV and 100 keV.

3.6 ^{58}Ni and ^{60}Ni Evaluations

New resonance region evaluations for ^{58}Ni and ^{60}Ni were performed with SAMMY [30]. The SAMMY analysis was started with the ENDF/BVII.0 resonance parameters as prior values, in the neutron energy ranges thermal to 812 keV for ^{58}Ni, and thermal to 450 keV for ^{60}Ni. For ^{58}Ni, all the neutron transmission data of the experimental database were consistent within 1% for the normalization and within less than 0.002 for the background. In order to fit the ORELA effective capture cross-section data, a background contribution not described by the resonance parameters was needed. Part of this background could be explained by the DC component, and by the contribution of nonidentified d-wave resonances in the high neutron energy range of the data. An example of SAMMY fit of the experimental data is shown on Fig. 4. The smooth curve represents the transmission data calculated with SAMMY. The Brusegan data were multiplied by a factor of 2 for clarity in the figure. For ^{58}Ni, a total of 487 resonances were used to fit the experimental database in the energy range thermal to 812 keV with 61 s-wave, 204 p-wave, and 222 d-wave resonances.

For ^{60}Ni, the ORELA low-energy range transmission data (^{6}Li glass detector) were consistent within 1% of the normalization, with a small background correction. The data at higher energies (NE-110 proton recoil detector) needed large normalization and background corrections in the energy range below about 200 keV. Above 200 keV, the corrections were very small, and the data were consistent with the GELINA transmission data in the energy range above 500 keV. The ORELA effective capture cross sections could not be fitted without an important residual background varying from 10 mb at 5 keV to about 0.5 mb at 400 keV. Part of this background was explained by the direct capture component and by the contribution of missing p-wave or d-wave resonances. An example SAMMY fit to the measured data is given in Fig. 5. The smooth curve represents the transmission calculated with SAMMY. The Brusegan data were multiplied by a factor of 2 for clarity in the figure. A total of 458 resonances was used to represent the experimental data in the energy range from thermal to 812 keV, including 61 s-wave, 236 p-wave, and 161 d-

wave. In addition, the new ^{58}Ni and ^{60}Ni resonance parameter evaluations also include the corresponding RPCMs that were derived as part of the resonance parameter evaluation effort.

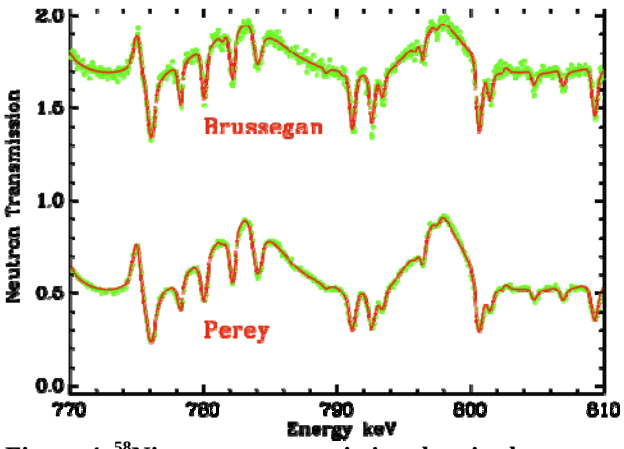

Figure 4. ^{58}Ni neutron transmission data in the energy range 770 keV to 810 keV from Grusegan et al. (upper fit), and Perey et al. (lower fit). The smooth curve represents the transmission data calculated with SAMMY. The Brusegan data were multiplied by a factor of 2 for clarity in the figure.

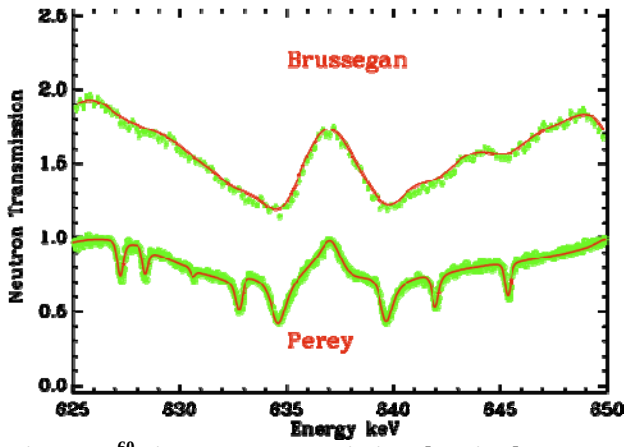

Figure 5. ^{60}Ni neutron transmission data in the energy range 625 keV to 650 keV from Brusegan et al. (upper fit), and Perey et al. (lower fit). The smooth curve represents the transmission calculated with SAMMY. The Brusegan data were multiplied by a factor of 2 for clarity in the figure.

3.7 ^{233}U, ^{235}U, ^{238}U, and ^{239}Pu Covariance Evaluations

For ENDF/B-VII.0, ORNL developed a new Reich-Moore resonance evaluation for ^{233}U in the energy range 0 to 600 eV using SAMMY. A total of 769 resonances, including the external levels, were used. At the time the evaluation was performed, the RPCM was generated; however, the RPCM matrix was not retained for the initial evaluation submittal. Therefore, the retroactive approach [31] was used to generate the ^{233}U RPCM. Each resonance of ^{233}U in the Reich-Moore formalism is described by five parameters (the resonance energy E_r, the gamma width Γ_γ, the neutron width Γ_n, and two fission widths Γ_{f1} and Γ_{f2}) for a total of 3845 parameters. The ^{233}U fission correlation matrix processed from the pre-release ENDF/B-VII.1 evaluation into the SCALE 44-group structure is shown in Fig. 6. Twenty-eight energy groups are in the energy region below 600 eV. Below 1 eV the fission cross-section data are highly correlated. Above the resonance region, the correlation data are based on the high-energy evaluation work at LANL. Together, ORNL and LANL have produced a complete covariance evaluation for ^{233}U that will be released with ENDF/B-VII.1.

With regard to ^{235}U, a Reich-Moore resonance evaluation has been performed in the energy range from 0 to 2250 eV using SAMMY [32]. A total of 3193 resonances, including the external levels, are provided in the ^{235}U resonance evaluation. As noted for ^{233}U, the ^{235}U covariance matrices were not retained when the latest resonance parameter evaluation was completed in the mid 1990s. In the current work, the retroactive approach was used to produce the ^{235}U covariance matrices. Each resonance of ^{235}U in the Reich-Moore formalism is described by five parameters that result in 15,965 parameters that must be considered in the covariance matrix analyses. The resulting RPCMs for ^{235}U have been submitted for inclusion with ENDF/B-VII.1. As an example, the ^{235}U capture cross-section covariance data processed with the AMPX module PUFF-IV [33] into the 44-group structure are shown in Fig. 7. The average uncertainty in the capture cross section as shown in Fig. 7 is about 1% up to 1 keV. In the covariance evaluation, a 1% uncertainty in the thermal cross section has been used. It should be noted that the 1% uncertainty in the thermal capture cross section has greatly influenced the correlation of the resolved resonance energy region up to 1 keV. As noted for ^{233}U, the high-energy correlation data above the energy region were prepared by LANL [34].

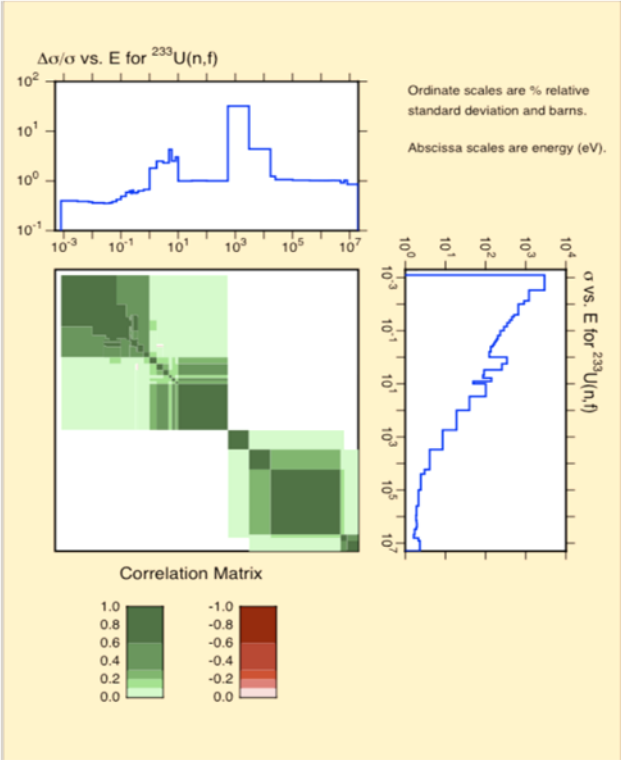

Figure 6. ^{233}U fission cross-section correlation matrix.

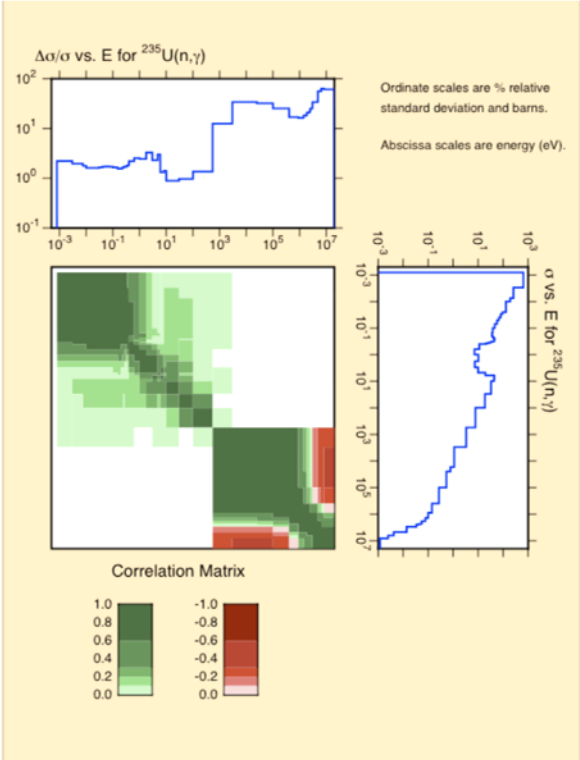

Figure 7. ^{235}U capture correlation matrix.

A detailed description of the cross-section evaluation for ^{238}U can be found in [35]. The ^{238}U resonance parameters are available in the ENDF/B-VII.0 library. The evaluation resulted in 3312 resonances in the energy range from 0 to 20 keV, 22 negative-energy resonances, and 9 resonances above 20 keV for a total of 3343 resonances. Because the fission cross section is negligible below 20 keV, each resonance of ^{238}U in the Reich-Moore formalism can be described by only three parameters: the resonance energy E_r, the gamma width Γ_γ, and the neutron width Γ_n. The resonance evaluation was performed in the energy range from 0 to 20 keV using SAMMY including corrections for experimental conditions (e.g., Doppler and resolution broadening, multiple scattering corrections, backgrounds, etc.). The experimental uncertainties are incorporated directly into the evaluation process in order to propagate those uncertainties into the resonance parameter results. Uncertainties treated during the ^{238}U evaluation process included statistical and systematic uncertainties for each of the differential data sets plus the quoted uncertainties for the resonance integral and thermal cross section. The result of the evaluation effort is a complete RPCM associated with the set of resonance parameters obtained in [35]. To illustrate the ^{238}U covariance data evaluation, capture cross-section correlation data for the 44-neutron energy groups calculated with the PUFF-IV code are shown in Fig. 8. Uncertainties in the capture data in the resonance region (up to 20 keV) range from 1 to 5%. Above 20 keV the uncertainty is on the order of 5% and reaches 25% near 20 MeV. The high-energy ^{238}U correlation data above the energy region were prepared by LANL [34].

A resolved resonance evaluation of the ^{239}Pu cross-section data has been completed with SAMMY in the energy region 0 to 2.5 keV. The evaluation of the RPCM was completed with a single SAMMY run including all the experimental data. The experimental database contains data in partial energy ranges. Since there is no experimental data set covering the entire energy range of the resonances, the RPCM was derived based on “experimental" fission and capture cross-section dataset generated in the energy range 0 to 2500 eV from the resonance parameters with resolution at least equivalent to the experimental resolution. To illustrate the ^{239}Pu covariance data, the PUFF-IV module was used to process the ^{239}Pu covariance data and generate covariance data in the 44-group structure. The ^{239}Pu capture cross-section correlation data are shown in Fig. 9. The high-energy

covariance evaluation (above 2500 eV) was performed at LANL [34]. In the resonance region the uncertainty in the capture cross section varies between 2 and 10%, whereas above the resonance region it can be as high as 40%.

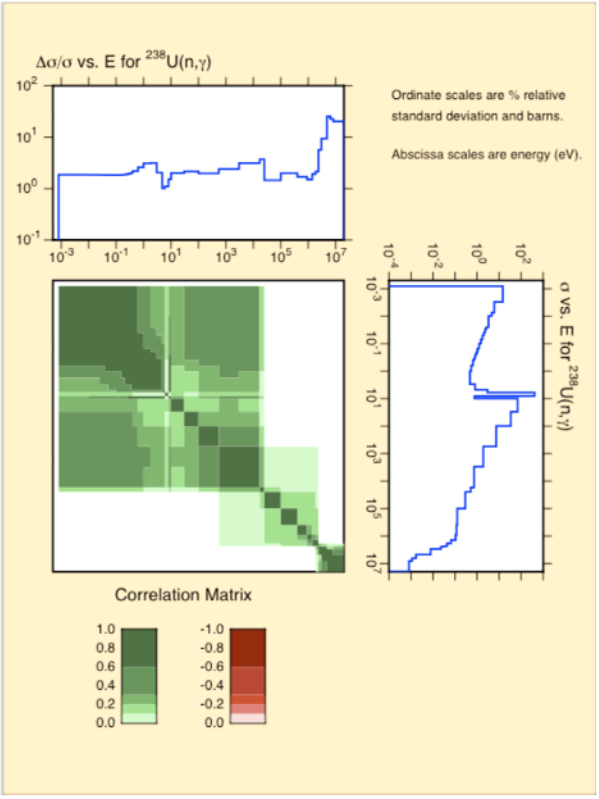

Figure 8. ^{238}U capture correlation matrix.

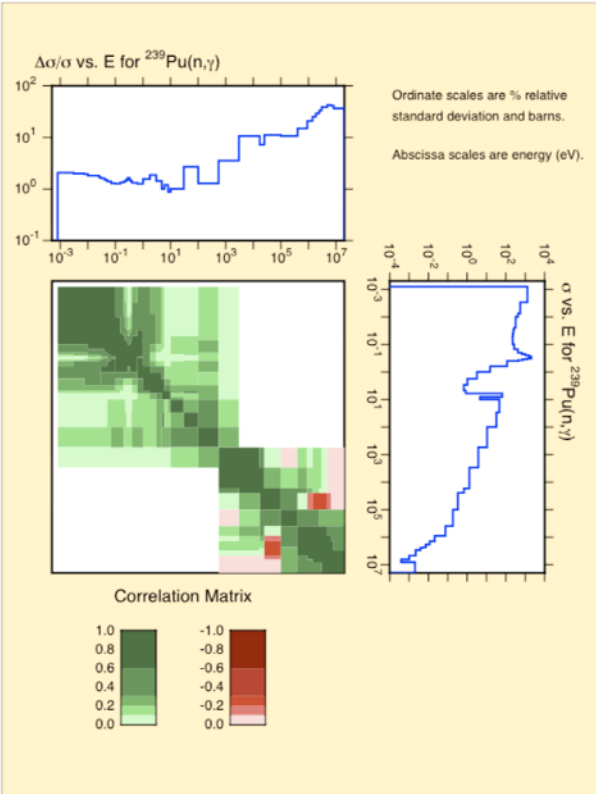

Figure 9. ^{239}Pu capture correlation matrix.

4 CONCLUSIONS

ORNL has performed nuclear data analysis work to prepare new cross-section evaluations for more than 20 priority isotopes of interest for nuclear criticality safety applications. This paper provides a summary of the nuclear data work for select nuclear data evaluations that have been prepared in support of the U.S. NCSP. As part of the work effort, ORNL has performed new differential data measurements at the ORELA facility in the United States and the GELINA facility in collaboration with IRMM in Geel, Belgium. Using the new cross-section measurements and measured cross-section data available in the literature, ORNL has produced new resonance region cross-section evaluations with corresponding covariance data. The new ORNL evaluations have been submitted to the U.S. NNDC for release with ENDF/B-VII.1.

5 ACKNOWLEDGMENTS

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