

## COVARIANCE DATA FOR $^{233}\text{U}$ IN THE RESOLVED RESONANCE REGION FOR CRITICALITY SAFETY APPLICATIONS

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### ABSTRACT

Uncertainties in nuclear data play an important role in determining the uncertainties in criticality safety calculations. The error estimation for calculated quantities relies on nuclear data uncertainty information available in the basic nuclear data libraries such as the U.S. Evaluated Nuclear Data File (ENDF/B); the uncertainty files (covariance matrices) in the ENDF/B library are generally obtained from analysis of experimental data. The computer code SAMMY, widely used for analyses of experimental data in the resolved and unresolved resonance energy regions, is used here to generate a physically-reasonable covariance matrix for the  $^{233}\text{U}$  resonance parameters in the resolved-energy region (0 to 600 eV) for use for criticality safety applications. SAMMY uses the generalized least-squares formalism (Bayes' method) together with the resonance formalism (R-matrix theory) for analysis of experimental data. Two approaches are available for creation of resonance-parameter covariance data: (1) During the data-evaluation process, SAMMY generates both a set of resonance parameters that fit the experimental data and the associated resonance-parameter covariance matrix. (2) For existing resonance-parameter evaluations for which no resonance-parameter covariance data are available, SAMMY can retroactively create an approximate resonance-parameter covariance matrix. In this report, our use of the retroactive method to generate covariance data for  $^{233}\text{U}$  is described. The resulting covariance matrix was then used as input to the ERRORJ code, which processed the covariance data into multigroup form, and to the TSUNAMI code, which calculated the uncertainty in the multiplication factor due to uncertainty in the experimental cross sections.

**KEYWORDS:** Resonance analysis, covariance generation, benchmark calculations

### 1. INTRODUCTION

Over the years, efforts have been made to improve the quality of basic nuclear data. Thermal reactor designs and applications have been the driving force for new data evaluations in the low-energy range. Data evaluations in the high-energy region have been accomplished primarily in support of shielding applications and fast reactor design. Although the accuracy of the nuclear data (such as neutron interaction cross sections) has significantly improved, little information

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exists on nuclear data uncertainties, and even less exists on nuclear data covariances. Efforts are now underway to correct these deficiencies.

At Oak Ridge National Laboratory (ORNL), data evaluations in the resolved and unresolved resonance energy regions are performed with the computer code SAMMY [1]. In a SAMMY evaluation, evaluation of experimental data (transmission or total cross section, capture, and fission cross section) incorporates uncertainties in the experimental data. Various sources of experimental uncertainties must be included; among these are normalization, background, and neutron time-of-flight, sample thickness, and temperature. Uncertainties in all of these are included in the evaluation process in order to properly determine the resonance-parameter covariance matrix.

## 2. COVARIANCE EVALUATION

Covariance matrices for resonance parameters have not been reported in the current evaluated data files because the dimension of the matrix can be very large and because, until recently, formats in the Evaluated Nuclear Data Files (ENDF) were often inadequate. These obstacles have been eliminated with the advent of the new “compact” resonance-parameter covariance matrix format proposed by these authors and adopted by the Cross Section Evaluation Working Group in 2004 for ENDF/B-VII [2]. In the compact formalism, correlation coefficients (between  $-1.0$  and  $+1.0$ ) are mapped into signed integer numbers from  $-99$  to  $99$  and correlations less than 2% are dropped. Further reduction of the storage of the covariance data can be achieved by dropping correlations less than an arbitrary value. To evaluate the impact of reducing the storage, flux-weighted group cross sections, based on the Bondarenko narrow resonance weighting scheme and the associated uncertainties, were calculated with the SAMMY code using the 44-group structure of the SCALE system.

### 2.1. Generating the Resonance Covariance Data for $^{233}\text{U}$ with SAMMY

As reported elsewhere [3], a Reich-Moore resonance evaluation (ENDF/B-VI) for  $^{233}\text{U}$  has been performed in the energy range 0 to 600 eV using the computer code SAMMY. A total of 769 resonances, including the external levels, were used. At the time the evaluation was performed, the resonance-parameter covariance matrix was generated; however, this matrix is no longer available. Unfortunately, this is also true for most existing evaluations. Therefore, an approach was developed within SAMMY to retroactively generate approximate covariance matrices for resonance parameters. This procedure has been used to generate the covariance matrix for the  $^{233}\text{U}$  parameters. Each resonance of  $^{233}\text{U}$  in the Reich-Moore formalism is described by five parameters (the resonance energy  $E_r$ , the gamma width  $\Gamma_\gamma$ , the neutron width  $\Gamma_n$ , and the two fission widths  $\Gamma_{f1}$ , and  $\Gamma_{f2}$ ), for a total of 3845 parameters. The large number of resonance parameters leads to two major issues when generating a resonance covariance: (a) the computer memory required to process the data and (b) the data storage for the resulting covariance file. The former has been addressed by using an in-house computer with 32 gigabytes of memory. To address the latter issue, the compact format was used. To illustrate the use of the compact format for  $^{233}\text{U}$ , the resulting covariance matrix in the existing ENDF format required a file of 100 megabytes. Conversion of the  $^{233}\text{U}$  covariance matrix into the new compact format requires only 2 megabytes. The remarkable reduction in the storage space permits the processing of SAMMY-

generated covariance data and the subsequent generation of covariance matrices for groupwise cross sections using processing codes.

## 2.2. Group Average Cross Section and Associated Covariance Matrix

The final result of a SAMMY evaluation is a parameterization of the neutron cross sections. If  $p_1, p_2, \dots, p_n$  are the resonance parameters, a reaction cross section  $\sigma_x$  is written as

$$\sigma_x = \sigma_x(p_1, p_2, \dots, p_n) \quad (1)$$

The covariance matrix elements  $\langle \delta p_i \delta p_j \rangle$  for the resonance parameters are calculated in SAMMY, as described above. Likewise, the derivatives of the cross section with respect to the resonance parameters (i.e., the sensitivity coefficients  $\partial \sigma_x / \partial p_i$ ) are also calculated.

Flux-weighted group cross sections  $\bar{\sigma}_{xg}$  are defined for a reaction cross section as

$$\bar{\sigma}_{xg} \Phi_g = \int_{E_g}^{E_{g+1}} \sigma_x(E) \Phi(E) dx \quad (2)$$

with  $\Phi_g$ , the neutron flux in the energy group  $g$ , given by

$$\Phi_g = \int_{E_g}^{E_{g+1}} \Phi(E) dx \quad (3)$$

In particular, for the Bondarenko narrow-resonance weighting scheme, the flux  $\Phi(E)$  is

$$\Phi(E) = \frac{C(E)}{\sigma_0 + \sigma_t(E, T)} \quad (4)$$

where  $C(E)$  is a smooth function of energy,  $\sigma_0$  is the background cross section, and  $\sigma_t(E, T)$  is the energy- and temperature-dependent total cross section. The covariance matrix for the group cross section is obtained by taking small increments in  $\bar{\sigma}_{xg}$  with respect to the resonance parameters as

$$\delta \bar{\sigma}_{xg} = \sum_j \frac{\partial \sigma_{xj}}{\partial p_j} \delta p_j \quad (5)$$

Squaring and taking expectation values give the covariance matrix elements as

$$\langle \delta \bar{\sigma}_{xg} \delta \bar{\sigma}_{xg'} \rangle = \sum_{j,k} \frac{\partial \sigma_{xj}}{\partial p_j} \langle \delta p_j \delta p_k \rangle \frac{\partial \sigma_{xk}}{\partial p_k} \quad (6)$$

The covariance of  $\bar{\sigma}_{xg}$  obtained from Eq. 5 is a function of the sensitivities (derivative of the cross sections with respect to the parameters  $p_j$ ) and of the covariance of the parameters  $p$  given as  $\langle \delta p_j \delta p_j \rangle$ . Sensitivities are calculated either analytically (as in SAMMY) or numerically (as in many processor codes). Covariance matrix elements are available from the SAMMY fitting of the experimental data, as described above.

Twenty-nine energy groups of the SCALE 44-group structure [4] are in the energy region below 600 eV; the group boundaries are shown in Table I. Average fission cross sections were calculated with SAMMY for 500 barns background cross section using Eqs. 2-4 with constant  $C(E)$ .

The impact of dropping correlations in the resonance covariance matrix on the group cross section covariance matrix has been studied for the fission cross section. The percent uncertainties in the calculated group fission cross section for the case of no correlations removed and for removal of correlations less than 2%, and 3% are shown in Table II. The correlation matrix for the group fission cross section is shown in Fig. 1. Below 1 eV the group fission cross sections are highly correlated.

**Table I. Boundaries for groups in the energy region below 600 eV**

Group number*	Lower boundary (eV)	Upper boundary (eV)
1	1.00000E-05	3.00000E-03
2	3.00000E-03	7.50000E-03
3	7.50000E-03	1.00000E-02
4	1.00000E-02	2.53000E-02
5	2.53000E-02	3.00000E-02
6	3.00000E-02	4.00000E-02
7	4.00000E-02	5.00000E-02
8	5.00000E-02	7.00000E-02
9	7.00000E-02	1.00000E-01
10	1.00000E-01	1.50000E-01
11	1.50000E-01	2.00000E-01
12	2.00000E-01	2.25000E-01
13	2.25000E-01	2.50000E-01
14	2.50000E-01	2.75000E-01
15	2.75000E-01	3.25000E-01
16	3.25000E-01	3.50000E-01
17	3.50000E-01	3.75000E-01
18	3.75000E-01	4.00000E-01
19	4.00000E-01	6.25000E-01
20	6.25000E-01	1.00000E+00
21	1.00000E+00	1.77000E+00
22	1.77000E+00	3.00000E+00
23	3.00000E+00	4.75000E+00
24	4.75000E+00	6.00000E+00
25	6.00000E+00	8.10000E+00
26	8.10000E+00	1.00000E+01
27	1.00000E+01	3.00000E+01
28	3.00000E+01	1.00000E+02
29	1.00000E+02	5.50000E+02

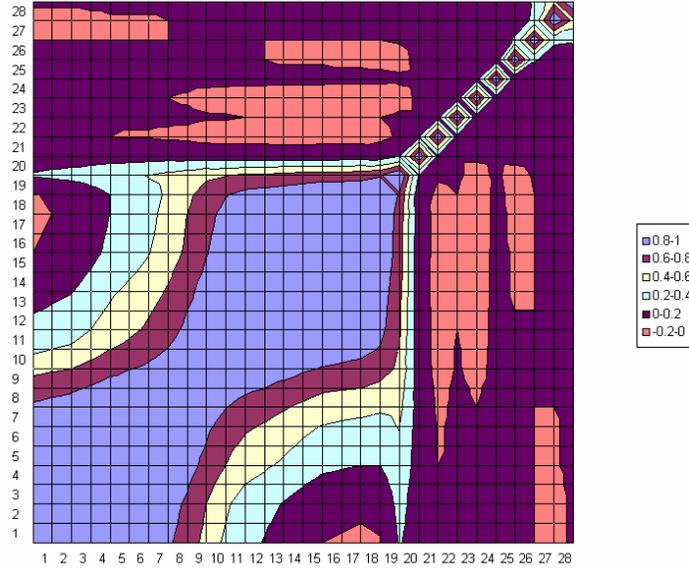
\*Group order reversed relative to the normal SCALE group structure

**Table II. Percent uncertainties in the calculated fission cross section**

Group number	No correlations deleted	Delete correlations < 2%	Delete correlations < 3%
1	1.5	1.9	2.1
2	1.5	1.9	2.1
3	1.5	1.9	2.1
4	1.5	1.9	2.1
5	1.5	1.9	2.1
6	1.5	1.9	2.1
7	1.5	1.9	2.1
8	1.5	1.9	2.0
9	1.5	1.8	2.0
10	1.4	1.8	1.9
11	1.3	1.7	1.8
12	1.3	1.7	1.8
13	1.4	1.7	1.9
14	1.4	1.7	1.9
15	1.4	1.7	1.9
16	1.4	1.7	1.9
17	1.4	1.7	1.8
18	1.4	1.7	1.8
19	1.3	1.7	1.8
20	1.3	1.6	1.7
21	1.4	1.4	1.4
22	1.6	1.6	1.6
23	1.1	1.1	1.2
24	1.8	2.0	2.0
25	1.7	1.7	1.7
26	1.7	1.7	1.8
27	0.4	0.4	0.4
28	0.2	0.2	0.2
29	0.2	0.1	0.1

### 3. APPLICATION OF DATA UNCERTAINTY TO CRITICALITY SAFETY BENCHMARK CALCULATION

Uncertainty in nuclear data is of vital importance in the determination of the uncertainty in the calculated system multiplication factor. Covariance data have been generated for  $^{233}\text{U}$  in the 44-neutron group structure of the SCALE [4] system using the processing code ERRORJ [5]. Group covariance data were converted to the standard COVERX [6] format for use in benchmark calculations using the computer code TSUNAMI [7]. All the calculations were done with the SCALE 44-group ENDF/B-V except the  $^{233}\text{U}$  from ENDF/B-VI.



**Fig. 1. Group cross-section correlations for the fission cross section.**

The sensitivity  $S$  of the calculated multiplication factor to the cross section is defined as

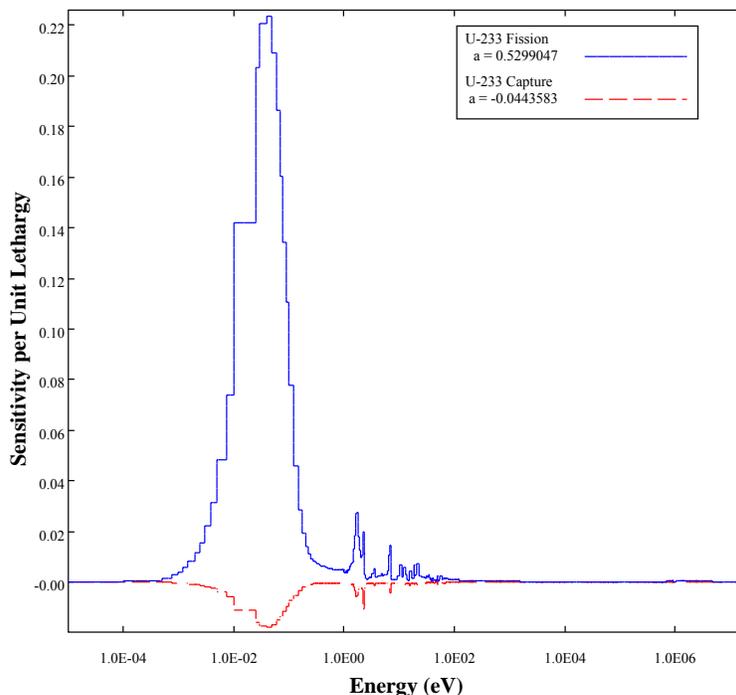
$$S = \frac{\sigma_x}{k} \frac{\partial k}{\partial \sigma_x} , \quad (7)$$

where  $k$  is the calculated multiplication factor and  $\sigma_x$  is the reaction cross section. If the covariance  $C$  determined from the experimental data is known, the variance  $V$  (or the standard deviation  $\sqrt{V}$ ) of the calculated multiplication factor is given by

$$V = S C S^t \quad (8)$$

where  $S^t$  is the transpose of  $S$ .

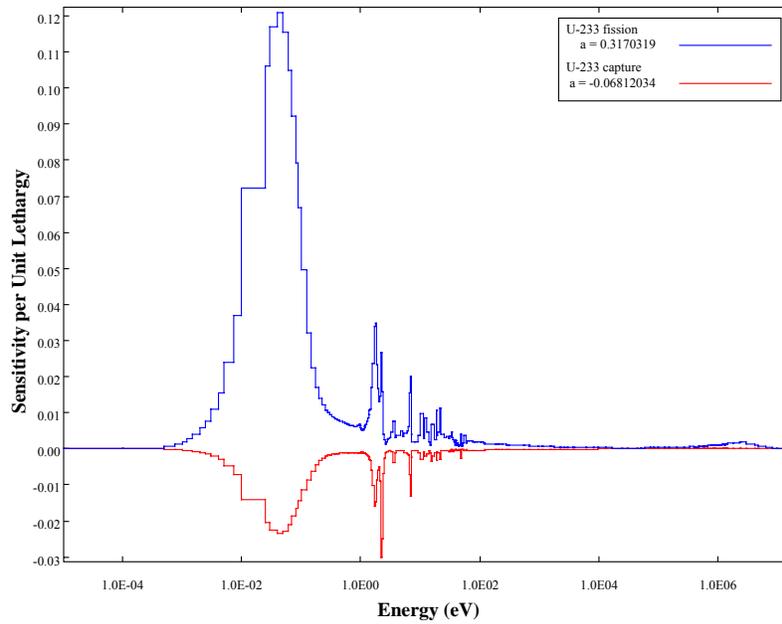
Sensitivity calculations for benchmarks with  $^{233}\text{U}$  have been performed at ORNL [8]. The sensitivity coefficients determined in the calculations and the 44-group covariance data generated for  $^{233}\text{U}$  with ERRORJ, converted into the COVERX format, were used to determine the variance and consequently the relative standard deviation in the multiplication factor using the code TSUNAMI. The first benchmark for which the uncertainty in the multiplication factor was studied was an unreflected homogeneous aqueous uranyl nitrate solution sphere of  $^{233}\text{U}$ , the so-called  $^{233}\text{U}$  ORNL spheres [9]. The  $\text{H}/^{233}\text{U}$  ratio for the case studied is 1533 with a  $^{233}\text{U}$  enrichment of 97.7 wt %. The multiplication factor calculated with the TSUNAMI code for the benchmark system is 0.9982. The sensitivity of the multiplication factor to the fission and capture cross sections, as given by Eq. 7, is shown in Fig. 2. As can be seen, this thermal benchmark system is very sensitive to the  $^{233}\text{U}$  fission cross section.



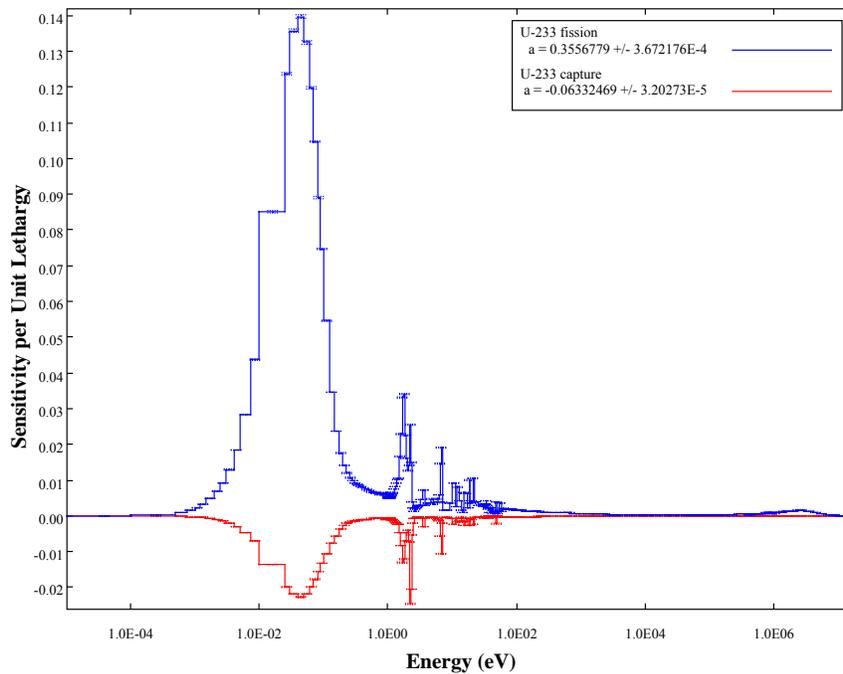
**Fig. 2. Sensitivity of the multiplication factor to the fission and capture cross section for  $^{233}\text{U}$  for the ORNL5 sphere benchmark system.**

According to Eq. 8, the resulting standard deviation for the multiplication factor calculated with the TSUNAMI code that is attributable to the  $^{233}\text{U}$  is 0.0191.

Sensitivity calculations were also performed for a water-reflected  $^{233}\text{U}$  nitrate solution benchmark referred to in the ICBSP book [10] as the U233-SOL-THERM-005 benchmark. This benchmark is part of an effort conducted during the 1950s at ORNL that was directed toward determining bounding values for the minimum critical mass, minimum critical volume, and maximum safe pipe size of water-moderated solutions. Calculations were performed with the TSUNAMI code for two benchmarks indicated in the ICBSP for these experiments: a spherical geometry with a  $\text{H}/^{233}\text{U}$  ratio of 405 and a cylindrical geometry with a  $\text{H}/^{233}\text{U}$  ratio of 514 with  $^{233}\text{U}$  enrichment of 98.7 wt %. The sensitivity to the  $^{233}\text{U}$  fission and capture cross sections for the spherical geometry benchmark system cases one and two are shown in Figs. 3 and 4, respectively. The systems are thermal benchmarks that are sensitive to the  $^{233}\text{U}$  cross section. The multiplication factors calculated with TSUNAMI for these systems are 0.9999 (for case one) and 1.0000 (for case 2). The resulting standard deviations for the multiplication factor calculated with the TSUNAMI code that are attributable to the  $^{233}\text{U}$  are 0.0137 (for case one) and 0.0147 (for case two). The TSUNAMI calculations for the sphere cases were done base on the TSUNAMI-1D which is deterministic. The cylindrical benchmark calculations were done with TSUNAMI-3d (Monte Carlo).



**Fig. 3. Sensitivity of the multiplication factor to the fission and capture cross section for  $^{233}\text{U}$  for the U233-SOL-THERM-005 case one benchmark system.**



**Fig. 4. Sensitivity of the multiplication factor to the fission and capture cross section for  $^{233}\text{U}$  for the U233-SOL-THERM-005 case two benchmark system.**

### 3. CONCLUSIONS

A methodology has been developed at ORNL to generate covariance data in the resolved energy region using the computer code SAMMY. It has been applied to  $^{233}\text{U}$  using the retroactive covariance generation option in the SAMMY code. To our knowledge, this is the first time that R-matrix covariance data were generated with SAMMY and effectively used in criticality safety calculations. Several difficulties related to computer storage have been resolved by using an in-house computer with 32 gigabytes of memory. The use of the covariance data in practical applications has been demonstrated by generating covariance data in the 44-group structure of the SCALE system. The code ERRORJ was used to generate the group covariance. Sensitivity calculations were performed with the code TSUNAMI using covariance data determined directly from experimental uncertainties. The benchmark calculations reported in this paper show that the relative standard deviations for the multiplication factors range from 1 to 2%. The nuclear criticality safety practitioner may use these results to help establishing margin for criticality safety calculations in practical applications.

### ACKNOWLEDGMENTS

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