Bias Assessment of ^{233}U Systems Using a GLLS Methodology with SCALE TSURFER

Bradley T. Rearden and Don Mueller

Oak Ridge National Laboratory, P.O. Box 2008, Bldg. 5700, Oak Ridge, TN 37831-6170, USA,*
ReardenB@ornl.gov and MuellerDE@ornl.gov

OVERVIEW OF TSURFER TECHNIQUES

A new capability for SCALE 6 [1] allows the prediction of computational biases and bias uncertainties with the Tool for Sensitivity and Uncertainty Analysis of Response Functions Using Experimental Results (TSURFER), which is based on a generalized linear least-squares approach.[2] The TSURFER approach to bias and bias uncertainty assessment is quite different from those traditionally used in criticality safety evaluations. The analysis is conducted with a suite of benchmark experiments for which the benchmark k_{eff} values and their uncertainties, as well as correlations in uncertainties between benchmarks, are quantified. The analysis considers the computed k_{eff} results and their uncertainties due to nuclear data uncertainties, as represented by the cross-section-covariance data. Where differences in the benchmark and computed k_{eff} values are realized, TSURFER identifies a single set of adjustments to nuclear data and benchmark k_{eff} values that will result in the computational models all producing computed k_{eff} values equivalent to the benchmark k_{eff} values. The unique solution is found using a GLLS procedure that minimizes the overall χ^2 of the adjustment. Then the same nuclear data adjustments are used to predict an adjusted k_{eff} value for the application system and an uncertainty on the adjusted k_{eff} value based on uncertainties remaining in the nuclear data after the adjustment. The unbiased or adjusted k_{eff} for the application is computed through sensitivity coefficients, not through a criticality calculation using adjusted nuclear data. The difference between the originally calculated k_{eff} value and the unbiased k_{eff} value represents the bias in the original calculation, and the uncertainty in the adjusted k_{eff} represents the uncertainty.

If similar experiments are available to validate the use of a particular nuclide in the application, the uncertainty of the bias for that nuclide is reduced through the generalized linear least-squares procedure. In TSURFER, experiments that are dissimilar from the application can still provide useful information for bias assessment if at least one material demonstrates similar sensitivities to those of the application. If similar experiments are not available to validate a particular nuclide, a high uncertainty in the bias for the given nuclide will result. Thus, with a complete set of experiments to validate important components in the application, a precise bias with a small uncertainty can be predicted. Where the experimental coverage is lacking, a bias can be predicted with an appropriately large uncertainty. As users gain experience with TSURFER, it is expected to become a preferred tool for rigorous bias and bias uncertainty determination, particularly for applications for which nearly identical critical experiments are not available.

REPRESENTATIVE ^{233}U SAFETY ANALYSIS MODELS

At the U.S. Department of Energy's request, Oak Ridge National Laboratory (ORNL) staff used the SCALE TSUNAMI tools to provide a demonstration evaluation of critical experiments considered for use in validation of current and anticipated operations involving ^{233}U at the Radiochemical Development Facility (RDF) at ORNL. This work was reported in ORNL/TM-2008/196, issued in January 2009.[3] This paper documents the TSURFER bias and bias uncertainty analysis techniques applied to two applications systems from the larger report and complements a previous publication related to the sensitivity, uncertainty, and trending analysis of these same applications.[4]

The model referred to as Application 1 in Ref. [3] is a 12.2-cm-radius sphere of 220 g U per liter uranyl nitrate solution with no excess acid. The uranium is 100 wt % ^{233}U. The solution sphere is reflected by 0.25 cm of Type 304 stainless steel (ss-304) and 2 cm of water. Calculations were conducted with SCALE 5.1 using KENO V.a in the TSUNAMI-3D-K5 sequence with ENDF/B-VI.7 cross-section data for all nuclides except ^{233}U where ENDF/B-VII.0 cross-section data were used. The energy of average lethargy of neutrons causing fission (EALF) calculated for this application was 0.282 eV. The k_{eff} calculated for this system is 1.0028 ± 0.0002.

The model referred to as Application 3 in Ref. [3] is a 53.0-cm-radius sphere of 600 g U per liter uranyl nitrate solution with no excess acid. The solution temperature is 80°C (353 K). The uranium is 3 wt % ^{233}U, 0.2 wt % ^{235}U, and 96.8 wt % ^{238}U. The fissile solution sphere is reflected by 0.25 cm of ss-304 and 2.0 cm of water. The

* Notice: This manuscript has been authored by UT-Battelle, LLC, under contract DE-AC05-00OR22725 with the U.S. Department of Energy. The United States Government retains and the publisher, by accepting the article 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.

EALF calculated for this application was 0.0631 eV. The k_{eff} calculated for this system is 0.9690 ± 0.0002.

The TSUNAMI similarity assessment found 82 benchmark experiments that were highly similar to Application 1, but no experiments that were even marginally similar to Application 3.

APPLICATION OF TSURFER TO ^{233}U SYSTEMS

Bias and Bias Uncertainty Assessment

A calculation was performed using the SCALE 6 version of TSURFER, the SCALE 6 covariance data library [5], and 1066 critical experiments, including those described in Ref. [4], to calculate the set of data adjustments that minimize the overall χ^2. An additional important input to the TSURFER calculation is the set of correlations between experiment k_{eff} values. For purposes of this study, it was assumed that all experiments within any one *International Handbook of Evaluated Criticality Safety Benchmark Experiments* [6] (IHECSBE) evaluation were 90% correlated and that there were no correlations between experiments in different evaluations. This approximation is arbitrary. In reality, the same fuel rods or plates are used in multiple evaluations and the correlations are far more complex than the simple assumption stated above.

The TSURFER-calculated bias and bias uncertainty values for Applications 1 and 3, identified in Refs. [3] and [4], are shown in Table I. Note that the bias uncertainty values are the uncertainties in the adjusted k_{eff} values. The reduction in these uncertainties relative to the original uncertainties in the applications is due to inclusion of additional information, the critical experiments, in the validation process. The original uncertainties in the k_{eff} of the applications, due to cross section covariance data, shown in Table II, are 0.937 and 0.515 %$\Delta k/k$, respectively. For Application 1, the uncertainty is reduced to approximately 10% of its original value, confirming that the benchmarks selected lead to adjustments in the cross sections that are the largest sources of uncertainty, and thus the largest potential sources of computational bias, for this case. For Application 3, the uncertainty in the bias is 31% of the original application uncertainty, indicating marginal coverage by the benchmark set. However, with this technique, several experiments, none of which exhibit an overall similarity to Application 3, can be used in combination to validate individual aspect of the system.

Table I. Bias, bias uncertainty, and USL values computed with TSURFER

Application	Bias (% $\Delta k/k$)	Bias uncertainty (% $\Delta k/k$)	USL
1	0.341	0.071	0.979
3	-0.091	0.159	0.976

Table II. Uncertainty in k_{eff} due to cross-section-covariance data

System	Standard deviation (%)	Top six contributors to standard deviation (%)	
Application 1	0.937	^{233}U χ to ^{233}U χ	0.819
		^{1}H elastic to ^{1}H elastic	0.320
		^{16}O elastic to ^{16}O elastic	0.194
		^{233}U n,γ to ^{233}U n,γ	0.174
		^{233}U nubar to ^{233}U nubar	0.145
		^{233}U fission to ^{233}U fission	0.117
Application 3	0.515	^{14}N n,p to ^{14}N n,p	0.346
		^{238}U n,γ to ^{238}U n,γ	0.233
		^{233}U fission to ^{233}U fission	0.173
		^{1}H n,γ to ^{1}H n,γ	0.145
		^{233}U χ to ^{233}U χ	0.136
		^{233}U nubar to ^{233}U nubar	0.135

Where the 2% additional margin used in Refs. [3] and [4] is applied, any positive bias disallowed, and a 95% confidence (2σ) applied for the bias uncertainty, the USL values shown in Table I result.

Detailed Bias Assessment

Through further examination of the results for Application 1, the individual sources of bias can be identified according to the impact of the adjusted cross sections on the application k_{eff} value. TSURFER ranks the bias contributions according to an L1 norm, which is based on the absolute values of the group-wise cross section adjustments. The top contributors to bias for Application 1 are shown in Table III. Comparing the TSURFER-predicted contribution to bias with the top sources of uncertainty shown in Table II, one finds that the ^{233}U reactions and ^{16}O elastic rank at the top of both tables. However, ^{1}H elastic scattering, the second highest source of uncertainty, appears as the 11th highest source of bias. For all nuclides and reactions, the bias contribution is approximately the same or less than the uncertainty shown in Table II, as the adjustments in the cross sections are bounded with a χ^2 of 1.2.

The energy-dependent adjustments for selected cross sections are shown in Fig. 1. A maximum adjustment of 7.3% is given for ^{233}U fission where ^{1}H elastic receives a maximum adjustment of <0.1%, confirming that it is not a significant source of bias. The magnitude and shape of the adjustments are constrained by the cross-section-covariance data.

Energy-Dependent Bias Assessment

The energy-dependent cross-section adjustments can be multiplied with the energy-dependent sensitivity of the application's k_{eff} to the cross sections to form an energy-dependent bias due to a specific nuclide-reaction pair. As shown in Fig. 2, the application bias is dependent on which cross sections were modified in the adjustment procedure, and how sensitive the application is to those adjustments. For example, the maximum adjustment for ^{233}U fission, shown in Fig. 1, occurs around 20 keV. However, the application is not very sensitive to the cross section in this energy range, and the bias due to this adjustment is small, as shown in Fig. 2. In the intermediate energies, the adjustment is on the order of 1%, but the application is more sensitive at these energies, so a larger bias contribution results. Also, as the thermal- and fast-energy cross sections are reduced where the intermediate-energy cross sections are increased, a positive contribution to the bias is observed for thermal and fast energies and a negative contribution to the bias is observed for intermediate energies.

Table III. Top contributors to TSURFER-determined bias for Application 1

Nuclide	Reaction	Contribution to bias % $\Delta k/k$	Fraction of bias L1-norm
^{233}U	chi	3.2738E-01	2.7907E-01
^{233}U	n,γ	9.8989E-02	2.5125E-01
^{16}O	elastic	-1.8796E-01	1.9356E-01
^{233}U	nubar	1.6175E-01	1.3797E-01
^{233}U	fission	-3.5477E-02	6.8533E-02
^{233}U	n,n'	-3.7597E-02	3.2049E-02
^{233}U	elastic	6.2944E-03	5.7039E-03
^{56}Fe	n,γ	6.4198E-03	5.4725E-03
^{14}N	n,p	6.1571E-03	5.2583E-03
^{1}H	n,γ	5.4157E-03	4.6187E-03
^{1}H	elastic	-4.9899E-03	4.4406E-03

Detailed Bias Uncertainty Assessment

As the TSURFER bias uncertainty is simply the uncertainty in the adjusted, or unbiased, k_{eff} value due to the adjusted cross-section data, the adjusted cross-section-covariance data resulting from the TSURFER procedure can be used to provide contributions to the bias uncertainty by individual covariance matrices using the extended uncertainty edit of TSUNAMI-IP. The

contributions to the uncertainty in the bias are shown in Table IV.

Here, anti-correlations in the adjusted covariance data become important components in the reduced uncertainty. Since all cross sections are adjusted from the same set of benchmark experiments, the number of cross-correlations in uncertainties for different nuclides and reactions greatly increases. Where sufficient quality benchmarks are available to validate a given nuclide reaction, the uncertainty in that nuclide reaction is reduced. However, due to correlations introduced in the adjustment procedure, the uncertainty in the cross section may be increased if only limited benchmarks that have significant sensitivities to a given nuclide reaction are available.

In comparing the initial uncertainties shown in Table II to those of Table IV, the uncertainties for all nuclide-reaction pairs in Applications 1 and 3 are reduced and new cross-correlations are introduced, which further reduce the uncertainty.

The detailed assessment of bias uncertainty is a gap analysis that guides the user to areas where validation coverage can be improved. If additional experiments are found to validate the processes identified in Table IV, the precision of the bias assessment can be improved by reducing the bias uncertainty.

CONCLUSIONS

The data adjustment techniques of the TSURFER tool of SCALE 6 have been applied to provide bias and bias uncertainty information for two ^{233}U applications. These results demonstrate the unique capabilities of TSURFER to combine information for many benchmarks where no overall similar benchmarks are available. Additionally, TSURFER's ability to provide detailed information regarding sources of bias and bias uncertainty to improve understanding of processes that contribute to bias and identify gaps in validation coverage has been demonstrated. This technique is also useful for prioritizing needs for better nuclear data.

Table IV. TSURFER bias uncertainty due to cross-section-covariance data

System	Standard deviation (%)	Top six contributors to standard deviation (%)	
Application 1	0.071	^{233}U chi to ^{233}U chi	0.201
		^{233}U n,γ to ^{233}U chi	-0.198
		^{233}U nubar to ^{233}U chi	-0.157
		^{233}U n,γ to ^{233}U n,γ	0.152
		^{233}U nubar to ^{233}U nubar	0.129
		^{16}O elastic to ^{233}U chi	-0.080
Application 3	0.159	^{233}U fission to ^{233}U fission	0.132
		^{233}U nubar to ^{233}U nubar	0.120
		^{14}N n,p to ^{14}N n,p	0.104
		^{233}U fission to ^{233}U nubar	-0.100
		^{1}H n,γ to ^{233}U fission	-0.086
		^{238}U n,γ to ^{238}U n,γ	0.078

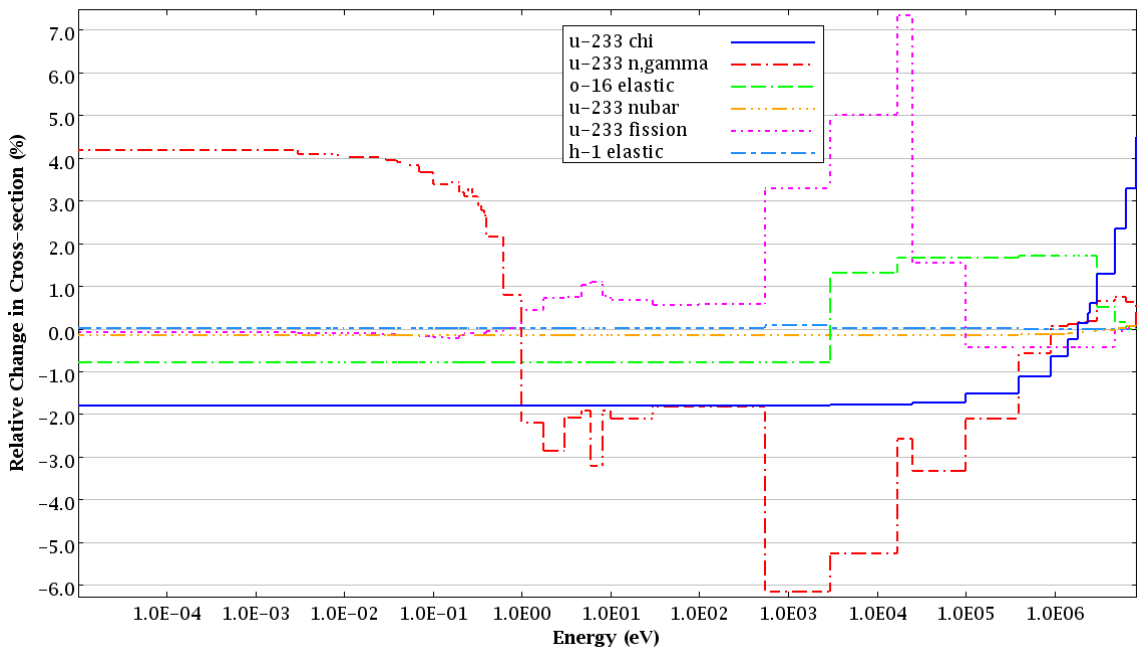

Fig. 1. Select cross-section adjustments from TSURFER analysis.

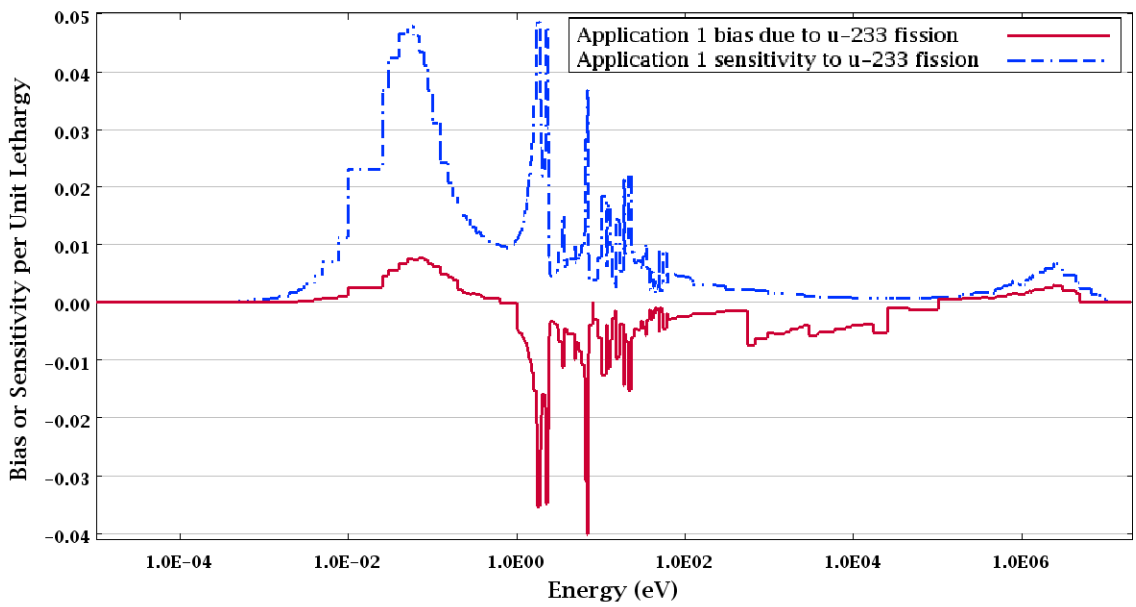

Fig. 2. Energy-dependent bias and sensitivity for ^{233}U fission for Application 1.

REFERENCES

1. *SCALE: A Modular Code System for Performing Standardized Computer Analyses for Licensing Evaluation*, ORNL/TM-2005/39, Version 6, Vols. I–III, Oak Ridge National Laboratory, Oak Ridge, Tennessee, January 2009. Available from Radiation Safety Information Computational Center at Oak Ridge National Laboratory as CCC-750.
2. B. L. BROADHEAD, B. T. REARDEN, C. M. HOPPER, J. J. WAGSCHAL, and C. V. PARKS, "Sensitivity- and Uncertainty-Based Criticality Safety Validation Techniques," *Nucl. Sci. Eng.*, **146**, 340–366 (2004).
3. D. E. MUELLER, B. T. REARDEN and D. F. HOLLENBACH, *Application of the SCALE*

TSUNAMI Tools for the Validation of Criticality Safety Calculations Involving ^{233}U, ORNL/TM-2008/196, Oak Ridge National Laboratory, Oak Ridge, Tennessee, January 2009.

4. D. E. MUELLER and B. T. REARDEN, "SCALE TSUNAMI Analysis of Critical Experiments for the Validation of ^{233}U System," *Trans. Am. Nucl. Soc.* **101**, 455–459 (2009).
5. M. L. WILLIAMS and B. T. REARDEN, "SCALE 6 Sensitivity/Uncertainty Methods and Covariance Data," *Nuclear Data Sheets* **109**, 2796–2800 (2008).
6. *International Handbook of Evaluated* Criticality *Safety Benchmark Experiments*, Organization for Economic Cooperation and Development—Nuclear Energy Agency, NEA/NSC/DOC(95)03 (2008 Edition).