SCALE TSUNAMI Analysis of Critical Experiments for Validation of ^{233}U Systems

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INTRODUCTION

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). This work was reported in ORNL/TM-2008/196 [1] issued in January 2009. This paper presents the analysis of two representative safety analysis models provided by RDF staff.

REPRESENTATIVE SAFETY ANALYSIS MODELS

Models representing three process conditions and one storage array configuration were provided by staff from the operations contractor Isotek, LLC [2] for use in analyses demonstrating the use of the SCALE TSUNAMI tools in computational method validation. This brief paper addresses only two of the four safety analysis models from Ref. [1] and a variant of a model that was excluded from the report.

The model referred to as Application 1 in Ref. [1], 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. The energy of average lethargy of neutrons causing fission (EALF) calculated for this application is 0.282 eV. The k_{eff} calculated for this system is 1.0028 ± 0.0002.

The model, referred to as Application 3 in Ref. [1], 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 EALF calculated for this application is 0.0631 eV. The k_{eff} calculated for this system is 0.9690 ± 0.0002.

A variant of Application 1 considered in preliminary studies was an infinite medium of 220 g U per liter uranyl nitrate solution with 9.5 M excess acid. In this model the uranium is 98 wt % ^{233}U, 1 wt % ^{235}U, and 1 wt% ^{238}U. The EALF calculated for this application is 0.446 eV. The k_{eff} calculated for this system is 2.055.

ANALYSIS METHODS

SCALE 5.1 [3] TSUNAMI tools TSUNAMI-3D, TSUNAMI-1D, and TSUNAMI-IP were used to generate k_{eff} sensitivity data for the applications and 672 critical experiments identified as being potentially useful for validation of these applications and to compare each application with each critical experiment. The candidate validation set included 232 ^{233}U configurations, 28 mixed U/Pu configurations, 4 intermediate uranium enrichment configurations, 153 high uranium enrichment configurations, and 255 low uranium enrichment configurations from 101 *International Handbook of Evaluated Criticality Safety Benchmark Experiments* [4] evaluations. Example upper subcritical limits (USLs) were generated for Application 1 based on trending of the TSUNAMI similarity parameters.

RESULTS

Uncertainty Quantification

The basis of the TSUNAMI validation techniques is that computational biases are primarily caused by errors in the cross-section data, which are quantified and bounded, with a 1σ confidence, by the cross-section-covariance data. The uncertainties in the computed k_{eff} values for Applications 1 and 3 due to uncertainties in the cross-section data were computed by combining the sensitivity of k_{eff} to the cross-section data with the cross-section-covariance data. This analysis phase gives insight as to potential contribution to system bias from nuclear data errors. Partial results for this analysis phase are presented in Table I. Benchmark experiments with similar uncertainties should have similar computational biases and be useful in the validation of these applications.

As all three systems are thermal solutions with EALF values of 0.0631–0.446 eV, one might expect k_{eff} to have a significant sensitivity to ^{1}H elastic scattering in each system. However, the ^{1}H elastic scattering is a significant source of cross-section uncertainty only in Application 1. The sensitivities of k_{eff} to ^{1}H elastic scattering for each system are shown in Fig. 1. Here is it observed that Application 1 has a significant sensitivity at fast energies. Application 3's larger size reduces the sensitivity at fast energies, but the presence of ^{238}U increases the

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importance of ^{1}H elastic scattering for the resonance escape probability. For the 220 g U per liter 9.5 M excess acid infinite medium model, the lack of leakage and low ^{238}U content limit the sensitivity across the entire spectrum. Although EALF values may indicate that these systems are similar, the physics indicate that different physical processes are driving k_{eff} in each system.

Similarity Assessment

The similarity of the selected critical experiments to each application was evaluated using the c_r parameter, which is a correlation coefficient that quantifies similarity in terms of shared uncertainties, excluding the fission spectrum sensitivities, χ. A summary of results for the similarity analysis is presented in Table II. For those familiar with the c_k parameter, the only difference between the c_r parameter used in this analysis and the c_k parameter is that c_k includes comparison of shared uncertainties in the χ distribution. See Ref. [1] for additional discussion.

The analysis identified 142 critical configurations with acceptable or high similarity to Application 1. For Application 3, none of the 672 critical configurations are considered similar. This result is not too surprising given that Application 3 has 3 wt % ^{233}U mixed with depleted uranium, and there are no experiments with a similar uranium composition.

For the variant of Application 1, the infinite medium of 220 g U per liter with 9.5 M excess acid, no experiments were similar. Recall that this system has a calculated k_{eff} value of 2.055, and many of its sensitivities, especially for ^{1}H, are significantly different from those of Application 1 and of any critical experiments. The variation in similarity results highlights the importance of comparing realistic safety analysis models to the critical experiments. Using a "simple," but unrealistic, representation of the true application can lead to incorrect selection of representative critical experiments and/or lead to improper results from the validation. In practice one would not use validation to suggest that a system with a calculated k_{eff} greater than 1 is safe. This system was not subjected to further analysis.

Bias Assessment with c_r Trending

Of the three applications shown here, experiments are available only for the validation of Application 1. USLs may be generated using standard statistical techniques such as those documented in NUREG/CR-6698, *Guide for Validation of Nuclear Criticality Safety Calculational Methodology* [5] and computer codes such as USLSTATS.[6]

For Application 1, trending is performed as a function of similarity with the results extrapolated to complete similarity (i.e., c_r = 1.0). USLSTATS accounts for the extrapolation with a quadratic confidence band, where the width of the confidence band increases as the extrapolation distance from the highest c_r value to unity increases. Note that USL_2 is a closed interval approach that is not valid outside the range of the experimental data. Thus, extrapolation to c_r = 1.0 for USL_2 is not valid.

The USLSTATS calculation was performed with the 82 critical experiments with $c_r \geq$ 0.95. The USLSTATS calculations all used the input parameters below.

- Proportion of population = 0.999
- Confidence of fit = 0.950
- Confidence on proportion = 0.950
- Average standard deviation of all input k_{eff} values = -1 (directing USLSTATS to use values input for each experiment)
- Additional margin (sometimes referred to as administrative margin) = 0.02

The results of the analysis are plotted in Fig. 2 and given below.

- Computational bias, β = 1.0 %Δk/k
- Uncertainty in the bias, $\Delta\beta$ = 1.5% Δk/k
- USL_1 (disallowing positive bias) = 0.965

Addressing Validation Gaps

No critical experiments were identified that are adequately similar to Application 3. In general, when an adequate set of critical experiments is not available to validate an application, the evaluator has a limited number of options in how to establish a USL.

The evaluator may be able to locate additional critical experiments that are more similar to the application based on knowledge gained through the similarity assessment. Alternatively, the evaluator may be able to revise the application model, or the application itself, such that it is more similar to available critical experiments and provide logic in the nuclear criticality safety evaluation as to why the new application is bounding compared to the originally identified application. For example, Application 3 has a very high uncertainty due to ^{14}N, which leads to dissimilarity with critical experiments. If the negative reactivity resulting from the presence of ^{14}N is not needed, one could revise Application 3, removing the ^{14}N and potentially increasing the similarity of additional critical configurations. Then the analyst would provide logic in the criticality evaluation justifying the revised Application 3 model as a conservative approximation.

A frequently used option is to supplement the margin of subcriticality with an additional margin penalty to cover uncertainties associated with having a validation set that is not adequately similar to the application. The TSUNAMI tools can assist with quantifying and defending such a penalty. The TSUNAMI tools combine the application sensitivity profiles with nuclear data

uncertainty information contained in the covariance data files to estimate the total uncertainty on k_{eff} due to uncertainty in the nuclear data. Where only partial validation coverage is available, the uncertainty analysis can supplement traditional validation methods to establish a USL.

Additional means of bias and bias uncertainty assessment using data adjustment techniques are available in the SCALE 6 code TSURFER [7], which is described in Ref. [1].

CONCLUSIONS

The use of the SCALE TSUNAMI sensitivity and uncertainty analysis tools on some representative safety analysis models has been performed. This work is documented in ORNL/TM-2008/196.[1]

ACKNOWLEDGMENTS

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

System	Standard deviation (%)	Top six contributors and 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
220 g U per liter with 9.5 M excess acid	0.293	^{233}U n,γ to ^{233}U n,γ	0.192
		^{233}U nubar to ^{233}U nubar	0.143
		^{14}N n,p to ^{14}N n,p	0.108
		^{233}U fission to ^{233}U fission	0.081
		^{16}O n,α to ^{16}O n,α	0.070
		^{233}U χ to ^{233}U χ	0.062

Table II. Summary of similarity analysis results

Similarity index range	Similarity	Application		
		1	3	220 g U per liter with 9.5 M excess acid
		Number of experiments in each category		
$c_r < 0.1$	Low	43	54	367
$0.1 \le c_r < 0.2$	Low	80	124	15
$0.2 \le c_r < 0.3$	Low	136	141	89
$0.3 \le c_r < 0.4$	Low	140	176	14
$0.4 \le c_r < 0.5$	Low	63	79	25
$0.5 \le c_r < 0.6$	Low	30	68	101
$0.6 \le c_r < 0.7$	Low	14	30	50
$0.7 \le c_r < 0.8$	Low	7	0	10
$0.8 \le c_r < 0.9$	Marginal	17	0	0
$0.9 \le c_r < 0.95$	Acceptable	60	0	0
$0.95 \le c_r < 1.0$	High	82	0	0

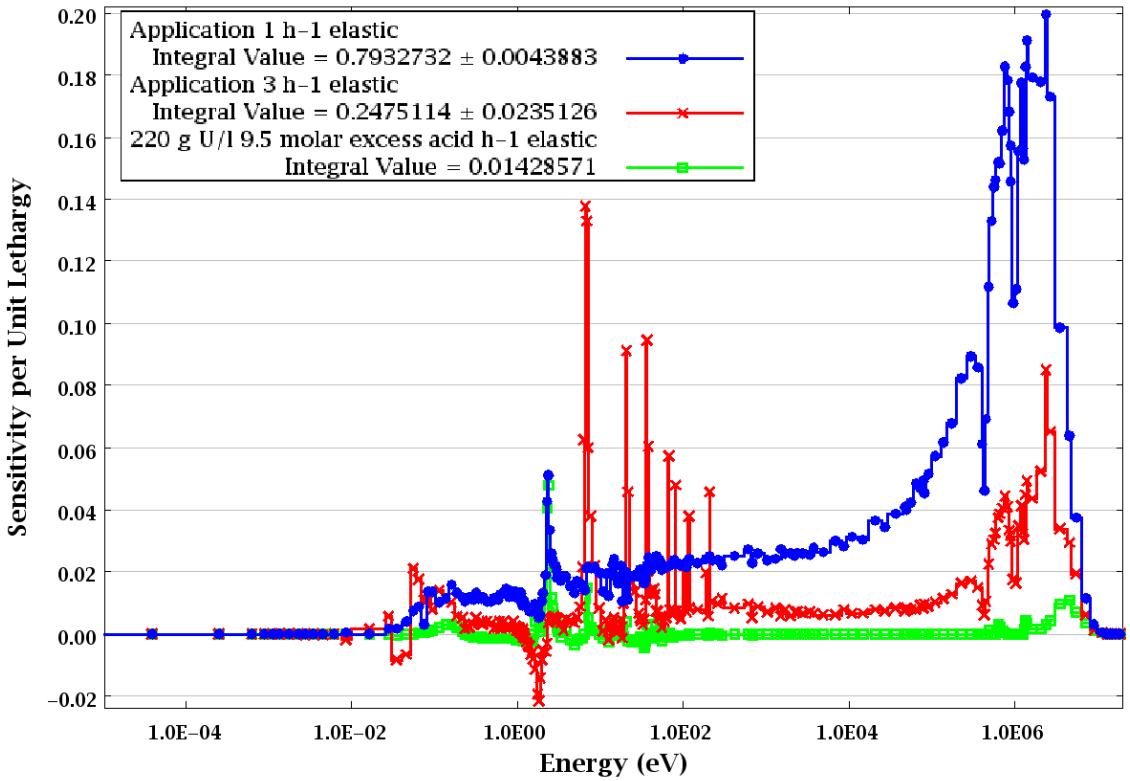

Fig. 1. Sensitivity of k_{eff} to ^{1}H elastic scattering.

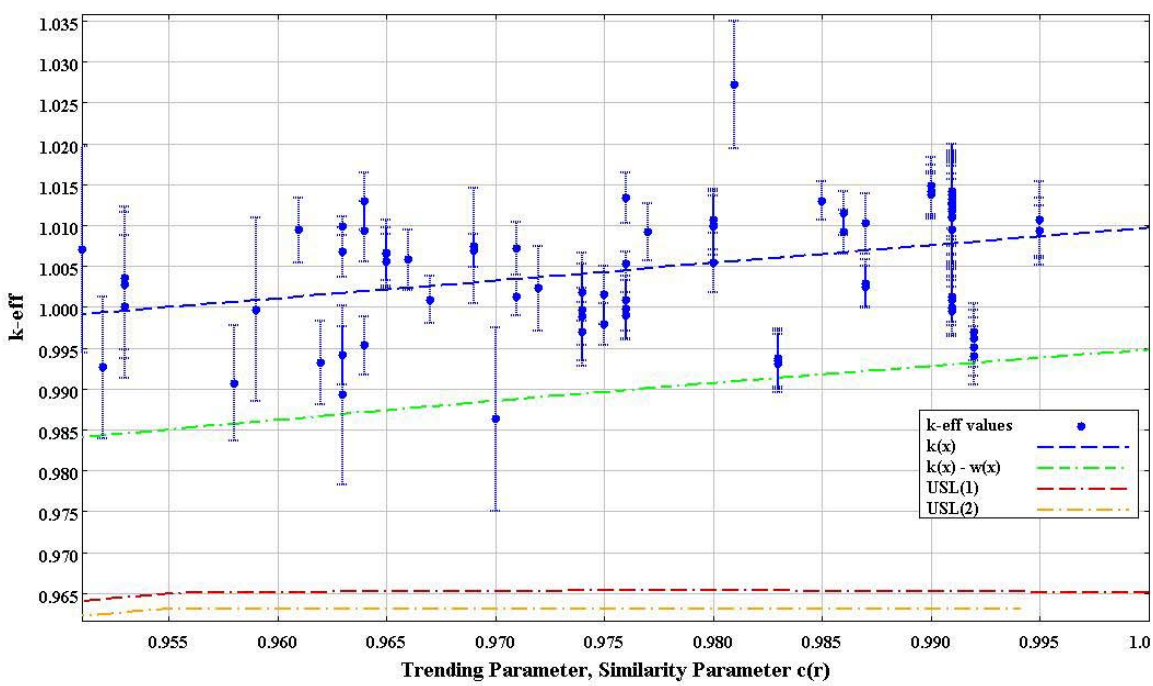

Fig. 2. USLSTATS plot for c_r trending of experiments with $c_r \geq 0.95$.