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VALIDATION OF CRITICAL PARAMETER CALCULATIONS FOR SODIUM DIURANATE USING SENSITIVITY AND UNCERTAINTY ANALYSIS

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ABSTRACT

Periodically, UF6 cylinders are cleaned and recertified for continued use. A new process being developed to reconvert the uranium recovered from the heels produces a sodium diuranate (SDU) compound, $Na_2U_2O_7$. A processing facility or transportation packaging for SDU is an application area for which no critical benchmark data exists. The validation of calculational methods described in ANSI/ANS-8.1-1998; R2007 and further amplified by ANSI/ANS-8.24-2007 requires establishment of bias by correlating the results of critical and exponential experiments with results obtained for these same systems by the calculational method being validated. These validation requirements define the area(s) of applicability as follows: the limiting ranges of material compositions, geometric arrangements, neutron energy spectra, and other relevant parameters (such as heterogeneity, leakage, interaction, absorption, etc.,) within which the bias of a calculation method is established. Benchmark data may exists for experimental systems that are different from the SDU application with respect to material composition but "look" very similar from the standpoint of other parameters, in particular, physical and spectral indices, like hydrogen-to-fissile ratio (H/X), energy of average lethargy causing fission (EALF), enrichment, solution concentration. The application of S/U techniques allows for a formal estimation of the applicability of critical benchmark experiments with different material compositions to the application area of SDU compounds. This study presents an illustrative application of sensitivity and uncertainty analysis (S/U) procedures to the validation of criticality safety calculations for sodium diurante compounds. The S/U procedures used in this study are distributed in the Standardized Computer Analyses for Licensing Evaluation (SCALE) computer software system developed at Oak Ridge National Laboratory (ORNL) system.

INTRODUCTION

Periodically, UF6 cylinders are cleaned and recertified for continued use. A new process being developed to reconvert the uranium recovered from the heels produces a sodium diuranate (SDU) compound, Na2U2O7. A processing facility or transportation packaging for SDU is an application area for which no critical benchmark data exists. The validation of calculational methods described in ANSI/ANS-8.1-1998; R2007 and further amplified by ANSI/ANS-8.24-2007 requires establishment of bias by correlating the results of critical and exponential

experiments with results obtained for these same systems by the calculational method being validated. Limits for the operation of the process are set using calculated minimum critical multiple parameters limits where enrichment is limited to 5wt% U-235. CSAS5 (KENOV.a) distributed as part of the SCALE code package is used to calculate the minimum critical mass, volume, and concentration for SDU. In addition, TSUNAMI-3D is used to calculate sensitivity coefficients for each application and critical experiment, and TSUNAMI-IP is used to compare the application to benchmarks and calculate a bias using USLSTATS. There are no critical experiments in the thermal energy range that include sodium, therefore, a noncoverage penalty calculated by TSUNAMI-IP is included in the bias calculation.

ANALYSIS METHODS

SCALE 6.1 [1] TSUNAMI tools TSUNAMI-3D and TSUNAMI-IP were used to generate keff sensitivity data for the applications and 50 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 low uranium enrichment, solution configurations from International Handbook of Evaluated Criticality Safety Benchmark Experiments [2] evaluations. Upper subcritical limits (USLs) were generated for the systems based on trending of the TSUNAMI similarity parameters.

Sensitivity Coefficients (TSUNAMI)

The SCALE code uses TSUNAMI-3D to calculate sensitivities, which are combined into one large sensitivity data file that is used to assess the similarity between desired applications and experiments using TSUNAMI-IP. Sensitivities are calculated for every appropriate reaction in different energy groups for each nuclide included in the experiment or application. The sensitivity is a measure how sensitive keff is to uncertainties in the cross sections. The sensitivities are used to quantify the similarity between an application and critical experiments in terms of nuclide, reaction, and energy group. The accuracy of the sensitivity coefficients calculated in the TSUNAMI is confirmed using direct perturbations. The sensitivities of the nuclides deemed “most sensitive” by TSUNAMI, i.e. the nuclides with the greatest sensitivity, should be confirmed by direct perturbations on the atom densities of those nuclides. This is necessary because the sensitivities calculated by TSUNAMI may be inaccurate due to insufficient meshing, lack of convergence for the forward or adjoint solutions, or a flux order moment that is too low.

Correlation Coefficient, ck (TSUNAMI-IP)

Sensitivity data files for the applications and each of the experiments are used to calculate the correlation coefficients known as ck values. This is an integral indices that is used to assess the similarity of uncertainty weighted sensitivity profiles between the application and the experiments. The ck values provide a comparison between the application and each individual experiment. These values are normalized such that a value of 1 indicates that the two systems that are identical, and a value of 0 indicates systems that the systems are completely different from each other. The correlation coefficient indicates the percentage of uncertainty that is common between the two systems, i.e. ck =0.7 indicates 70% common uncertainty between the two. For experiments to be considered applicable for comparison and validation, a ck value of at least 0.8 is required, while 0.9 is preferred. TSUNAMI-IP can calculate these correlation

coefficients, along with some other integral indices that can be useful when comparing systems. With at least ten experiments with a ck value that meet the acceptance criteria, TSUNAMI-IP will calculate an noncoverage penalty. This penalty should be added to the administrative margin in order to account for areas where there is insufficient data from experiments to make a complete comparison to the application. In other words, when the benchmarking experiments do not fully cover the uncertainty in the cross sections used in the application, there must be some penalty added on to account for that gap in validation.

Bias Trending Analysis (USLSTATS)

While the use of trending analysis in criticality validation is not a new concept, the method utilized in this validation was developed with the use of TSUNAMI-IP for validation. Typically, trending analysis has been done using descriptive parameters such as energy of average lethargy of fission (EALF), moderator-fuel-ratios (H/X or H/U), enrichment, or poison concentration. However, after establishing correlation coefficients between an application and a set of benchmarking experiments, a bias can be established by trending keff against these ck values. USLSTATS, which is provided in the SCALE code package, can be used to perform the trending analysis on the ck values. A user can input values for the administrative margin and penalty, and USLSTATS will output plots showing the trending curves establishing a bias, as well as an upper safety limit.

RESULTS

Fifty critical experiments were selected from the International Handbook of Evaluated Criticality Safety Benchmark Experiments (IHECSBE) for low enriched uranium, solutions, and thermal energy spectra represented the systems considered. None of the critical experiments include sodium in the uranium fuel material. A slab, cylinder, sphere, and infinite homogeneous systems were used to calculate minimum critical dimensions that can be used to define safe geometries for the uranium recovery process. TSUNAMI-3D using KENO-Va is used to calculate the forward and adjoint flux solutions needed to calculated the sensitivity coefficients for both the one-dimensional systems and critical experiments. The system characteristics are provided in Table I.

Sensitivity data files for each of the systems is compared to the critical experiment using TSUNAMI-IP and an integral index calculated to characterize the applicability of the critical experiment to the systems considered. A summary of the correlation coefficients is provided in Table II.

The similarity of the selected critical experiments to each application was evaluated using the ck parameter, which is a correlation coefficient that quantifies similarity in terms of shared uncertainties, including the fission spectrum sensitivities. A summary of results for the similarity analysis is presented in Table II.

A large number of critical experiments show similarity to the applications, i.e. ck values greater than 0.9, with exception of the concentration. The lower ck values are due primarily to the differences in the H/X between the application and critical experiments. Calculation of minimum critical concentration occurs at large H/X where there not many large volume solution critical experiments. The H/X of the 50 critical experiments ranged from 469 to1438. Critical

experiments with H/X greater than about 1000 had ck values less than 0.9 for the slab, cylinder and sphere minimum critical dimension calculations. The critical experiments with the large H/X had ck values greater than 0.7 for the minimum critical concentration calculation. A plot of the comparing the sensitivity 1H total cross section for minimum critical concentration application to an experiment with the lowest had ck values, i.e. 0.61, is presented in Figure 1. Most of the dissimilarity for total hydrogen cross section sensitivity is due to the 1H absorption cross section.

Table I. System Case Characteristics

Parameter	Slab thickness	Cylinder diameter	Sphere diameter	Concentration
Minimum Critical Value	29.638 cm	14.543 cm	41.455 l	282.1 gU/l
^{235}U (g/l)	75.5	67.8	67.8	14.1
^{238}U (g/l)	1435.0	1287.5	1287.5	268.0
Na (g/l)	146.0	131.0	131.0	27.3
H/X	237	277	277	1739
EALF (eV)	0.08295	0.07516	0.07607	0.03405
^{23}Na capture sensitivity	-0.0124 ± 2.85E-6	-0.0123 ± 2.99E-6	-0.0123 ± 3.24E-6	-0.00802 ± 1.47E-6

Table II. Correlation Coefficients

Application	Number of Critical Experiments		
	c_k **< 0.8**	**0.8** $\geq c_k \leq$ **0.9**	c_k **> 0.9**
Slab	0	6	44
Cylinder	2	7	41
Sphere	2	9	39
Concentration	42	6	2

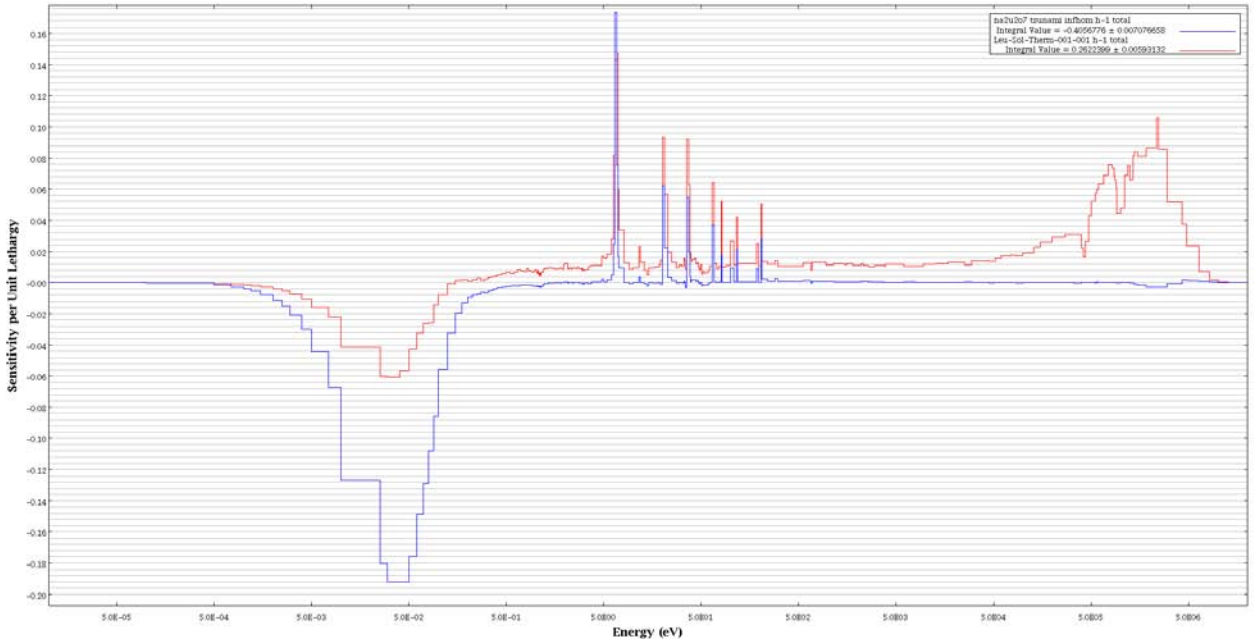

Figure 1. Sensitivity of 1H total reaction for minimum critical concentration

The margin of subcriticality is supplemented with an additional margin penalty to cover uncertainties associated with having a validation set that is not adequately similar to the application. There are no low enriched uranium, solution, and thermal energy spectra critical experiments that contain sodium in the fuel or other materials. The TSUNAMI tools can quantify a penalty for this gap in the validation. 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 keff due to uncertainty in the nuclear data. The penalty for non-coverage of 23Na is presented in Table III.

For the application of USL to calculating minimum critical parameters, trending is performed as a function of similarity with the results extrapolated to complete similarity (i.e., ck = 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 ck value to unity increases. Note that USL2 is a closed interval approach that is not valid outside the range of the experimental data. Thus, extrapolation to ck = 1.0 for USL2 is not valid.

The results of the analysis are plotted in Fig. 2 for the minimum critical concentration and given below. The number of experiments with ck > 0.9 was insufficient to perform a parametric analysis; therefore the criteria was relaxed to ck > 0.7 to include sufficient critical experiments to demostrate the USLSTATS method for bias assessment.

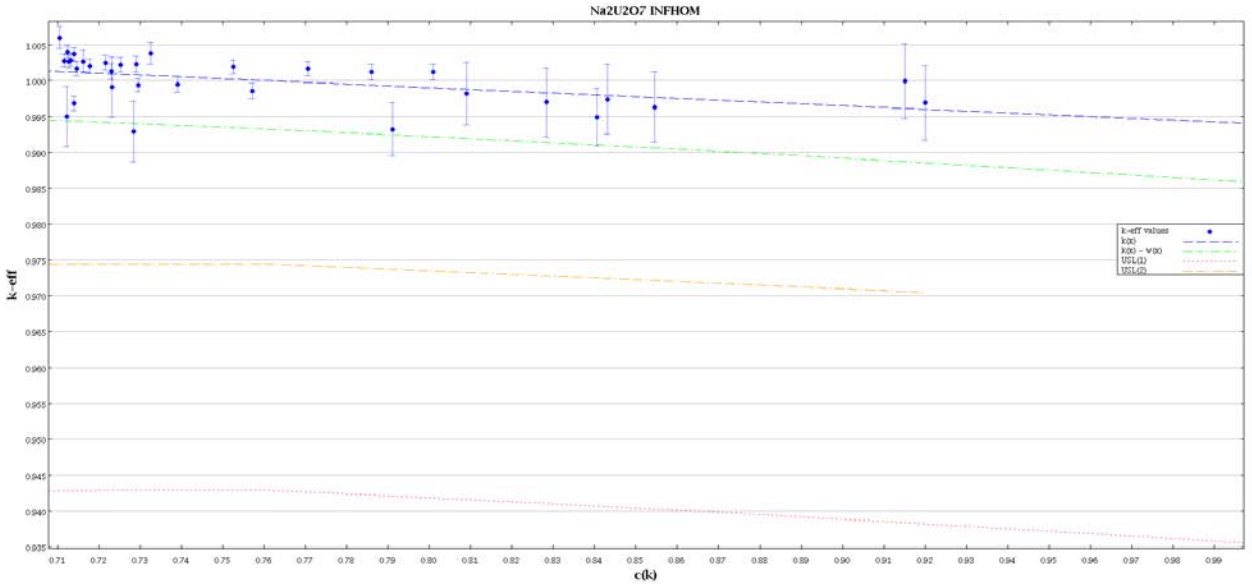

Figure 2. USLSTATS bias trending with c_k value for minimum critical concentration

The calculated keff values for the application cases have been adjusted by applying a penalty for not having complete coverage by the benchmarks. Calculated keff values for the application cases, along with the adjusted keff values, are listed in Table III. The adjusted keff is the calculated keff increased by a penalty value. This penalty is computed by multiplying the value of the portion of the application sensitivity that is not covered by any benchmarks by the uncertainty in the keff of the cask due to 23Na capture cross-section uncertainties. The penalty due to noncoverage by the benchmarks (i.e., the penalty due to the application not being in the area of applicability of benchmarks completely) is used as an additional subcritical margin in licensing calculations.

Table III. USL and Penalty Summary

	USL_1	**Penalty Δk**	**Adjusted USL_1**
Slab	0.9405	4.0E-4	0.9409
Cylinder	0.9407	6.4E-4	0.9413
Sphere	0.9402	8.8E-4	0.9410
Concentration	0.9352	3.4E-4	0.9355

CONCLUSIONS

TSUNAMI tools can be used to perform a more quantitative analysis for validation. The sensitivity data provides a useful comparison of applications to critical experiments based on

specific nuclides, reactions and energy groups. Bias assessment with ck trending extrapolates the benchmark to complete similarity with the application rather than relying on physical or spectral indices. Another option is to calculate the minimum critical parameters by removing the 23Na from the application cases. This would be conservative, but is not necessary if the penalty for noncoverage is quantified. For calculation with SDU the dissimilarity is due to the differences in H/X and not the lack of 23Na in the critical experiments. The penalty due to noncoverage of 23Na capture cross sections is small, with a maximum value that is less than 0.1% in keff. Therefore, it is concluded that although sufficient benchmark experiments did not exist to provide coverage for all design scenarios, the potential impact of the noncoverage on the criticality safety of the uranium recovery process is minimal.

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