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Application of Sensitivity/Uncertainty Methods to Burnup Credit Validation

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Abstract. The responsible use of calculational methods in nuclear criticality safety includes a determination of bias and bias uncertainty that may exist between the calculated results and reality. Such biases exist due to approximations used to model the real world, uncertainties in nuclear data, and approximations associated with the calculational method (e.g., Monte Carlo method). The bias and bias uncertainty are typically determined by using the modeling approximations, nuclear data, and calculational method to model well-known, usually critical, systems. Unfortunately, the bias and bias uncertainty determined in this manner can vary significantly depending on the characteristics of the known "benchmark" systems. The most accurate determination of bias and bias uncertainty is obtained by using benchmark systems that are very similar to the real-world operational configuration, the subcriticality of which must be safely ensured. Historically, similarity has typically been determined using comparisons of gross integral parameters (e.g., lethargy of average energy of neutrons causing fission and hydrogen to fissile nuclide ratio) and on qualitative comparisons of the geometry and materials present in the benchmark and application systems. The development of sensitivity/uncertainty methods permits detailed quantitative comparison of these systems. The work presented in this paper is a sensitivity/uncertainty-based study of the similarity or applicability of many critical experiment models to a model of a high-capacity transportation cask that is loaded with spent commercial nuclear fuel and flooded with water. This paper includes descriptions of the sensitivity/uncertainty methods used, the operational configuration of interest, benchmark critical configurations used for comparisons, and discussion of the results from the sensitivity/uncertainty analyses.

1. Introduction

Historically, criticality safety analyses for commercial light-water reactor spent nuclear fuel (SNF) storage and transportation casks have assumed the SNF to be fresh (unirradiated) with uniform isotopic compositions corresponding to the maximum allowable enrichment. This fresh-fuel assumption provides a simple bounding approach to the criticality analysis and eliminates concerns related to modeling the fuel operating history. However, because this assumption ignores the decrease in reactivity as a result of irradiation, it is very conservative and can result in a significant reduction in SNF capacity for a given storage or cask volume. Numerous publications, an extensive set of which is listed in the reference section of NUREG/CR-6800 [1], have demonstrated that increases in SNF cask capacities from the use of burnup credit can enable a reduction in the number of casks and shipments and thus have notable financial benefits while providing a risk-based approach to improving overall safety. The concept of taking credit for the reduction in reactivity due to irradiation of nuclear fuel (i.e., fuel burnup) is commonly referred to as burnup credit. The reduction in reactivity that occurs with fuel burnup is due to the change in concentration (net reduction) of fissile nuclides and the production of parasitic neutron-absorbing nuclides (nonfissile actinides and fission products). The work presented in this paper used sensitivity/uncertainty (S/U) analysis to explore the potential applicability of critical experiments to validation of burnup credit calculations. The S/U analysis was performed using TSUNAMI-3D [2] sequence and TSUNAMI-IP [3] module from SCALE 5 [4].

TSUNAMI-3D is a Monte Carlo–based eigenvalue sensitivity analysis sequence that was released with SCALE 5. This software tool permits energy-, mixture-, nuclide- and region-dependent examination of the sensitivity of the system k_{eff} to variations in nuclear data of modeled materials.

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TSUNAMI-3D uses first-order linear-perturbation theory to produce sensitivity coefficients. As such, the sensitivity coefficients are valid only for small perturbations.

TSUNAMI-IP uses sensitivity data generated by TSUNAMI-1D and/or TSUNAMI-3D and cross-section uncertainty data to generate several relational parameters and indices that can be used to determine the degree of similarity between two systems. The sensitivity profiles generated for a particular system of interest may be compared with the sensitivity profiles for critical experiments used to generate subcritical limits in validation studies. Such comparisons enable the analyst to reach conclusions regarding the adequacy or applicability of critical experiments used in the validation study.

Work is being performed at Oak Ridge National Laboratory (ORNL) to generate recommendations and develop computational methods related to taking credit for the in-reactor burnup of commercial nuclear fuel during out-of-reactor storage and transport. The work reported in this paper involved an evaluation of the applicability of more than 1000 critical configurations to the validation of criticality calculations for a high-capacity rail cask loaded with 32 spent pressurized-water reactor (PWR) fuel assemblies.

2. Applicability of benchmark critical experiments

The complexity of many systems having criticality accident potential necessitates heavy reliance on computer calculations to establish the safety of the system under normal and upset conditions. One of the responsibilities of the safety analyst using computer calculations is to use validated computational methods to determine the system multiplication factor (k_{eff}) and the maximum k_{eff} considered safely subcritical. Typically, validation is performed by using a computational method to calculate the k_{eff} for a set of applicable critical experiments. A computational method is defined by the modeling approximations, the computer code, computer code input options, and the nuclear data used. The analyst performing the validation then uses the calculated k_{eff} values and associated uncertainties and the critical experiment measured k_{eff} and uncertainties in a statistical analysis to determine the bias and bias uncertainty for the computational method. This bias quantifies the relationship between the calculated and actual k_{eff} values for a modeled system.

For any given computational method, the values of the bias and bias uncertainty can vary significantly depending on geometry and materials present in the modeled critical experiments. For example, the computational method bias for an array of fuel rods may be significantly different if the system is dry as compared to the bias for a similar array flooded with water. Consequently, the most accurate values for bias and bias uncertainty for use in a criticality analysis are determined using critical experiments that are similar to the calculations used in the criticality analysis. It would not be appropriate to use dry, high-enriched uranium critical experiments to determine a bias and bias uncertainty for a safety analysis of an optimally moderated low-enrichment uranium system. The bias developed in this way is a total bias that combines the biases resulting from individual bias sources. For example, the total bias may include, among many others, a bias resulting from the modeled presence of fission product ^{103}Rh and a bias related to hydrogen scattering. The size and sign of such biases vary depending upon the neutronic environment of the modeled systems. These biases may be of opposite sign, thereby partially compensating for each other. Critical experiments may also contain extra features or materials not present in the criticality analysis model, also referred to in this paper as the application. The extra biases associated with the features present only in the experiments may cancel out other biases that are present in both the experiment and application, thus hiding a real bias that should be applied to the application. The calculated partial biases vary with conditions. A bias calculated for a fission product using experiments that include only that fission product or include it in a system that is rather different than the application may not be correct. Neutron energy spectrum shifts, associated with the presence of other fission products or other materials, may significantly affect the bias associated with the fission product of interest.

A conclusion drawn from this discussion is that it is important to include only critical experiments that are similar to the evaluation case when determining bias and bias uncertainty that are appropriate for the evaluation case. Sensitivity/Uncertainty tools developed at ORNL and distributed as part of the SCALE 5 package permit a detailed, quantitative comparison of modeled systems. This comparison can be used to determine how similar a critical experiment model is to an application.

3. Sensitivity/uncertainty analysis tools and concepts

TSUNAMI-3D calculates total and partial sensitivity coefficients for various neutron interactions with each nuclide in each region. A sensitivity coefficient is the relative impact of a change in some nuclear data (e.g., Σ_a) on the system k_{eff} and is defined as $S_\alpha = (dk/k)/(d\alpha/\alpha)$, where α is the nuclear data of interest.

In some cases, these sensitivity coefficients may be further broken down into explicit and implicit components. The explicit component results from the sensitivity of k_{eff} to variation of the resonance self-shielded macroscopic cross section. The implicit component results from cross-section adjustments in the resonance self-shielding calculations. For example, the explicit sensitivity of hydrogen in the moderator around a fuel pin results directly from the sensitivity of k_{eff} to changes in the hydrogen cross section. The implicit sensitivity includes the effects of the sensitivity of the fuel macroscopic cross sections to changes in the moderator cross sections. The implicit component is calculated using derivatives produced during problem-dependent cross-section processing.

The TSUNAMI-3D sequence uses KENO V.a to perform forward and adjoint calculations. Then the SAMS program uses the forward and adjoint solutions in a standard linear perturbation theory method to produce neutron energy-dependent sensitivity profiles. The profiles for each modeled system are saved into a sensitivity data file (SDF).

TSUNAMI-IP, also a part of the SCALE 5 computer software package, is used to compare the sensitivity data for two systems. It generates a variety of total and partial relational parameters that quantify the similarities between the two systems. The work reported in this paper utilizes the c_k parameter. The c_k parameter is a single-valued parameter used to assess similarity of uncertainty-weighted sensitivity profiles for all nuclide-reactions between a design system and a criticality experiment. The value of c_k varies between zero, for two completely dissimilar systems, and 1.0, for two identical systems. The premise behind the c_k parameter is that biases are primarily due to cross-section data with larger uncertainties. Systems that demonstrate similarly high sensitivities to highly uncertain cross section data will have similar computational biases. The current guidance based on experience at ORNL is that a critical configuration is applicable to an evaluation case if the c_k value is ≥ 0.9, a critical configuration is considered marginally applicable if c_k is ≥ 0.8 and <0.9, and a critical configuration is not applicable if $c_k <0.8$.

Another use of sensitivity data is to evaluate "coverage". Coverage for a specific nuclide and cross section is a measure of whether a critical configuration is at least as sensitive to change in the cross section as is the application. Figure 1 below shows that the ^1H total sensitivity for critical configuration LEU-COMP-THERM-050, case 18, from the *International Handbook of Evaluated Criticality Safety Benchmark Experiments* [5] (IHECSBE); it covers the sensitivity for the GBC-32 for most of the energy range. It also shows that there is significant noncoverage in the 0.1- to 10-eV range. For a fully covered sensitivity profile, the blue curve in Fig. 1 would completely cover the red curve.

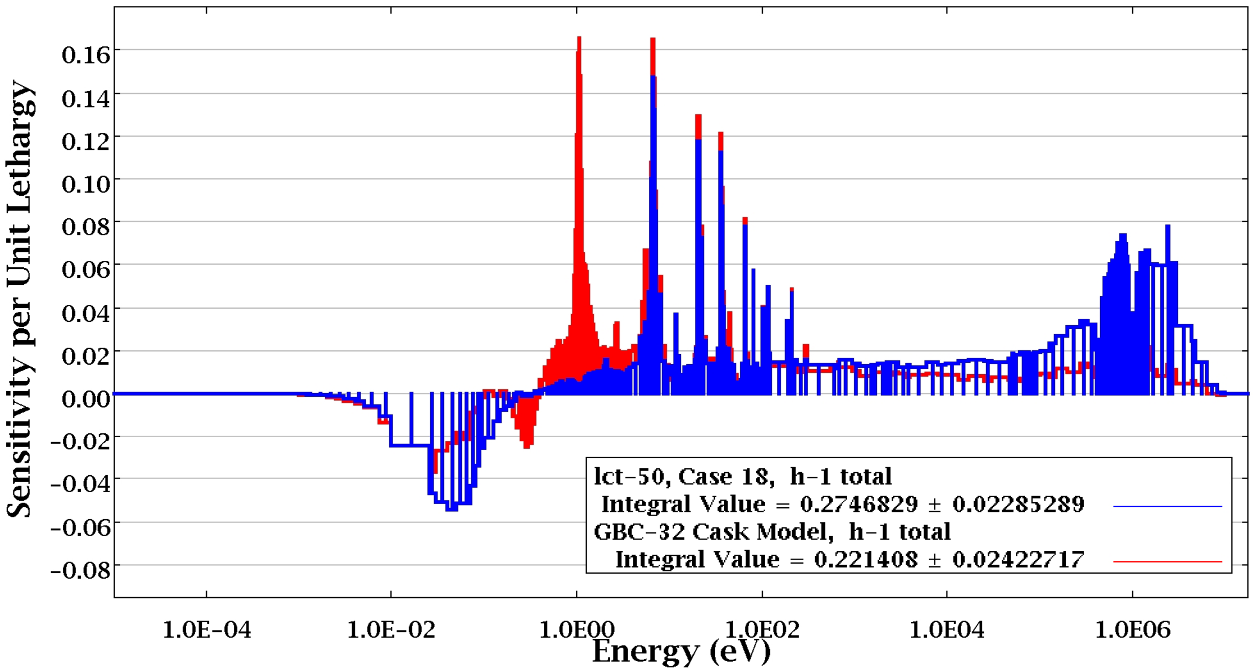

FIG. 1. The two curves show the sensitivity of k_{eff} to changes in the total hydrogen macroscopic cross sections for a burnup credit cask model (red curve) and for a critical experiment model (blue curve). "Coverage" is provided by the experiment wherever the blue curve bounds the red curve.

4. Burnup credit cask model and methods

A generic cask model with a 32-PWR assembly capacity was previously developed and is described in NUREG/CR-6747 [6]. This model, referred to as the GBC-32, was created to serve as a computational benchmark. The features of the GBC-32 include 32 cells with 365.76-cm-tall and 19.05-cm-wide Boral (0.0225 g $^{10}B/cm^2$) panels between and on the external faces of each cell. The cell walls are constructed of stainless steel having inner dimensions of 22 by 22 cm and are spaced on 23.76-cm centers. The cells sit 15 cm above the bottom of a stainless steel cask having an inner radius of 87.5 cm and internal height of 410.76 cm. The radial thickness of the side walls is 20 cm, and the cask bottom and lid are 30 cm thick. Figure 2 shows a half-cask model with the top removed.

For purposes of the analyses documented in this paper, the cask was modeled as loaded with Westinghouse 17 by 17 Optimized Fuel Assemblies (W17×17OFA). The dimensions for the W17×17OFA were taken from Table 3 of Ref. [6]. The interior of the cask was modeled as filled with water.

The fuel had an initial enrichment of 4 wt % ^{235}U and was burned to 40 GWd/MTU. The STARBUCS [7] sequence in SCALE 5 was used to generate 18 axial location-dependent burned fuel compositions. The STARBUCS sequence and available input parameters are discussed in Ref. [7]. The normalized burnup profile from Table 5 of Ref. [6] was used. The fuel burnup was modeled at a power density of 40 MW/MTU for 1000 d, with a postshutdown cooling period of 5 years. The fuel burnup calculations model the depletion of the ^{235}U and the in-growth of actinide and fission product nuclides. From the depletion calculations, fuel compositions for the following nuclides were retained for the criticality calculations: ^{234}U, ^{235}U, ^{236}U, ^{238}U, ^{237}Np, ^{238}Pu, ^{239}Pu, ^{240}Pu, ^{241}Pu, ^{242}Pu, ^{241}Am, ^{243}Am, ^{95}Mo, ^{99}Tc, ^{101}Ru, ^{103}Rh, ^{109}Ag, ^{133}Cs, ^{147}Sm, ^{149}Sm, ^{150}Sm, ^{151}Sm, ^{152}Sm, ^{143}Nd, ^{145}Nd, ^{151}Eu, ^{153}Eu, and ^{155}Gd.

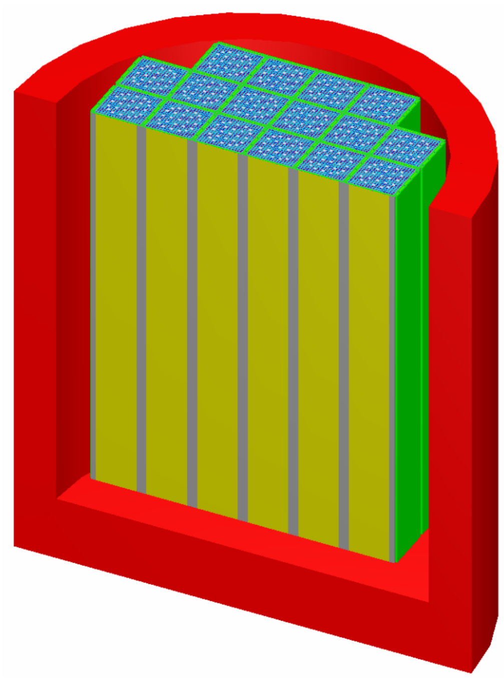

FIG. 2. GBC-32 cask model.

Sensitivity analysis for the GBC-32 cask was performed with TSUNAMI-3D and was checked using direct perturbation calculations. As is noted in the SCALE documentation, the TSUNAMI-3D version distributed with SCALE 5 will not work if the model has more than 50 nuclides that have resonance information. Each occurrence of the same resonance nuclide in multiple mixtures is counted toward this limit. The GBC-32 cask model included different mixtures of uranium, plutonium, other actinides, and 15 fission products in each of 18 axial zones. There were in excess of 500 resonance nuclides in all mixtures. Consequently, special development versions of some of the TSUNAMI-3D sequence programs were created by the SCALE development staff to support this work. The 50-resonance-nuclide limitation will be removed in SCALE 5.1. However, users will need to keep this 50-nuclide limit in mind during model development with SCALE 5.

5. Comparison with critical experiments

Sensitivity analyses have been performed by ORNL staff for more than 1,000 critical configurations. The sensitivity data files have been accumulated as a resource for identifying critical configurations that may be useful for validation studies. The critical configurations are primarily from the IHECSBE [5]. At the time this work was presented at the International Atomic Energy Agency (IAEA) meeting in London, the collection of sensitivity data files included 170 ^{233}U, 150 high-enrichment uranium (HEU), 4 intermediate-enrichment uranium (IEU), 256 low-enrichment uranium (LEU), 197 Pu, 201 mixed-oxide (MOX), and 156 MOX configurations from the French Haut Taux de Combustion (HTC) experiment series.

The French HTC experiment data were purchased from the French under an agreement that limits release of the information. This series was designed to support validation of actinide-only burnup credit. The uranium and plutonium compositions of the rods were selected to be consistent with PWR U(4.5 wt %)O_2 rods with 37,500 MWd/MTU burnup. The French categorized these experiments into four groups. The first group was 18 configurations involving a single square-pitched array of rods with rod pitch varying from 1.3 to 2.3 cm. The arrays were flooded and reflected with clean water. The second group was 41 configurations that were similar to the first group except that the water used as moderator and reflector included either boron or gadolinium in solution. The third group was 26 configurations with the rods arranged into four assemblies that were arranged in a 2×2 array. The spacing between assemblies was varied, and some of the assemblies had borated steel, Boral®, or cadmium plates attached to the sides of the four assemblies. The fourth group was 71 configurations similar to the group 3 configurations except thick steel or lead shields were placed around the outside of the 2×2 array of fuel assemblies.

One of the primary objectives of the work reported in this paper was to evaluate the applicability of the HTC experiments to burnup credit calculations.

The TSUNAMI-IP code from SCALE 5 was used to compare the sensitivity data file for the GBC-32 cask with the sensitivity data files for 1134 critical configurations. The results from this comparison are presented in Fig. 3.

The critical configurations are grouped by type of fissionable material. The trend of c_k values with fissionable material composition appears reasonable. The GBC-32 model has LEU mixed with Pu. The nearly zero c_k values for ^{233}U configurations show that these configurations have very little in common with the GBC-32. The HEU, IEU, and LEU have ^{235}U, but no Pu. Consequently, their c_k values are higher than for the ^{233}U configurations but are still significantly below the 0.8 cut-off value for being considered marginally applicable. The MOX critical configurations are the most similar to the GBC-32 model. Figure 4 shows a closer look at the MOX data from Fig. 3 and includes some annotations identifying specific sets of critical configurations.

The results for the HTC MOX configurations show that these configurations are generally very similar to the burned fuel in the GBC-32 cask model. The HTC configurations with lower c_k values are from HTC group 2, and all have a significant quantity of gadolinium dissolved in the moderator/reflector. The amount of gadolinium present far exceeds that present in the burned fuel as a fission product. The results from the S/U analyses confirm the value of the HTC MOX critical configurations for validation of burnup credit calculations. The set of MOX critical configurations considered in this study is not complete. There are additional configurations in the IHECSBE [5] not considered in this study that are expected to be at least marginally applicable.

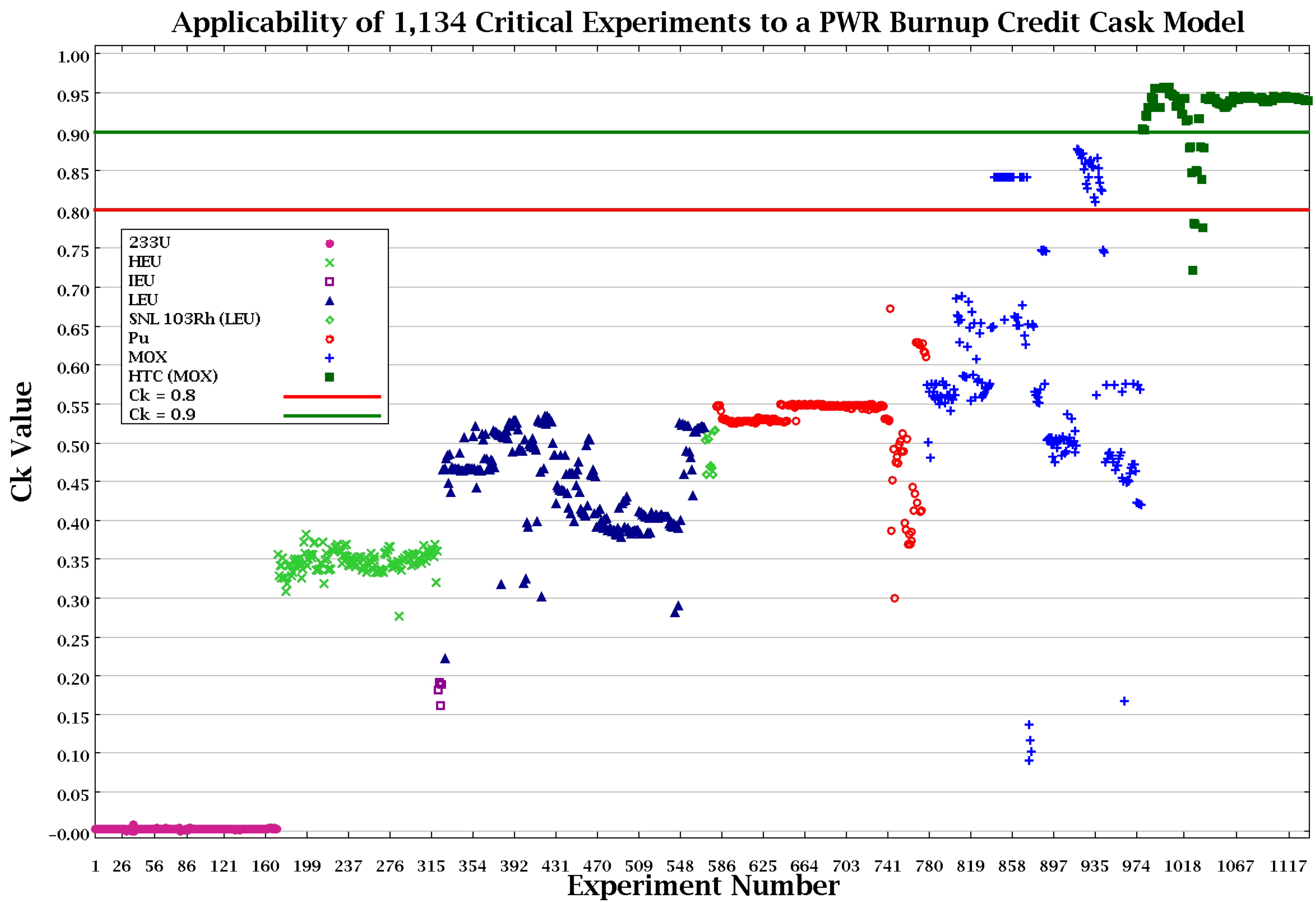

FIG. 3. Sensitivity/Uncertainty analysis results.

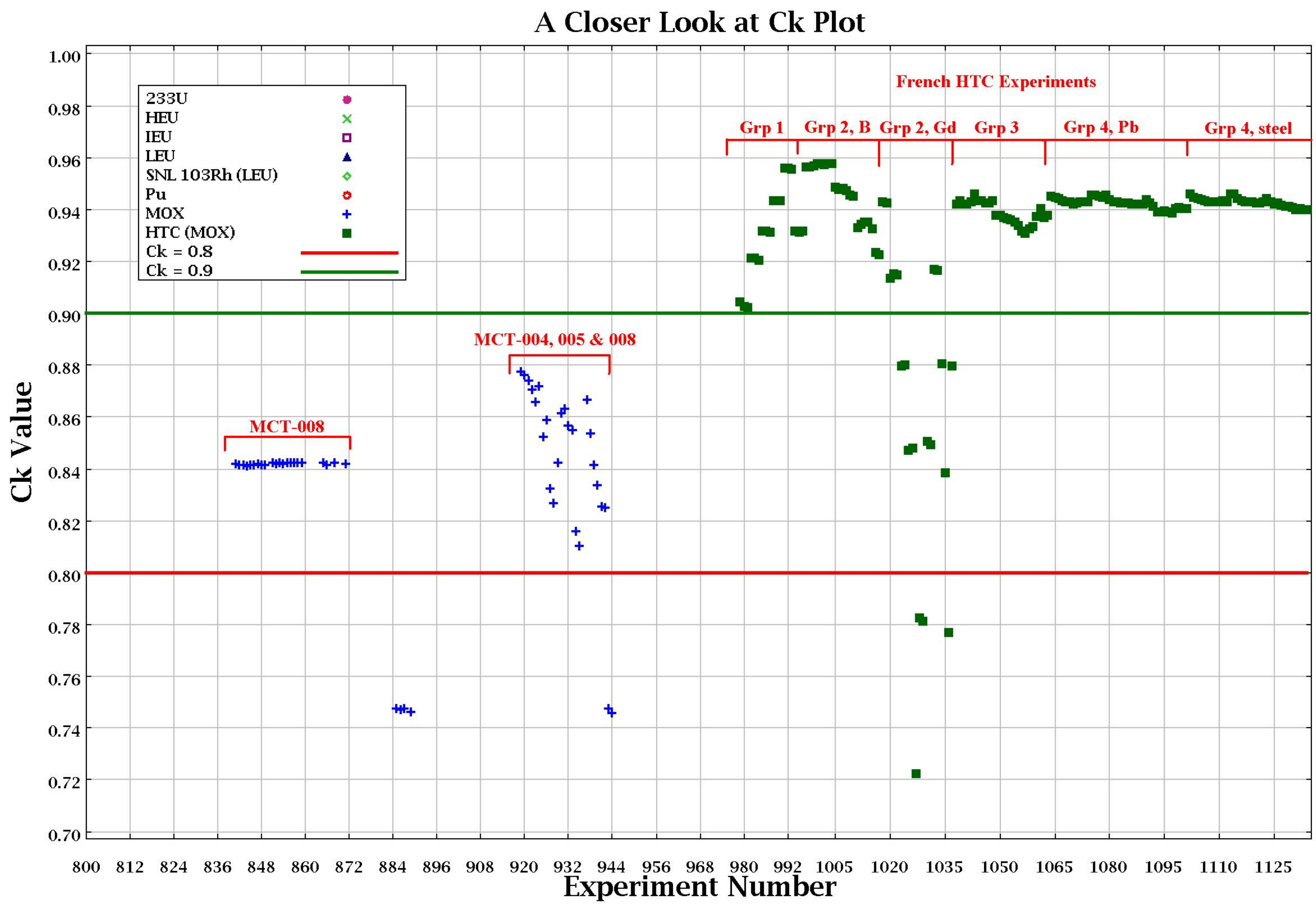

FIG. 4. Closer look at S/U analysis results.

Of the 1134 critical configurations considered, 937 had c_k values lower than 0.8 and, by the guidelines presented earlier in this paper, would not be considered applicable to the validation of burnup credit cask calculations. Table I below presents the S/U analysis results for the 1134 critical configurations considered. Of the 978 non-HTC experiment configurations, none were identified as applicable ($c_k \geq 0.9$), 45 configurations were marginally applicable ($0.9 > c_k \geq 0.8$), and 933 were not applicable. Of the 156 HTC configurations, 143 were applicable, 9 were marginally applicable, and 4 were not applicable.

Table I. Sensitivity/uncertainty analysis results for 1134 critical configurations

	^{233}U	HEU	IEU	LEU	Pu	MOX	HTC	All
$c_k \geq 0.95$	0	0	0	0	0	0	11	11
$c_k \geq 0.9$	0	0	0	0	0	0	143	143
$c_k \geq 0.8$	0	0	0	0	0	45	152	197
$c_k < 0.8$	170	150	4	256	197	156	4	937
Total	170	150	4	256	197	201	156	1134

5.1. Coverage

An additional consideration is whether all of the nuclides in an application are represented in the critical configurations. If they are not present in the experiments, any bias associated with the missing nuclides will not be present in the bias calculated using the critical experiments. If the missing nuclides are a significant contributor to the k_{eff} of the application, the c_k values for the experiments should as a result be lower. If they do not contribute significantly to the application multiplication factor, then any associated bias should be insignificant, too. The degree to which the experiments cover the sensitivities for a nuclide is referred to as "coverage." If the experiment or group of experiments have sensitivity profiles that are at least as large as those for the application at all neutron energies, the application is considered covered by the experiments.

The most important fission product for burnup credit is ^{149}Sm. With current S/U methods, critical experiments should have a c_k value of at least 0.9 and provide coverage for all significant nuclides. In this context, significant means that the presence of the nuclide has a statistically significant effect on the application k_{eff} value. Of the 1134 critical configurations evaluated for applicability, none containing ^{149}Sm had a c_k value greater than or equal to 0.8. However, the critical configurations did include cases 8 through 18 of LEU-COMP-THERM-050 from the 2004 IHECSBE [5], which include ^{149}Sm in solution in a tank in the middle of an array of LEU fuel rods. These critical configurations are experiments 560 through 570 in Fig. 3. Figure 5 shows the ^{149}Sm sensitivity profiles for 11 cases from LCT-050 and for the GBC-32 cask loaded with fuel burned to 40 GWd/MTU. The thick black curve shows the total sensitivity curve for ^{149}Sm in the GBC-32 cask. The other curves in Fig. 5 show the ^{149}Sm sensitivity profiles for cases 8 through 18 of LCT-050. Note that the LCT-050 ^{149}Sm curves show that the experiments are at least as sensitive to the presence of ^{149}Sm as is the GBC-32 cask. Also note that the magnitude and shape of the LCT-050 curves are similar to those from the GBC-32. If the c_k value had been high enough, the user could have some confidence that any bias caused by the presence of ^{149}Sm would be adequately included in the overall bias. However, since the c_k values of the LCT-050 configurations are around 0.45, a ^{149}Sm bias calculated using the LCT-050 configurations may not be appropriate for the GBC-32. If the LCT-050 configurations were modified to contain fissionable materials, moderators, and absorbers more similar to the GBC-32, thus yielding a higher c_k value, the ^{149}Sm bias calculated from the modified experiments might be significantly different.

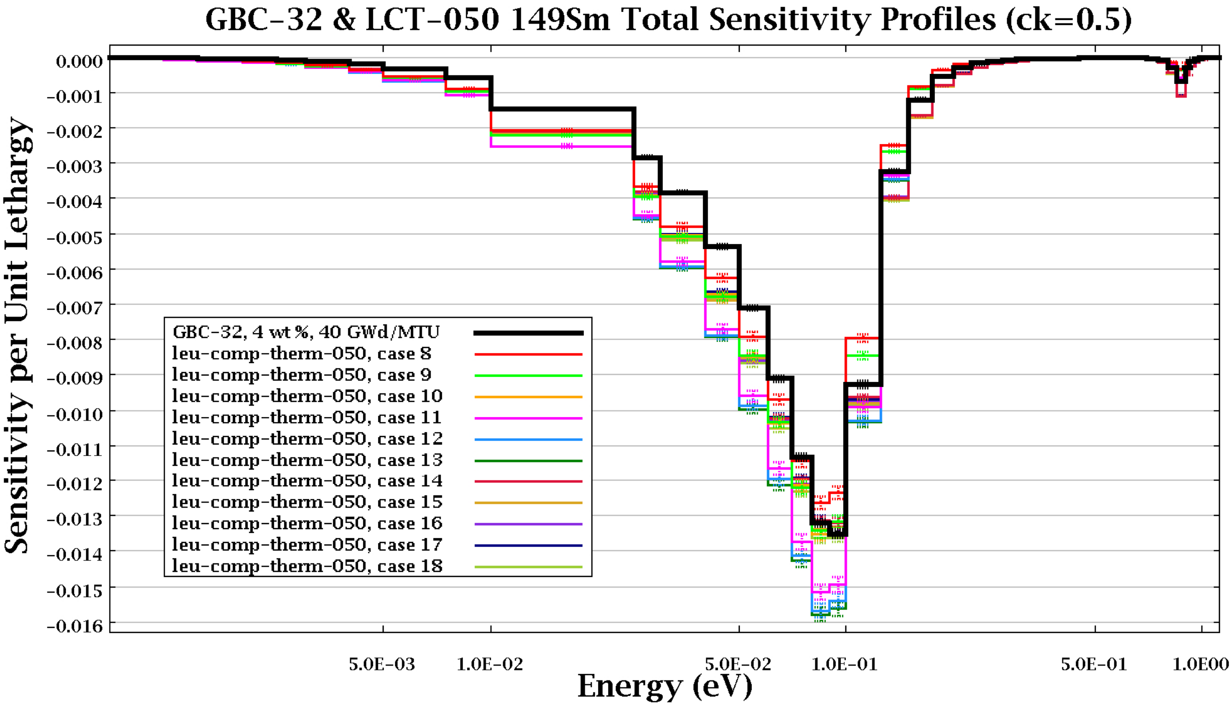

FIG. 5. ^{149}Sm sensitivity profile coverage.

The next coverage example examines the ^{155}Gd present in the GBC-32 and in some of the French HTC experiments. Note from Fig. 4 that the c_k values for the group 2 Gd experiments vary significantly from the non-Gd HTC experiments. From the S/U analysis, the c_k values decreased as the Gd concentration increased. Figure 6 shows the ^{155}Gd sensitivity curves from the GBC-32 and from a subset of the HTC group 2 Gd solution experiments. The figure shows that the sensitivity profiles from the HTC group 2 Gd configurations completely cover the ^{155}Gd sensitivity profile from the GBC-32 model. The peak sensitivity for the HTC group 2, Gd case 8 configuration is nearly 4 times as great as the peak sensitivity for the GBC-32. Consequently, the ^{155}Gd bias calculated using this experiment could be significantly different than a bias calculated using experiments having more similar ^{155}Gd sensitivity profiles.

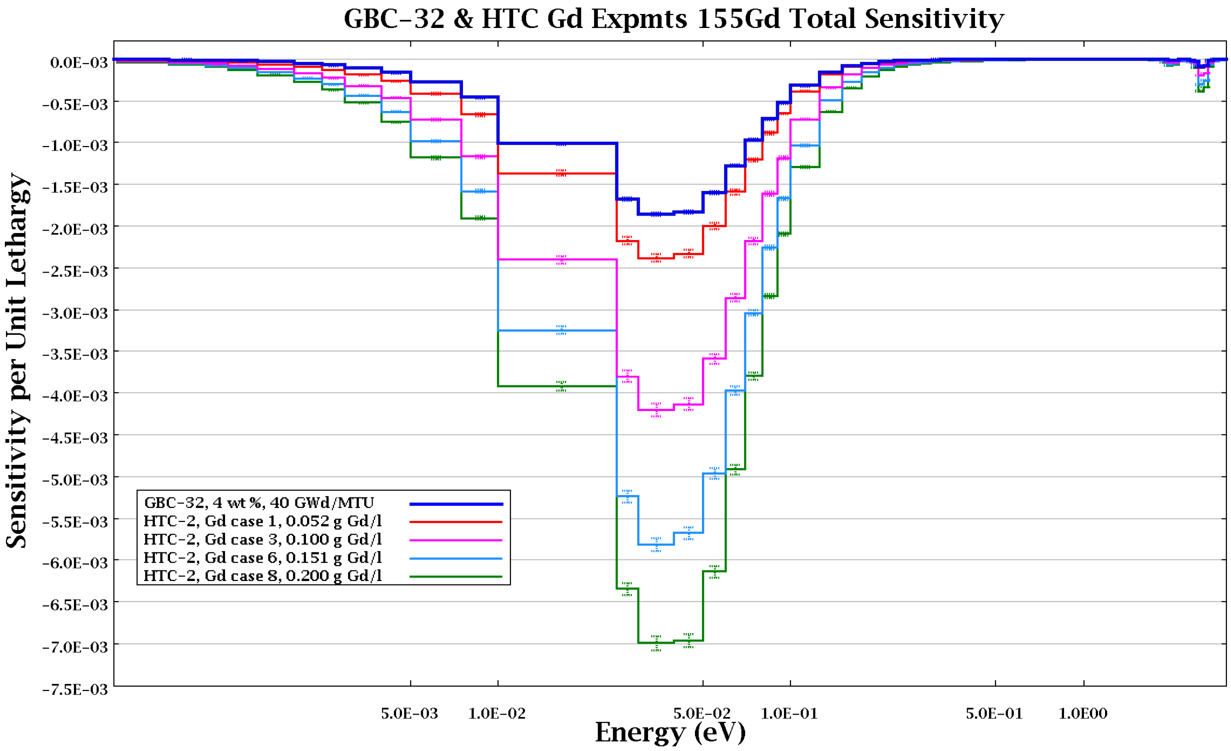

FIG. 6. ^{155}Gd sensitivity profile coverage.

As a final example of how coverage may be used, let us examine the ^{103}Rh sensitivity profiles from the GBC-32 model and a series critical experiments performed at Sandia National Laboratories (SNL). These experiments, shown as a separate group in Fig. 3, are documented in the 2005 version of the IHECSBE [5] as evaluation LEU-COMP-THERM-079. These experiments were designed to support validation of ^{103}Rh in burnup credit applications. The experiments have thin (25-, 50- or 100-microns thick) ^{103}Rh foils stacked between the U(4.32 wt %)O_2 pellets in some of the rods in a water moderated and reflected triangular pitched (2.0- or 2.8-cm) array. Figure 7 shows the ^{103}Rh sensitivity profiles for the GBC-32 and four of the SNL ^{103}Rh experiments. Generally, the ^{103}Rh coverage provided by the SNL experiments is good, except in the 1- to 2-eV range. In this neutron energy range it appears that the GBC-32 model is significantly more sensitive than the experiments. Consequently, it is not clear that a bias calculated using these experiments would be correct. Note that for the SNL experiments, the sensitivity around the 1- to 2-eV range increases with decreasing foil thickness. This indicates that even thinner or more lightly loaded ^{103}Rh foils might produce a more similar sensitivity profile. Based on these observations, conceptual critical experiment models were developed using thinner foils. The resulting sensitivity profiles are shown in Fig. 8. This figure shows that 5-micron thick foils in the modified experiment model produced a ^{103}Rh sensitivity profile that is very similar to the GBC-32 cask model ^{103}Rh sensitivity profile.

This exercise demonstrated how the S/U tools could be used to design critical experiments that are intended to support a specific application, such as a high-capacity burnup credit cask model. The S/U tools were not available when the SNL ^{103}Rh experiments were designed and conducted. The S/U tools will be used to evaluate and, where appropriate, modify future SNL fission product experiment designs. This will maximize the usefulness of the experiments and maximize the return on the experiment sponsor's investment.

Due primarily to the absence of Pu in the SNL ^{103}Rh experiments, the c_k values generated by comparing the experiments to the GBC-32 cask model were around 0.5 and would not be considered "applicable" for a traditional calculation of bias and bias uncertainty. Methods based upon

Generalized Linear Least-Squares Methods [8] are being developed at ORNL to utilize the relevant information from experiments with relatively low c_k values for validation.

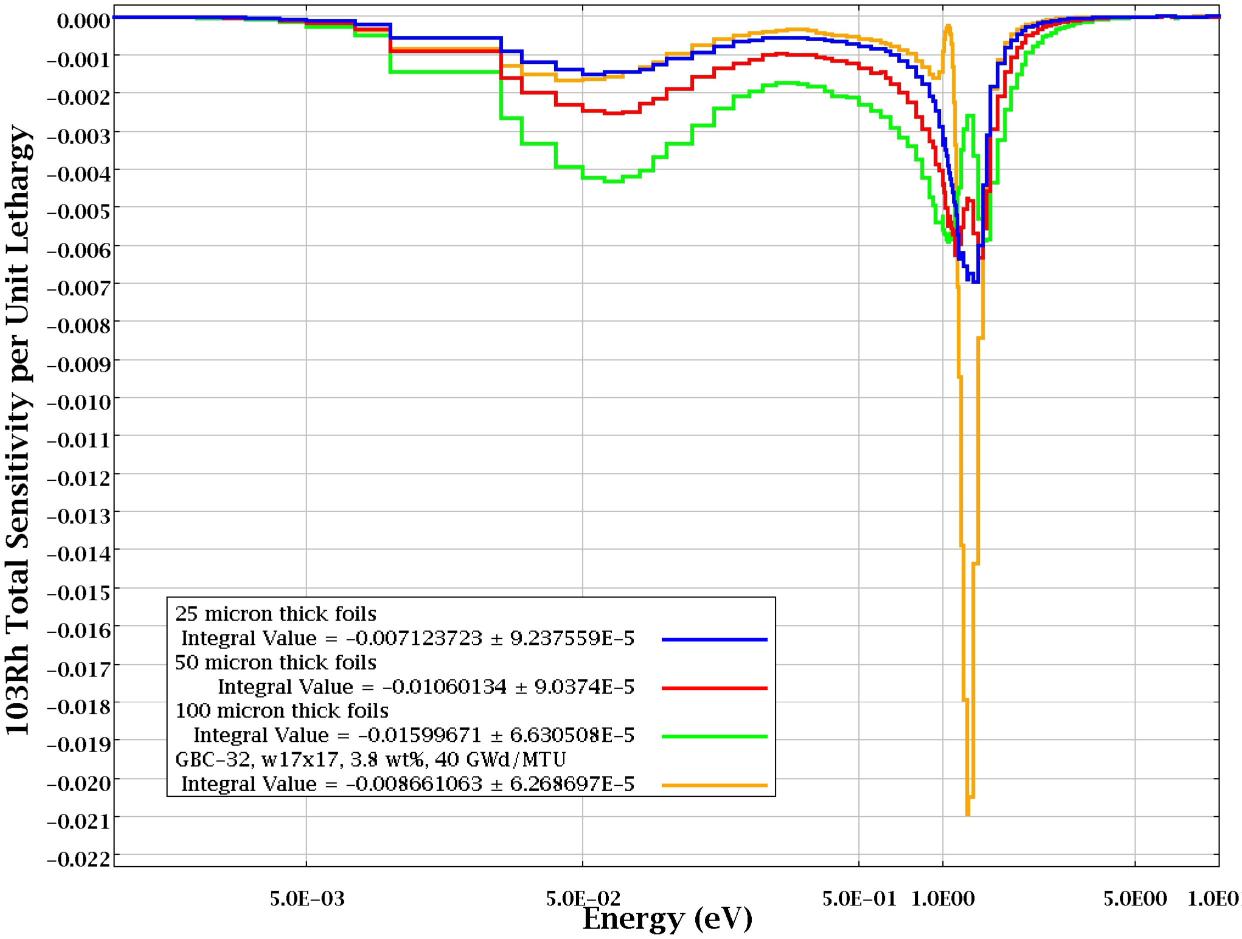

FIG. 7. GBC-32 and SNL experiment ^{103}Rh sensitivity profiles.

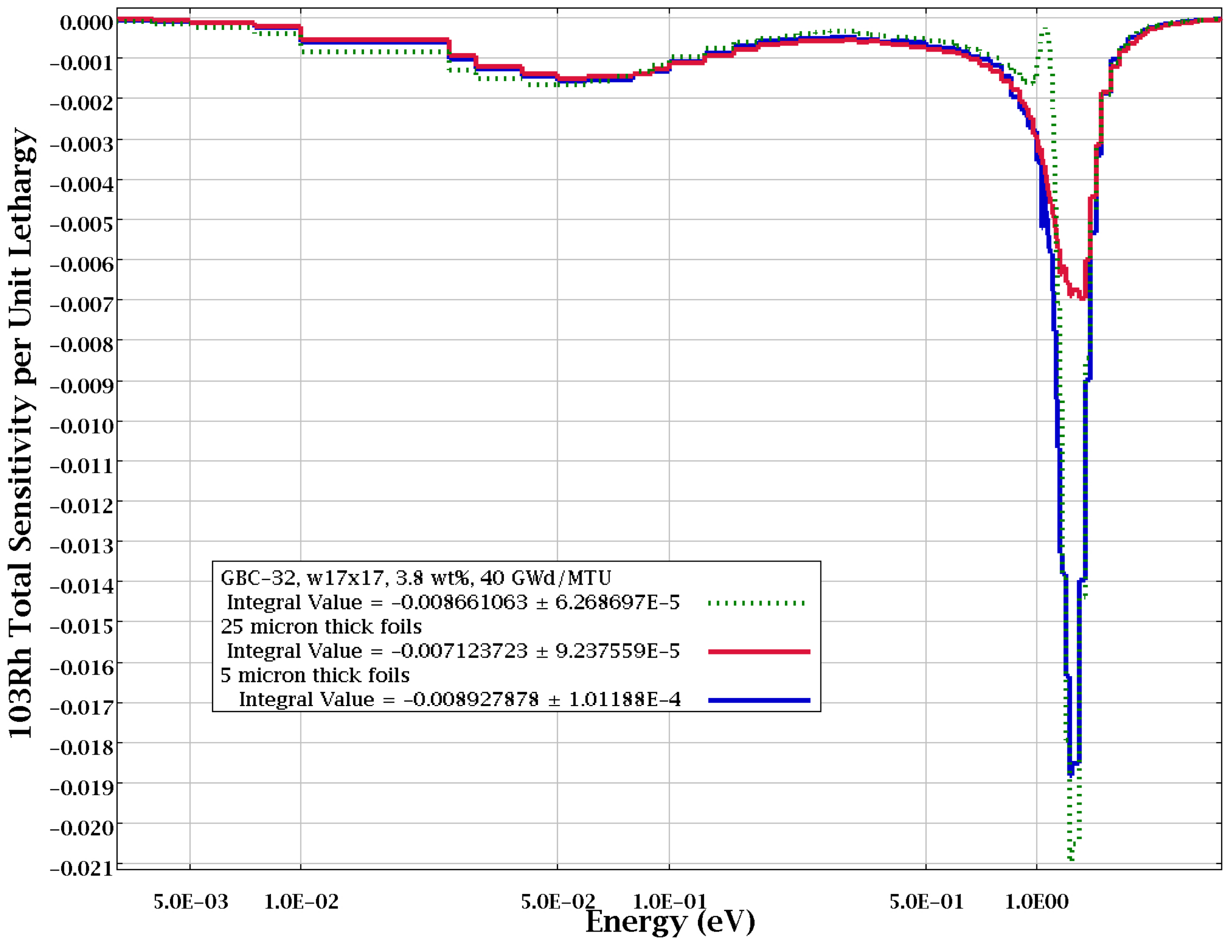

FIG. 8. ^{103}Rh sensitivity profiles with modified SNL ^{103}Rh experiment.

6. Conclusions

The objective of the work reported in this paper was to use S/U analysis to identify critical configurations that could be used to validate burnup credit calculations. Sensitivity calculations have been performed on a generic burnup credit cask and on 1134 critical configurations, 156 of which are from the French HTC series. The S/U analysis shows that 45 of the non-HTC critical configurations are marginally applicable for validation. The analyses also show that 143 of the 156 HTC experiments are applicable for validation, and an additional 9 HTC critical configurations are marginally applicable. The HTC experiments provide an excellent source of data for validating burnup credit calculations.

Discussion of the concept of coverage was included in this paper and an example of how S/U tools could be used in the review and design of critical experiments.

Future S/U analysis work will include evaluation of French critical experiments with fission products, evaluation of casks loaded with spent boiling water reactor fuel, design and evaluation of future SNL fission product critical experiments, and application of S/U tools to commercial reactor criticals and to the REBUS experiment [9].

7. Acknowledgment

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