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Criticality Safety Applications of S/U Validation Methods

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1. Introduction

This paper applies sensitivity and uncertainty (S/U) methods described in the previous papers in this session[1-3] to example validation exercises for a series of criticality safety application areas. These application areas were taken from a sampling of Department of Energy (DOE) sites. While not intended to be fully documented validation sets, these results are designed to illustrate the methods and procedures for areas that would be difficult to validate using traditional methods. Situations where validation is difficult using standard methods include: (1) unusual moderator or reflector materials are present with little or no support from critical experiments, (2) presence of two or more fissile materials in ratios where few experimental criticals exist, and (3) poisoned systems where the poison is not well characterized.

The first step in any validation exercise is to establish a benchmark database of critical experiments. This work has built a set of criticals that span several application areas: low-enriched ^{235}U (LEU) systems, high-enriched ^{235}U (HEU) systems, intermediate-enriched ^{235}U (IEU) systems, plutonium systems, mixed plutonium and uranium systems, and ^{233}U systems. An added value of the S/U approach is the possibility of including many benchmark types and allowing the procedure to automatically pick only those experiments that are determined to be applicable. A set of 419 benchmark systems is analyzed via the S/U approach in this work.

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Application of the S/U validation approach to four criticality safety scenarios is discussed in this paper. These systems include a mixed Pu/U/Fe waste tank system, a U(10)SiO_2 system, a U(100)SiO_2 system, and a PuSiO_2 system. These applications are derived from areas currently under study at DOE facilities.

2. Description of Benchmark Dataset

The key to any criticality safety data validation procedure is the generation of a database of critical benchmark experiments that typically covers a broad range of systems that are in some way similar to the particular application(s) of interest. This section will briefly describe the 419 critical benchmark systems included in this analysis. These 419 systems can be broken into several groups, which include low-enriched ^{235}U (LEU) systems, high-enriched ^{235}U (HEU) systems, intermediate-enriched ^{235}U (IEU) systems, plutonium systems, mixed plutonium and uranium systems, and ^{233}U systems. This list is not intended to be exhaustive, but does contain experiments that cover all of the major fissile isotopes, as well as a full range of enrichment and moderation.

LEU Systems - This group consists of 168 experiments, including solution systems, fuel pin lattices, and solid oxide and fluoride systems moderated by water, paraffin or sterotex.

HEU Systems - This group consists of 75 experiments, and includes solution systems, metal systems, and uranium hydride systems.

IEU Systems - This group includes twenty experiments with an intermediate ^{235}U enrichment. There are eleven systems with uranium metal, and 9 systems with either UO_2 or UF_4.

Pu Systems - Seventy eight plutonium critical experiments are included in this set of benchmarks. This set includes solution systems, metal systems, and oxide systems.

Mixed Pu/U - Seventy six mixed plutonium and uranium systems are included in this set of benchmarks. These experiments involve solution systems, fuel pin lattices, solid mixed-oxide systems moderated by polystyrene, and one mixed metal sphere.

^{233}U Systems - Two systems containing ^{233}U fuel.

3. S/U and GLLSM Analysis Examples

The ultimate goal of these procedures is a "simple" approach which either utilizes the S/U-based parameters described in Refs. 1 and 2 as a trending tool or an automated GLLSM (Generalized Linear Least Squares Method, Ref. 3) approach in which the detailed GLLSM input requirements are minimized. The techniques that will be studied in this section include the use of trending with the S/U-based c_k parameters under two different assumptions as compared with the use of GLLSM. The two assumptions for the c_k trending are the use of the entire benchmark set (419 experiments) as opposed to trending with only those experiments that have a c_k value greater than 0.65. The value of 0.65 is chosen arbitrarily to include values outside of 0.8–1.0 range (systems considered to be similar) but not inclusive of all systems. Comparisons of these three methods will allow for overall conclusions to be made regarding their performance.

Generally, the GLLSM procedure is the most rigorous and should be the most accurate. However, the procedure is limited by the quality and quantity of the data available. Hence, the completeness parameter, R, will be used to judge the adequacy of the benchmark dataset used in the GLLSM procedure. The completeness parameter, R, is defined as follows:

$$R = S_a / S_t, \text{ where } S_t = \sum_j \sum_i \sum_x S_{xaij}, \; S_a = \sum_j \sum_i \sum_x d\, S_{xaij},$$

$$\text{and} \quad d = \begin{cases} 1, \text{ if } N_{ix} > 9 \\ 0, \text{ if } N_{ix} \leq 9 \end{cases}$$

$$N_{ix} = \text{number of systems for which } S_{xeij} > 0.9 * S_{xaij},$$

N_{ix} = number of systems for which $S_{xeij} > 0.9 * S_{xaij}$,

Where e refers to experiment, a refers to application, S_{xij} is the sensitivity of k_{eff} to the cross sections of the constituent material nuclides j, and i and x are the indices by energy group and nuclide/reaction. Analyses are planned but have not been completed to determine the magnitude of the R parameter, such that completeness is assured. Until then, the R value is useful only in relative comparisons, i.e., the higher the number the better the quality of the GLLSM bias prediction.

Hanford waste tanks

Three application scenarios were considered for the Hanford waste tanks. The scenarios correspond to the base case (base), contingency of a factor of three less iron than planned (Fe), and a contingency of a factor of three more plutonium than the base case (Pu). Models taken from the summary documentation[4] consist of infinite homogeneous materials corresponding to limiting conditions.

These models were used in the SEN1 code[5] to generate sensitivity coefficients corresponding to the predicted changes in k_{eff} due to cross section changes over all groups. Additionally, a database of sensitivity profiles was generated for each of the 419 benchmark experiments described in Section 2 above. These benchmark experiments were processed with either the SEN1 or SEN3[6,7] sensitivity modules. These resulting sensitivity profiles were subsequently analyzed with the CANDD code[7] to generate values of c_k for each of the three applications to each of the benchmark experiments.

In Fig. 1, the calculated k_{eff} values of each of the 419 benchmark experiments are trended versus its value of c_k for the Hanford Tank Base case. Three different estimates of the bias for this case are shown in the figure. These estimates are obtained from the intersection of the solid line (all c_k values) and dotted line (c_k values above 0.65 only) with the c_k of unity line, and the rectangular box also shown for $c_k = 1$ (GLLSM result). The predicted bias from the dotted line is clearly much different from the other two predicted values, which are in general agreement with each other considering each has an uncertainty of about 0.5 % in k_{eff}. The c_k trending method for obtaining estimates of the bias is also useful in that the applicability of the benchmark set to this problem can be easily seen from the plot shown. In this case, the criterion of 15–20 systems with a c_k value greater than 0.8 is not met. Thus, the guidelines indicate that neither the solid nor the dotted line should be trusted for c_k values near unity. The reliability of the GLLSM result can now be estimated by noting the value of R, the completeness parameter.

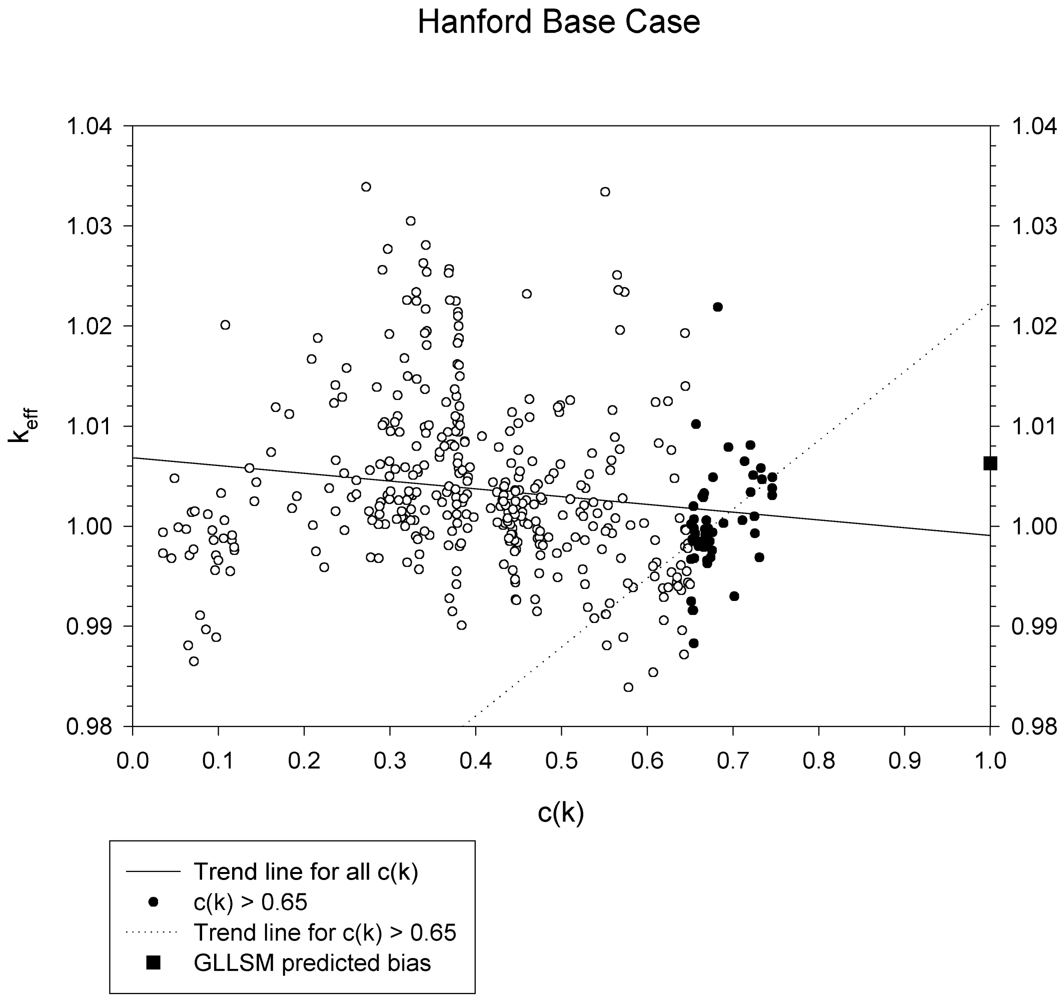

Figure 1. Trend plot of k_{eff} versus c_k for Hanford Base Case.

As stated earlier, an acceptable value of the completeness parameter has not yet been determined. For this initial study, a value of 0.8 is assumed to be acceptable. This value is chosen to match the similarity criterion, which is based on a c_k value of 0.8. The value of R for the Hanford Tank Base case to the set of 419 benchmarks is 0.81, which is just above the acceptable level. This example very nicely shows the advantage of the GLLSM technique combined with the completeness parameter. Validation is possible using the generalized trending procedure, GLLSM, when the other methods appear to be unacceptable.

Certainly situations exist where the set of available benchmarks is not felt to be adequate. For these systems steps must be taken to ensure the subcriticality of the defined application. Typically, under these circumstances a value for the upper subcritical limit (USL) that is much lower than expected is chosen. The value of USL that was obtained from the analysis of this case was 0.90.[4] Certainly this is a value that is quite low. However, under the circumstances that existed for this problem, it is likely a prudent choice. Clearly, based on the GLLSM result given above (corresponds to a USL of about 0.98) it is extremely conservative, but it does show the steps that must be taken when sufficient experimental data is judged to be not available to justify a higher USL value.

Similar analyses for the two contingency cases, labeled Fe and Pu along with the Base case, are given in Fig. 2. The results for the Fe case indicate that the 3 methods are very consistent with each other. Indeed, the criterion of about 10-20 cases with c_k values of 0.8 or higher are met for the Fe case. It is indeed encouraging that the two methods of trending with the c_k values (c_k greater than 0.65 and all c_k values) agree with each other quite well. Also, the GLLSM result should be reliable since the value of the completeness parameter, R, for this case is 0.90. The final case, labeled Pu, does not meet the similarity criterion for c_k values, however the trend with all c_k values and the GLLSM result compare very well. The value of R for this case is 0.84, which indicates acceptable reliability of the GLLSM result.

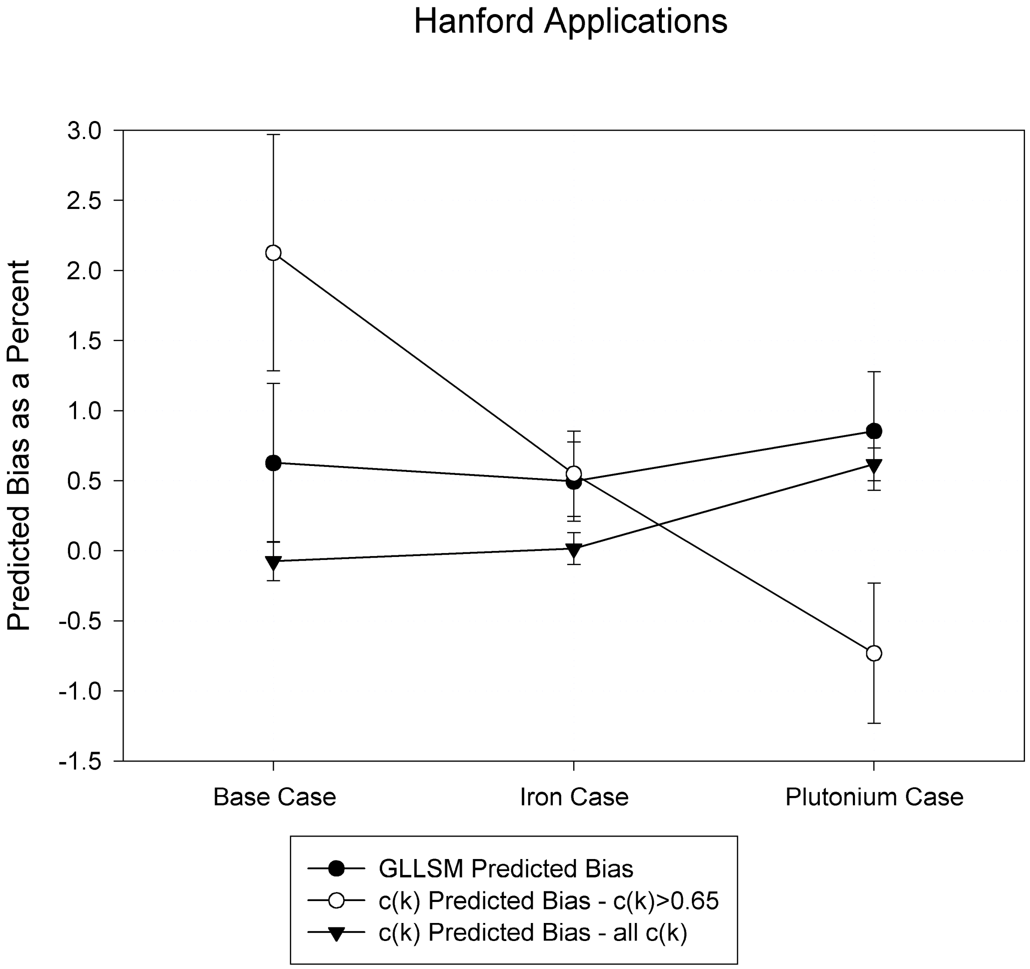

Figure 2. Bias predictions for three Hanford Tank Applications.

In summary, it can be seen that for the Fe case, where the c_k criterion is meet, the three methods all give very comparable results. For the other two situations, the GLLSM and the trending with all c_k values appear to agree quite well with each other.

Application to U(10)-SiO_2 Systems

This series of applications are designed to model a wide range of conditions that could exist in a low-level waste facility containing a maximum of 10 wt % U-bearing materials. The models for these applications were generated via a critical radius search for the conditions shown in Table 1.

Table 1. Specifications for U(10)-SiO_2 Systems[a]

	SiO2-1	SiO2-2	SiO2-3
Sphere radius (cm)	311.071	778.157	154.42
Si/X	2084	1355	818
H/X	219	1140	1351
	SiO2-4	SiO2-5	SiO2-6
Sphere radius (cm)	249.714	31.5479	24.0187
Si/X	29	20	15
H/X	0	22	34
	SiO2-7	SiO2-8	SiO2-9
Sphere radius (cm)	84.6371	30.66	24.2359
Si/X	12	8	5
H/X	0	11	17

[a] Each system was surrounded by a 4-m-thick SiO_2 + variable H_2O reflector, where the H_2O concentration matches the core concentration, see Ref. 8 for further details.

In the same manner as the Hanford tank problem previously, these models were used in the SEN1 code to generate sensitivity coefficients corresponding to the predicted changes in k_{eff} due to cross section changes over all groups. These sensitivity profiles were then processed with the CANDD code to generate values of c_k for each of the 9 applications to each of the 419 benchmark experiments.

The results of c_k trending and GLLSM for all nine of these systems are shown in Fig. 3. The SiO2-1 system has an H/X value of 219, which indicates that it is well moderated. Experience has shown that for well-moderated systems, typically the primary contributors to the system sensitivities are the fissile and moderating nuclides. Thus, the large number of similar systems (209) was expected. Similarly, the good agreement between the three different bias prediction procedures was also expected based on the large number of systems (108) with c_k values greater than 0.9. The trend plots for cases SiO2-2 and SiO2-3 are not shown but follow the general trends of the first case since these systems are also highly moderated. However, the value of the completeness parameter, R, for the SiO2-1 case was a quite surprising value of 0.68. A large number of systems with c_k values above 0.9 should be indicative of a very high value of R. The exception could be a high sensitivity nuclide with a very low uncertainty associated with it. Upon checking the sensitivities for the SiO_2 systems, it was noted that the silicon capture sensitivities were quite large, i.e., –0.29, –0.17, and –0.10, respectively. Thus the uncertainties for these systems would *have* to be small in order for the c_k values to be appropriate. The Si covariance files were then examined and it was discovered that the capture (MT=102) component of the covariance was omitted from the file (the file contained MT=1, 2, 4, 51, 62, 103, and 107). Work is currently underway to add the MT=102 component to the covariance file for silicon. Caution on the use of the results for these first three SiO_2 cases is urged, since the addition of the MT=102 component could have a large impact on the results for these systems. Note that the remaining systems, i.e., SiO2-4 through SiO2-9 are not expected to suffer from this limitation, since the largest silicon capture sensitivity for these systems is –0.02 which corresponds to system SiO2-5.

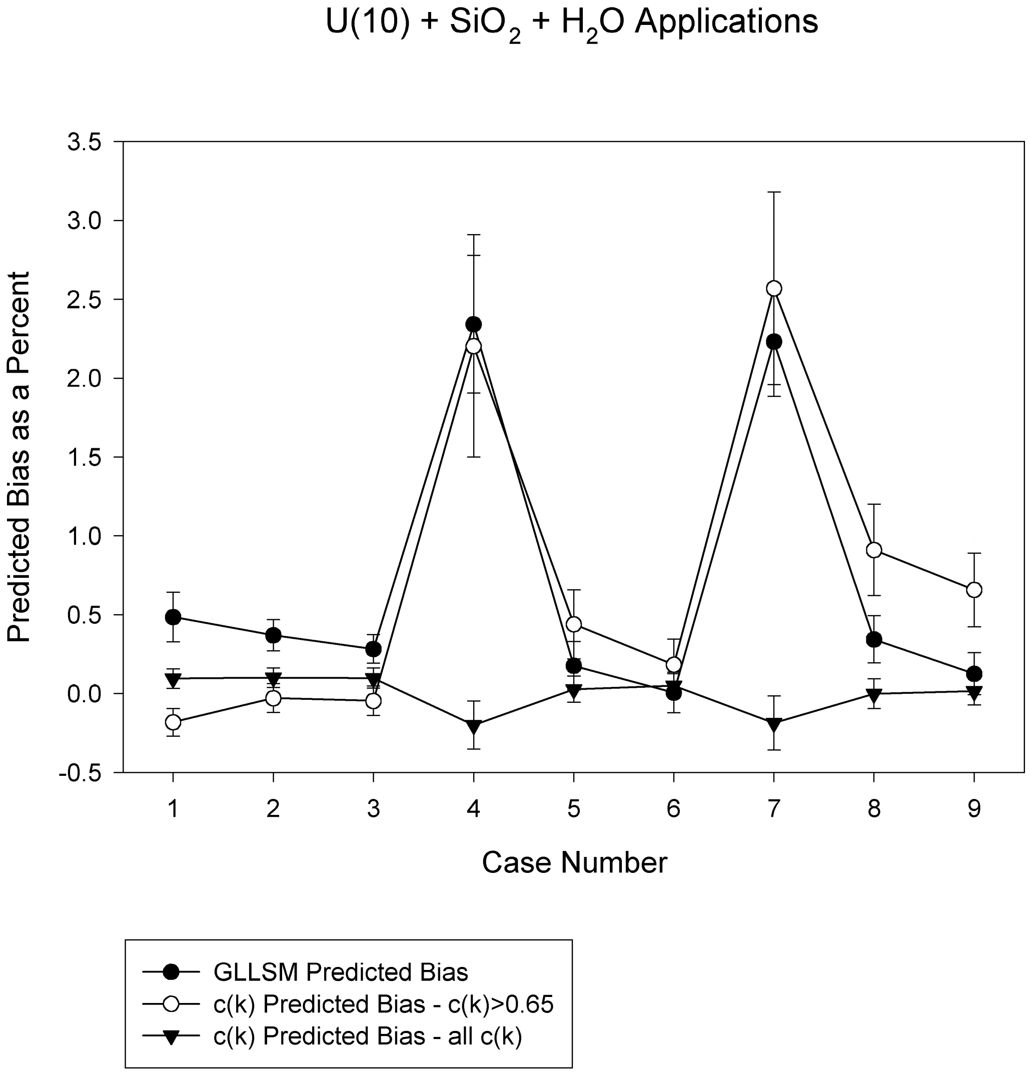

Figure 3. Bias predictions for nine U(10)SiO_2 applications.

The bias results for system SiO2-4 is also shown in Fig. 3. As noted in Table 1, this system has an H/X value of 0. The analysis for this system indicated that there are only two systems in the benchmark set with c_k values above 0.8 (only 8 systems with c_k above 0.65). Hence the trending with c_k for this case is not expected to be reliable without additional relevant experiments. This appears to be the case since the predictions for all c_k values and only those greater than 0.65 produce very different results. The GLLSM result appears to agree in this case with the trend with c_k values above 0.65, however, the R value for this application is only 0.33. Hence very little meaning can be attached to the GLLSM result as well. For cases SiO2-5 and SiO2-6, the addition of water quickly changes the characteristics of these systems, and the resulting trend plots. For case SiO2-5, the bias predictions should be reasonably reliable since; a large number of systems have c_k values above 0.8, the value of R is 0.72, and all three bias predictors agree quite well with each other. While not discussed, the conclusions regarding the SiO2-6 system are expected to be very similar to those of system SiO2-5, since they contain similar Si/X and H/X values.

Lastly, the SiO2-7, SiO2-8, and SiO2-9 systems are expected to very closely follow the SiO2-4, SiO2-5, and SiO2-6 systems discussed above. Each pair of systems, i.e., SiO2-4 vs SiO2-7, have very similar characteristics and are expected to behave in a similar manner with respect to the bias predictions. In summary, general agreement is seen for the various methods except the cases SiO2-4 and SiO2-7 where a sufficient number of experiments is not available based on the various reliability criteria. Also, caution is urged in the interpretation of the results from cases SiO2-1, SiO2-2 and SiO2-3.

Application to U(100)-SiO_2 Systems

This series of applications are designed to model a wide range of conditions that could exist in a low-level waste facility containing a maximum of 100 wt % U-bearing materials. The models for these applications were generated via a critical radius search for the conditions shown in Table 2.

Table 2. Specifications for U(100)-SiO_2 Systems[a]

	SiO2-1F	SiO2-2F	SiO2-3F
Sphere radius (cm)	1150.16	77.96	72.12
Si/X	3914	1165	783
H/X	0	977	1286
	SiO2-4F	SiO2-5F	SiO2-6F
Sphere radius (cm)	82.69	33.39	29.89
Si/X	543	407	329
H/X	0	342	540
	SiO2-7F	SiO2-8F	SiO2-9F
Sphere radius (cm)	49.45	21.79	17.92
Si/X	54	40	34
H/X	0	34	54

[a] Each system was surrounded by a 4-m-thick SiO_2 + variable H_2O reflector, where the H_2O concentration matches the core concentration

The results presented below were generated in the same manner as those of the Hanford tanks, and U(10)SiO_2 cases, previously. The general features of these 9 systems closely mimic the features of the 9 U(10)SiO_2 systems. The SiO2-1F, SiO2-4F, and SiO2-7F systems are completely dry with H/X values of 0. Each trio of systems, i.e., 1–3, 4–6, 7–9, exhibit increasing moderation along with decreasing SiO_2 concentrations.

As was noted in the U(10)SiO_2 systems, the deficiencies in the silicon covariance data will also have an impact on the results for the U(100)SiO_2 systems. The silicon capture sensitivities for cases 1F through 4F are generally larger than those for the corresponding 10 wt % systems, i.e., −0.504, −0.164, −0.105, −0.172, respectively. Thus for the same reasons stated earlier, the results for cases 1F through 4F should be used with caution.

Due to the likeness of the results for the U(100)SiO_2 systems to those of the U(10)SiO_2 systems, only the summary results for cases 1F through 9F are presented in Fig. 4. The general trends are the same as those shown in Fig. 3 in that the moderated systems have predicted biases near zero, while the dry systems have predicted biases of about 2%. Each of the moderated systems, have a large number of systems with c_k values greater than 0.8 and typically completeness parameter values greater than 0.8. Thus, the results for cases, 2F, 3F, 5F, 6F, 8F, and 9F appear to be quite reliable, except for the previously mentioned problems with 2F and 3F. This is borne out by the good agreement between the various bias prediction methods for these cases.

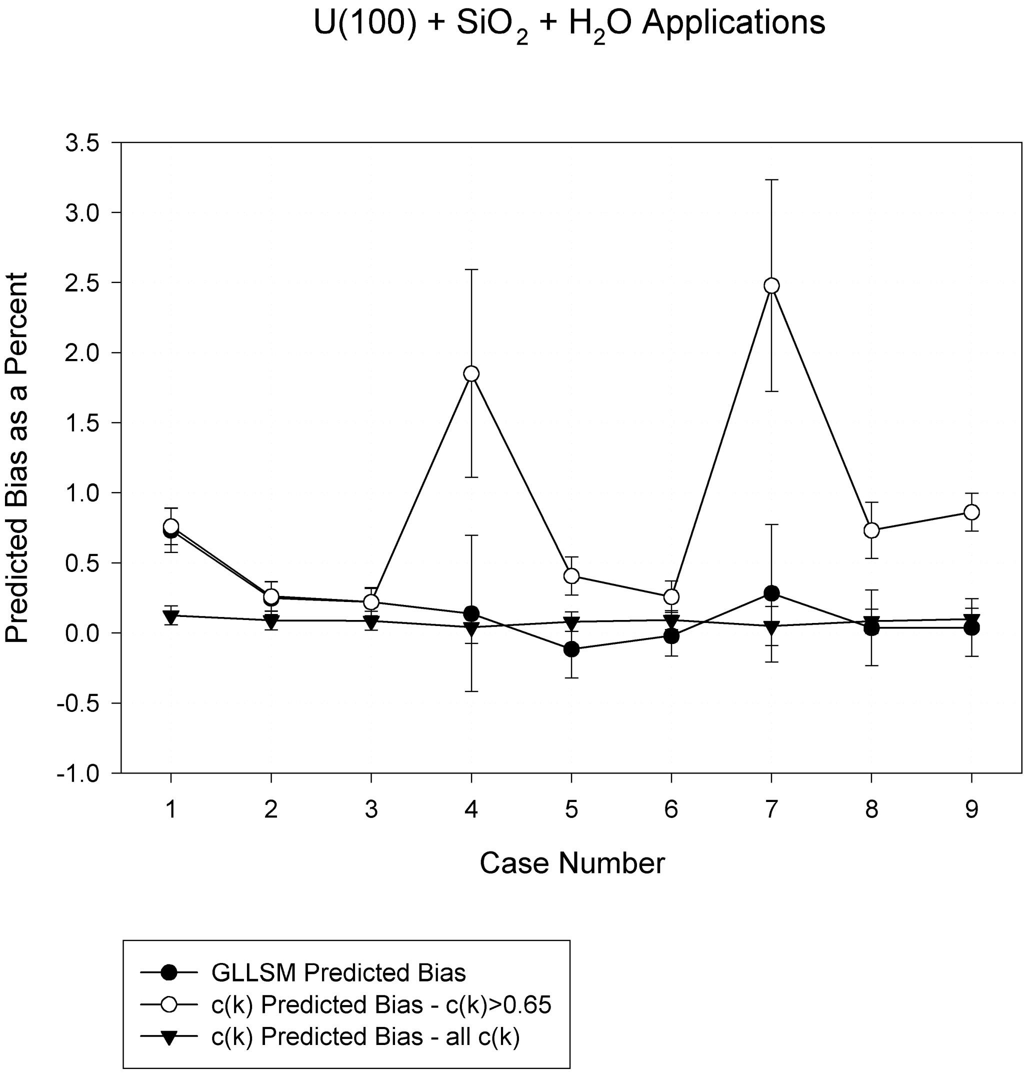

Figure 4. Bias predictions for nine U(100)SiO_2 applications.

However, for the U(100)-driven systems, the GLLSM versus trending approaches produce differing conclusions for cases 1F, 4F and 7F. Previously for the U(10)-based dry systems, the predicted biases using GLLSM versus trending with c_k for values greater than 0.65 agreed very well with each other. However, for this set of applications, the GLLSM results agree more closely with those predicted via trends versus all c_k values. This is not totally unexpected, since *none* of these cases meet the criterion for reliable c_k trending procedures (i.e., at least 15–20 systems with c_k values above 0.8). The value of the completeness c_k parameter, R, for cases 1F, 4F, and 7F are 0.47, 0.20, and 0.21, respectively. Thus, the GLLSM results are not likely to be reliable as well for these cases.

Application to Yucca Mountain PuO_2

The previous two criticality-safety application areas were intended to simulate a range of conditions that could occur following the release of uranium-bearing materials into the ground substructure. The intent of this application is similar, except the fissile material under study is plutonium. Two application problems were provided by the Yucca Mountain Project (YMP) support staff from a large number of configurations under study.[9] The two applications correspond to simulated PuO_2 residing in fracture areas of porous tuff and contain varying quantities of assumed water moderator.

The models used in this study were developed from three-dimensional models supplied by the YMP staff. The starting point for development of one-dimensional models used in this study was an array of cubes, each of which consisted of a 2.90 cm inner cube of tuff, inside by a 3 cm on a side outer cubic shell containing varying amounts of PuO_2. These small cubes were placed into a cubic array of dimension 1 m on each side, with an external one meter thick, cubic shell reflector of tuff. The two cases analyzed in this study contain 0 and 4 volume percents of interstitial water in the tuff regions. The fissile volume fractions for the two systems, designated e100p20 and e96p03, are 0.20 and 0.03, respectively. The 1-D models used in this study were developed by creating a cell-weighted material corresponding to the 3 cm cubes, then placing the cell-weighted mixture into spheres with volumes equivalent to the large 1 m and reflector cubes.

The 1-D models were used in the SEN1 module to generate sensitivity coefficients of k_{eff} to each of the cross sections of the constituent material nuclides. These sensitivity coefficients are then utilized in the CANDD code to generate values of c_k to each of 419 benchmark experiments. The results of these trending studies are shown in Fig. 5 corresponding to the e100p20 and e96p03 cases. In both cases the agreement between the two trending techniques and the GLLSM predictions is very good. The c_k criterion appears to also be met for both cases, with approximately 15 and 50 cases, respectively, with c_k values greater than 0.8. Thus, the bias estimates should be well predicted by the various methods, based on the criterion established thus far. However, the values of R for these two cases are 0.17 and 0.53, indicating that the benchmark database is not complete with respect to *either* of the two applications areas. Further investigation was necessary to understand this apparent discrepancy.

The very low value of R for the e100p20 case is caused by a combination of effects. The first effect is the high sensitivity of this system to the plutonium fission and capture cross sections in the intermediate-energy range (1 to 1000 eV). The value of R is calculated based on the sum of the absolute values of the fission and capture sensitivities, while c_k is based on the propagated cross section uncertainties to the value of k_{eff}. Secondly, in the intermediate-energy range, the uncertainties in the fission and capture cross sections are highly anti-correlated due to the cross section evaluations being based on measurements of alpha (capture-to-fission ratio). The result of this correlation is to lessen the impact of individual uncertainties in the fission and capture components of the cross section in this range. However, the summation of the absolute value of the sensitivities in this range, as in the R calculation, tends to increase the impact of this region, since cancellation of positive and negative sensitivity components does not occur. This application also suffers from a lack of a sufficient number of similar systems, which also contributes to the very low value of R. Thus, the predicted bias for this system should not be considered reliable, even though the various methods tend to agree very well with each other.

The second system, e96p03, has primarily the same type of effects as the e100p20 case, however, to a lesser extent. This system is primarily sensitive to the thermal cross sections for fission and capture of plutonium. The correlation of the capture and fission uncertainties is much smaller in the thermal range for plutonium than in the intermediate range. The primary cause of the relatively low value of R for this case is the use of pure ^{239}Pu as the fissile nuclide in this case. The use of pure material causes the ^{239}Pu sensitivities to be enhanced and thus the comparison of sensitivities shown in definition of the R parameter fails for systems that are indeed similar to this system. This effect was also a contributor to the effects seen for the e100p20 system. Due to the somewhat superficial under prediction of the value of R for this case, the bias prediction for this system should be reliable for validation efforts. The predicted biases and their uncertainties are shown in summary form in Fig. 5 for both of these YMP systems.

4. Summary

This paper has presented details of the current state of the validation techniques that apply sensitivity and uncertainty methods to criticality safety studies. Validation parameters D, E and c_k have been developed to be utilized in these new methods. This work has only dealt with the c_k parameters and the GLLSM method.

Additionally, a new parameter, R, has been defined that measures the “completeness” of the benchmark dataset for a given application. The value of this new parameter is that it gives a measure of the applicability of the benchmark set as a whole, even if the individual systems do not under standard procedures meet the conditions of applicability. A value of R of about 0.8 seems to indicate that dataset is capable of being used as a validation of the given application. Work is currently underway to further refine the meaning of the magnitude of the R parameter.

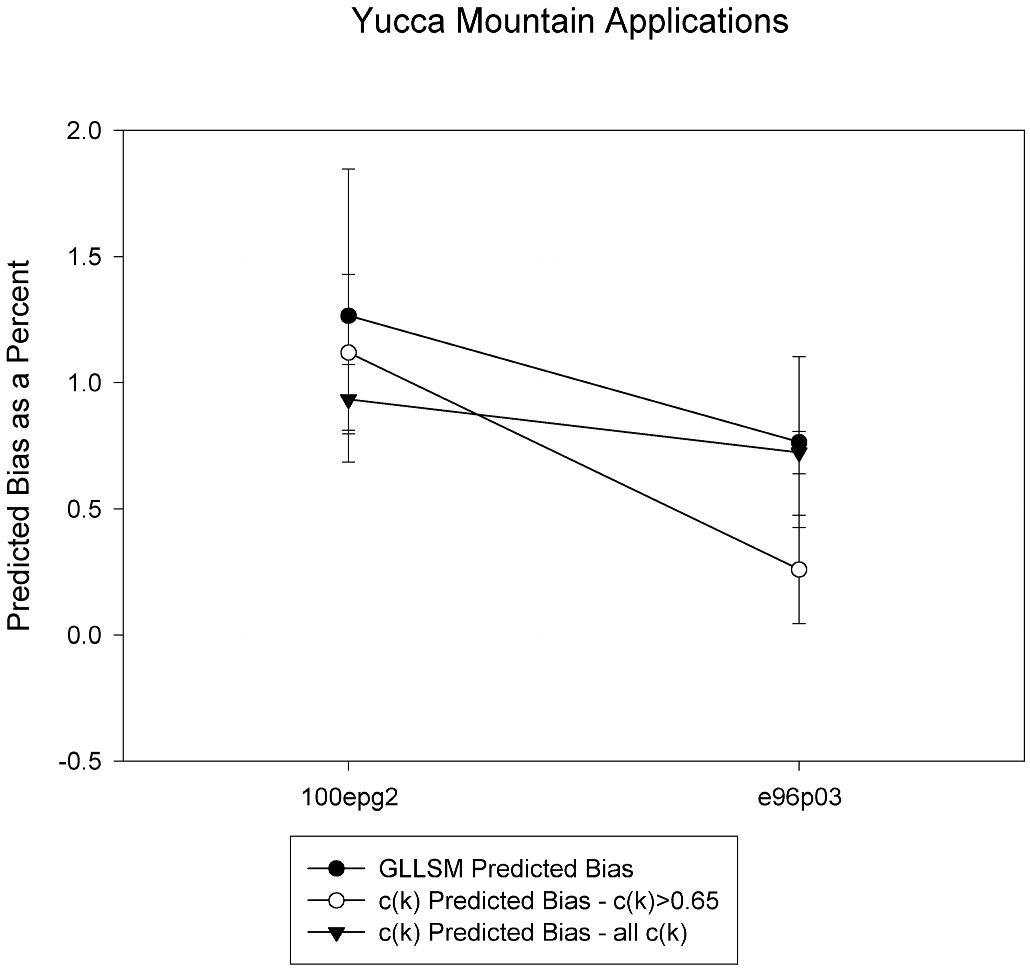

Figure 5. Bias predictions for two Yucca Mountain $PuSiO_2$ applications.

Current versions of the S/U techniques have been applied to four different application areas; Hanford tanks, U(10)-SiO_2 systems, U(100)-SiO_2 systems, and YMP Pu-SiO_2 systems. These studies are not complete, but it is apparent from the results presented thus far that the techniques are quite beneficial in their prediction of biases for a number of unusual systems. The techniques are also valuable in determining whether a set of benchmark experiments is adequate to validate a given set of applications. In the case of the Hanford tanks, validation was illustrated using these techniques where validation was difficult without these techniques. The SiO_2 application areas exposed that well-moderated systems can typically be validated quite readily, however, dry systems need additional experiments including SiO_2 in order for validation to be possible. The applicability of the new Russian and LANL experiments with SiO_2 to these applications are currently under evaluation using these techniques. These are valuable tools to have available for general use. This goal has not been realized as of yet, but is the aim of current and future studies.

This study has shown that the GLLSM and trending with c_k for all 419 benchmark systems produced generally consistent results. The exceptions to this are also when the GLLSM was deemed to be unreliable due to non-completeness. Further work will be performed to assess these observations, and to determine perhaps a more appropriate method for estimating biases for general systems.

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