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**VALIDATION OF A METHOD FOR PREDICTION OF ISOTOPIC
CONCENTRATIONS IN BURNUP CREDIT APPLICATIONS**

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VALIDATION OF A METHOD FOR PREDICTION OF ISOTOPIC CONCENTRATIONS IN BURNUP CREDIT APPLICATIONS

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SUMMARY

Unlike fresh fuel assumptions typically employed in the criticality safety analysis of spent fuel configurations, burnup credit applications rely on depletion and decay calculations to predict the isotopic composition of spent fuel. These isotopics are used in subsequent criticality calculations to assess the reduced worth of the spent fuel. To validate the codes and data used in depletion approaches, experimental measurements are compared with numerical predictions for relevant spent fuel samples. This paper describes a set of experimentally characterized pressurized-water-reactor (PWR) fuel samples and provides a comparison to results of SCALE-4 [1] depletion calculations. An approach to determine biases and uncertainties between calculated and measured isotopic concentrations is discussed, together with a method to statistically combine these terms to obtain a conservative estimate of spent fuel isotopic concentrations.

INTRODUCTION

The radionuclide characteristics of light-water-reactor (LWR) spent fuel play key roles in the design and licensing activities for radioactive waste systems that are intended to take credit for fuel burnup. Burnup credit applications rely on an accurate prediction of spent fuel isotopic concentrations to correctly estimate the reduced reactivity of spent fuel. At the Oak Ridge National Laboratory (ORNL), burnup credit studies for application with PWR spent fuel have generally used the SCALE-4 code system to predict the radionuclide composition of depleted fuel. However, analytical methods (including those for isotopic prediction) used in spent fuel criticality analyses should be validated as outlined in the ANSI/ANS 8.1

criticality safety standard [2]. Under the guidance of the standard, comparison of calculational results with experimental measurements is used in the validation of SCALE-4 for calculation of PWR spent fuel isotopic compositions. This paper presents and discusses a set of PWR spent fuel samples for which numerical depletion models have been developed. Spent fuel isotopic concentrations computed using these models are compared with measured nuclide densities to determine the adequacy of the numerical estimates. Additionally, a statistical method is described in which biases and uncertainties are determined based on these comparisons, and are conservatively combined to provide a bounding estimate of isotopic concentration for use in subsequent criticality safety analyses.

OVERVIEW OF THE ANALYSIS METHOD

Calculations performed in this work used various computational modules in version 4.2 of the SCALE-4 code system. Spent fuel characterization is performed by the SAS2H analysis sequence of SCALE-4 [3]. SAS2H is a multicode sequence that determines the isotopic composition of spent fuel using the ORIGEN-S code for depletion and decay calculations and the XSDRNPM one-dimensional (1-D) deterministic transport code to prepare burnup-dependent cross sections for ORIGEN-S. Calculations were performed using two different cross-section libraries: (1) the SCALE-4 27-group burnup library, a hybrid library that contains ENDF/B-IV (actinide) and ENDF/B-V (fission product) data for isotopes important in spent fuel from commercial reactor fuel designs; and (2) the SCALE 44-group ENDF/B-V library, which contains the 300+ nuclides available in the ENDF/B-V data files, and was specifically tailored for the

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needs of LWR spent fuel analysis. The 44-group library also contains ENDF/B-VI evaluations for ^{16}O , ^{154}Eu , and ^{155}Eu ; evaluated data for these isotopes has been significantly improved in ENDF/B-VI.

SAS2H input provides data describing a fuel assembly as it is initially loaded into a particular reactor. The geometry, initial material composition, average zone temperatures, and time-dependent specific power of the fuel assembly are required input. Although ORIGEN-S is a point-depletion code and contains no spatial dependence, spatial effects can be treated by using XSDRNPM to generate cross sections that are weighted by assembly design characteristics to capture the effects of assembly nonuniformities (e.g., water holes). Such weighting is generally performed based on an assembly averaged basis to provide assembly averaged results. However, it is possible to refine the weighting scheme to estimate the effects of a particular location within an assembly by specifying local temperatures, material densities and specific powers.

SAS2H/ORIGEN calculations are performed in burnup stages based on the operational history of the assembly being modeled, and provide isotopic concentrations for a specified cooling time after the end of the final depletion cycle. In burnup credit methods, the concentrations of selected nuclides from such calculations are used in criticality models of spent fuel configurations; however, in this work the computed concentrations are compared with the experimentally measured concentrations of available nuclides. Sources of experimental measurements are described in the following section.

CHEMICAL ASSAY MEASUREMENTS

The majority of the radiochemical assays of PWR spent fuel included in this study [4-7] were conducted by the Material Characteristics Center (MCC) at Pacific Northwest Laboratory (PNL) using discharged PWR fuel from Calvert Cliffs Unit 1 and H. B. Robinson Unit 2. Additional spent fuel characterizations [8,9] presented herein were conducted by four research laboratories in Europe using fuel elements

from the Obrigheim (KWO) PWR.

A wide range of parameters was included in the problems analyzed for this study. The fuel exposures, or burnups, were in the range 16.02 to 46.46 GWd/MTU. The PWR designs were substantially different: Calvert Cliffs assemblies were of a Combustion Engineering 14×14 design; the H. B. Robinson assembly was a Westinghouse 15×15 design; and Obrigheim used a Siemens-designed 14×14 assembly. In addition, there were significant variations in fuel ^{235}U enrichments (2.45 to 3.04 wt %), assembly operational histories, average fuel temperatures (743 to 923 K), specific powers (13.1 to 44.7 MW/MTU), and other pertinent operational conditions among the cases considered.

Fuel assemblies taken from the three reactors were used to obtain 19 different fuel samples. Three fuel pins from each of three different assemblies were acquired from the Calvert Cliffs PWR. From each pin, fuel pellets were removed from three different axial locations to provide a total of nine Calvert Cliffs samples with a wide range of burnups. Similarly, a single fuel pin removed from an H. B. Robinson assembly was disassembled, and fuel pellets were selected from four axial locations distributed along the length of the pin. Obrigheim assemblies were examined by a significantly different approach: each of six assemblies studied were cut into full-length, half-width units. One half-width unit was then dissolved. Thus unlike the spent fuel characterizations performed at MCC in which fuel represented a specific position in the assembly, Obrigheim spent fuel characterization represented the average contents over the full length of the assembly.

Table I describes some of the significant aspects of each fuel sample. The last column of the table lists the isotopes measured from each sample. For the most part, fission-product measurements were performed only for the three samples obtained from the Calvert Cliffs D047 fuel pin; thus fission-product measurements are somewhat limited. In some cases, specific isotopes within a given mass number could not be chemically separated (i.e., mass numbers 147, 151, and 155). In such cases, isotopic comparisons are made

Table I. Summary of 19 Spent Fuel Samples Analysed.

Reactor	Sample ID (sample location ^a)	Burnup (GWd/MTU)	Initial enrichment (wt %)	Average effective fuel temperature (K)	Isotopes measured
Calvert Cliffs	D047 (13.20 cm)	27.35	3.038	790	⁹⁹ Tc, ¹³³ Cs, ¹³⁵ Cs, ¹⁴³ Nd, ¹⁴⁵ Nd, ¹⁴⁷ Pm+ ¹⁴⁷ Sm, ¹⁴⁹ Sm, ¹⁵⁰ Sm, ¹⁵¹ Sm+ ¹⁵¹ Eu, ¹⁵² Sm, ¹⁵³ Eu, ¹⁵⁵ Eu+ ¹⁵⁵ Gd, ²³⁴ U, ²³⁵ U, ²³⁶ U, ²³⁸ U, ²³⁸ Pu, ²³⁹ Pu, ²⁴⁰ Pu, ²⁴¹ Pu, ²⁴² Pu, ²⁴¹ Am
Calvert Cliffs	D047 (27.70 cm)	37.12	3.038	841	⁹⁹ Tc, ¹³³ Cs, ¹³⁵ Cs, ¹⁴³ Nd, ¹⁴⁵ Nd, ¹⁴⁷ Pm+ ¹⁴⁷ Sm, ¹⁴⁹ Sm, ¹⁵⁰ Sm, ¹⁵¹ Sm+ ¹⁵¹ Eu, ¹⁵² Sm, ¹⁵³ Eu, ¹⁵⁵ Eu+ ¹⁵⁵ Gd, ²³⁴ U, ²³⁵ U, ²³⁶ U, ²³⁸ U, ²³⁸ Pu, ²³⁹ Pu, ²⁴⁰ Pu, ²⁴¹ Pu, ²⁴² Pu, ²⁴¹ Am
Calvert Cliffs	D047 (165.22 cm)	44.34	3.038	873	⁹⁹ Tc, ¹³³ Cs, ¹³⁵ Cs, ¹⁴³ Nd, ¹⁴⁵ Nd, ¹⁴⁷ Pm+ ¹⁴⁷ Sm, ¹⁴⁹ Sm, ¹⁵⁰ Sm, ¹⁵¹ Sm+ ¹⁵¹ Eu, ¹⁵² Sm, ¹⁵³ Eu, ¹⁵⁵ Eu+ ¹⁵⁵ Gd, ²³⁴ U, ²³⁵ U, ²³⁶ U, ²³⁸ U, ²³⁸ Pu, ²³⁹ Pu, ²⁴⁰ Pu, ²⁴¹ Pu, ²⁴² Pu, ²⁴¹ Am
Calvert Cliffs	D101 (9.10 cm)	18.68	2.72	816	⁹⁹ Tc, ¹³⁵ Cs, ²³⁴ U, ²³⁵ U, ²³⁶ U, ²³⁸ U, ²³⁸ Pu, ²³⁹ Pu, ²⁴⁰ Pu, ²⁴¹ Pu, ²⁴² Pu, ²⁴¹ Am
Calvert Cliffs	D101 (24.50 cm)	26.62	2.72	880	⁹⁹ Tc, ¹³⁵ Cs, ²³⁴ U, ²³⁵ U, ²³⁶ U, ²³⁸ U, ²³⁸ Pu, ²³⁹ Pu, ²⁴⁰ Pu, ²⁴¹ Pu, ²⁴² Pu, ²⁴¹ Am
Calvert Cliffs	D101 (161.90 cm)	33.17	2.72	910	⁹⁹ Tc, ¹³⁵ Cs, ²³⁴ U, ²³⁵ U, ²³⁶ U, ²³⁸ U, ²³⁸ Pu, ²³⁹ Pu, ²⁴⁰ Pu, ²⁴¹ Pu, ²⁴² Pu, ²⁴¹ Am
Calvert Cliffs	BT03 (11.28 cm)	31.40	2.453	790	⁹⁹ Tc, ¹³⁵ Cs, ²³⁴ U, ²³⁵ U, ²³⁶ U, ²³⁸ U, ²³⁸ Pu, ²³⁹ Pu, ²⁴⁰ Pu, ²⁴¹ Pu, ²⁴² Pu, ²⁴¹ Am
Calvert Cliffs	BT03 (19.92 cm)	37.27	2.453	841	⁹⁹ Tc, ¹³⁵ Cs, ²³⁴ U, ²³⁵ U, ²³⁶ U, ²³⁸ U, ²³⁸ Pu, ²³⁹ Pu, ²⁴⁰ Pu, ²⁴¹ Pu, ²⁴² Pu, ²⁴¹ Am
Calvert Cliffs	BT03 (161.21 cm)	46.46	2.453	873	⁹⁹ Tc, ¹³⁵ Cs, ²³⁴ U, ²³⁵ U, ²³⁶ U, ²³⁸ U, ²³⁸ Pu, ²³⁹ Pu, ²⁴⁰ Pu, ²⁴¹ Pu, ²⁴² Pu, ²⁴¹ Am
H. B. Robinson	N-9B-S (11 cm)	16.02	2.561	743	⁹⁹ Tc, ²³⁵ U, ²³⁶ U, ²³⁸ U, ²³⁸ Pu, ²³⁹ Pu, ²⁴⁰ Pu, ²⁴¹ Pu
H. B. Robinson	N-9B-N (26 cm)	23.81	2.561	830	⁹⁹ Tc, ²³⁵ U, ²³⁶ U, ²³⁸ U, ²³⁸ Pu, ²³⁹ Pu, ²⁴⁰ Pu, ²⁴¹ Pu
H. B. Robinson	N-9C-J (199 cm)	28.47	2.561	883	⁹⁹ Tc, ²³⁵ U, ²³⁶ U, ²³⁸ U, ²³⁸ Pu, ²³⁹ Pu, ²⁴⁰ Pu, ²⁴¹ Pu
H. B. Robinson	N-9C-D (226 cm)	31.66	2.561	923	⁹⁹ Tc, ²³⁵ U, ²³⁶ U, ²³⁸ U, ²³⁸ Pu, ²³⁹ Pu, ²⁴⁰ Pu, ²⁴¹ Pu
Obrigheim	94-170	25.93	3.13	846	²³⁵ U, ²³⁶ U, ²³⁸ Pu, ²³⁹ Pu, ²⁴⁰ Pu, ²⁴¹ Pu, ²⁴² Pu
Obrigheim	92-172	26.54	3.13	841	²³⁵ U, ²³⁶ U, ²³⁸ Pu, ²³⁹ Pu, ²⁴⁰ Pu, ²⁴¹ Pu, ²⁴² Pu
Obrigheim	91-176	27.99	3.13	849	²³⁵ U, ²³⁶ U, ²³⁸ Pu, ²³⁹ Pu, ²⁴⁰ Pu, ²⁴¹ Pu, ²⁴² Pu
Obrigheim	86-168	28.40	3.13	859	²³⁵ U, ²³⁶ U, ²³⁸ Pu, ²³⁹ Pu, ²⁴⁰ Pu, ²⁴¹ Pu, ²⁴² Pu
Obrigheim	89-171	29.04	3.13	867	²³⁵ U, ²³⁶ U, ²³⁸ Pu, ²³⁹ Pu, ²⁴⁰ Pu, ²⁴¹ Pu, ²⁴² Pu
Obrigheim	90-176	29.52	3.13	867	²³⁵ U, ²³⁶ U, ²³⁸ Pu, ²³⁹ Pu, ²⁴⁰ Pu, ²⁴¹ Pu, ²⁴² Pu

^a Position of sample pellet measured from the bottom of the fuel. Does not apply to Obrigheim data, which is assembly averaged.

relative to the combined concentrations of the two isotopes. Table I shows the combined isotopes. Note that this table lists only those isotopes determined to be important in burnup credit criticality analyses [10]. Additional isotopic measurements were performed and are available in Refs. 4-9.

COMPARISON TO CALCULATED ISOTOPIC CONCENTRATIONS

Using the SCALE SAS2H analytical sequence described earlier, isotopic concen-

trations were computed for each of the nuclides for which isotopic measurements were available, as listed in Table I. Results are illustrated in Figs. 1 and 2 in terms of the relative difference between calculated and measured concentrations. These results demonstrate that using either cross-section library, SAS2H calculations are able to predict actinide concentrations to within approximately $\pm 10\%$ relative to experimental measurements, although there is significant spread in ^{234}U data. Many fission-product concentrations are calculated to within the

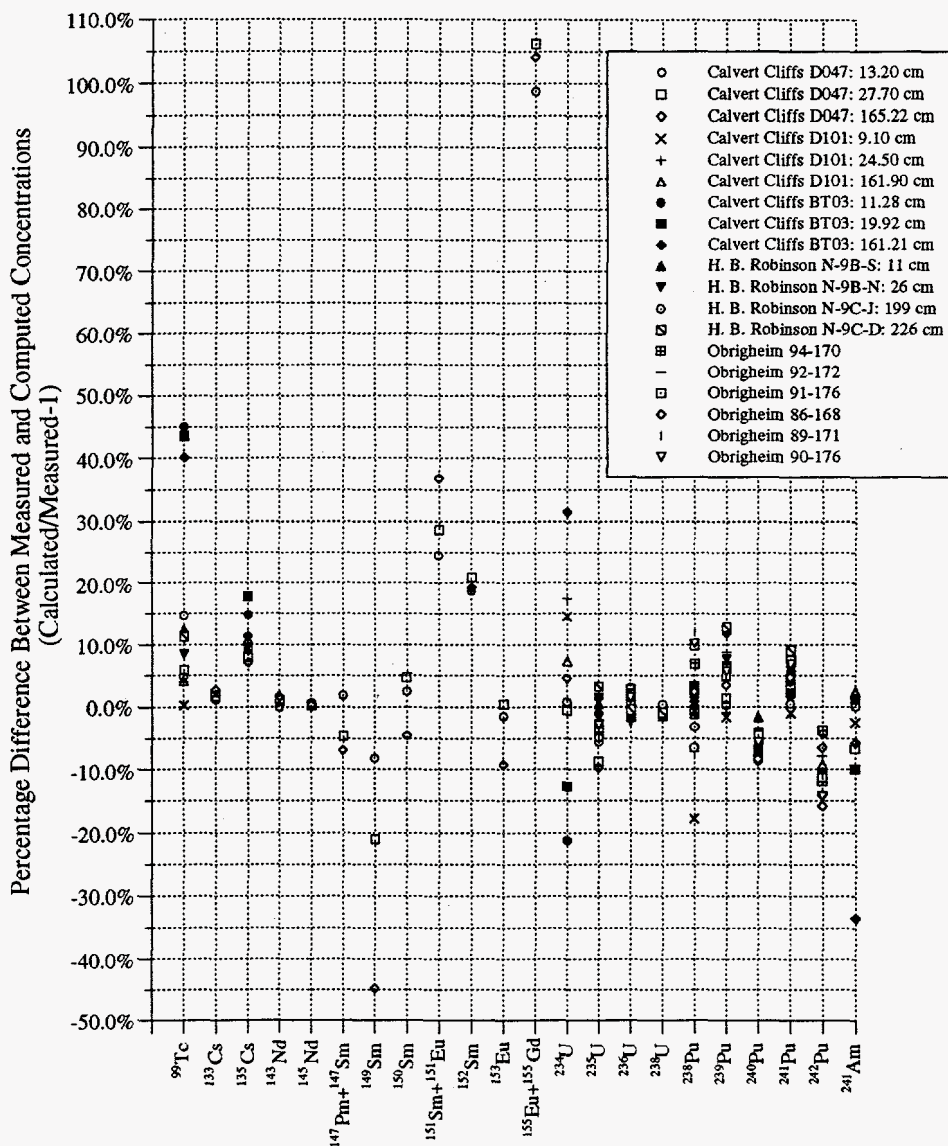


Figure 1. Comparison of Measured to Computed Isotopic Concentrations (27-Group Library).

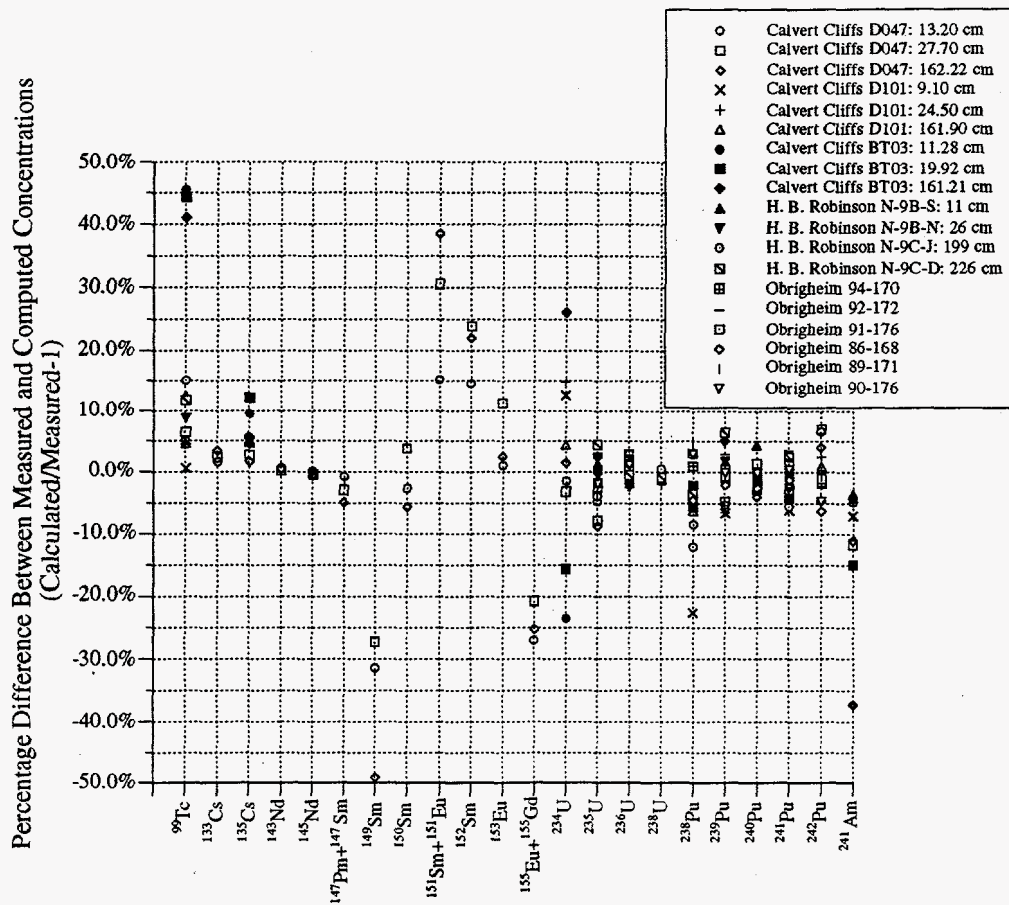


Figure 2. Comparison of Measured to Computed Isotopic Concentrations (44-Group Library).

same accuracy, and almost all are calculated to within $\pm 15\%$. Results obtained using the 44-group library are generally in better agreement with measurements than 27-group-library results, although there are minor exceptions. For both libraries, the principal outliers are ^{99}Tc , ^{149}Sm , mass chain 151 ($^{151}\text{Sm} + ^{151}\text{Eu}$), ^{152}Sm , and mass chain 155 ($^{155}\text{Eu} + ^{155}\text{Gd}$). The error in several of the fission-product isotopes (i.e., ^{99}Tc , ^{149}Sm , and mass chain 151) may be due to the relatively small isotopic abundance of these nuclides in the spent fuel samples, which would make experimental measurements more difficult and therefore would contain more measurement error. In addition, it is observed that the outliers for several of the isotopes (i.e., ^{99}Tc , ^{135}Cs , ^{234}U , and ^{241}Am) are primarily from the Calvert Cliffs BT03 sample. This fact suggests that some aspect of the burnup of this particular sample may have been inade-

quately characterized in the depletion models. Finally, it can be seen that the results for several of the samples are relatively closely clustered, indicating a constant bias in the prediction and/or measurement of the concentrations for these isotopes. For example, all three measurements made for mass chain 155 show a consistent overprediction of the concentrations of these isotopes (by roughly 100%) relative to measured concentrations. This particular behavior has been traced to a cross-section-induced bias due to a lack of resonance cross-section data in the ^{155}Eu evaluation in ENDF/B-V. Since ^{155}Eu decays to ^{155}Gd , an overprediction of ^{155}Eu concentration will result in a corresponding overprediction of ^{155}Gd . Use of an improved ENDF/B-VI evaluation for ^{155}Eu results in a much-improved estimate of mass chain 155 concentrations, as shown in Fig. 2.

CONSERVATIVE LIMITS FOR ISOTOPIC CONCENTRATIONS

Differences between calculated and measured isotopic concentrations for any given fuel sample result from biases and uncertainties in both the calculated and measured results. Isotopic uncertainty is a consequence of the variation of physical parameters in a random fashion relative to modeled parameters, combined with the random error associated with experimental measurements. Isotopic bias, on the other hand, is the offset between the measured nuclide concentration for a given nuclide and the corresponding calculational prediction of the concentration of that nuclide, after accounting for uncertainties. Isotopic bias can result from both the method used in experimental measurements and from the assumptions, data, and method used in computational prediction of nuclide densities. Isotopic bias therefore represents the difference between the computed concentration for a given isotope and the actual isotopic content in a spent fuel unit. Isotopic uncertainty represents the confidence with which the corresponding isotopic bias is known.

In the approach described here, an isotopic correction factor is derived based on the statistical variations in the measured isotopic concentration relative to calculated concentrations for the same isotope over the range of all experimental measurements for the selected isotope. For any given isotope, a ratio between experimentally measured and the corresponding calculated isotopic concentration may be defined as

$$x_i = M_i/C_i, \quad (1)$$

where C_i is the calculated concentration for a given isotope in sample i , and M_i is the measured concentration for the same isotope in sample i . (If more than one measurement is performed on sample i , M_i is determined as the average of the multiple measurements.) Given a population of ratios for all fuel samples for which spent fuel analysis is considered, the population mean and variance are defined as μ and σ^2 . For a given level of confidence, any member of the population will be found within the range $\mu \pm N_\alpha\sigma$, where N_α is determined based on an α confidence level (generally α is selected as

95% or 99%). However, μ and σ^2 cannot be directly ascertained without the destructive assay of every available fuel sample. Thus it is necessary to estimate the mean and variance in terms of the mean and variance of a subset of samples selected to represent the entire population. The mean of the sample set (and the estimate for μ) is given as \bar{x} , where for a sample set comprised of n samples,

$$\bar{x} = \frac{1}{n} \sum_{i=1}^n x_i \approx \mu, \quad (2)$$

Assuming the set of x_i values for a given isotope have a normal distribution about the mean of the sample set, the sample variance is given as

$$s^2 = \frac{1}{(n-1)} \sum_{i=1}^n (x_i - \bar{x})^2 \approx \sigma^2. \quad (3)$$

Given a set of measured-to-calculated ratios for a given isotope, one can determine \bar{x} and s^2 as estimates of the population's true mean and variance. If \bar{x} and s^2 are good estimates of μ and σ^2 , then bounding limits of \bar{x} are given by $\bar{x} \pm N_\alpha s$. Again, the value of N depends on the statistical confidence desired for a bounding limit. Clearly, as n (the number of samples in the population subset used to establish the estimated parameters) becomes large, one has a reasonable confidence that the sample parameters are reasonable estimates of the population parameters. However, when the sample population is small, there is limited confidence that the sample parameters represent the population parameters. Thus a second statistical parameter is required to relate a second level of confidence, ρ , to the estimate of the population mean and variance. For a set of normally distributed data points, one may use a tolerance interval approach [11] to assign a "tolerance factor" $T_{\alpha/\rho}$ value for N . Under this approach, for a given α and ρ , there is a probability ρ that a future value of x_i will lie within the range $\bar{x} \pm T_{\alpha/\rho} s$, with an α confidence. Reference 12 provides tabulated values of $T_{\alpha/\rho}$ for common values of α and ρ and for a range of values of n . In such tables, it is observed

that $T_{\alpha/\rho}$ is large relative to N_{α} for small values of n , but approaches N_{α} as n becomes larger.

Recall that x represents the ratio of measured-to-calculated isotopic concentrations for a given isotope. Thus the limits of $\bar{x} \pm T_{\alpha/\rho} \cdot s$ represent the expected upper and lower bounds for measured-to-calculated ratios with a confidence of α with a probability ρ . In a spent fuel criticality calculation, a conservative approach in the prediction of a neutron multiplication factor, k , is to assume the maximum concentration of fissile isotopes combined with the minimum concentration of nonfissile isotopes, to set an uppermost expected limit on k . Thus for a set of isotopes present in a criticality calculation, one must determine the limiting isotopic concentration for each isotope by using a correction factor appropriate for the isotope. These correction factors may be written for each isotope I as

$$f_{I,\text{fissile}} = \bar{x}_I + T_{I,\alpha/\rho} \cdot s_I \quad (4)$$

and

$$f_{I,\text{nonfissile}} = \bar{x}_I - T_{I,\alpha/\rho} \cdot s_I \quad (5)$$

The estimated bounding concentration of isotope I , ϵ_I , is then simply given as

$$\epsilon_I = f_I \cdot C_I \quad (6)$$

Clearly, to be included in a criticality calculation, isotopic measurements must be available for each isotope I such that f_I can be computed for the isotope. As the number of experimental measurements is increased, the term s_I will generally decrease or remain constant, and $T_{I,\alpha/\rho}$ will decrease; hence the margin between the average ratio and the bounding limit will decrease with an increased number of experimental measurements. Isotopic correction factors based on a limited number of experimental measurements are therefore generally excessively conservative, although the conservatism is statistically justified.

Based on the calculated and measured concentrations illustrated in Figs. 1 and 2, isotopic correction factors have been calculated for each of the available nuclides. These results are provided in Table II for each of the isotopes. The correction factors

computed based on available isotopic data demonstrate the earlier observation that actinide predictions are in general very good; correction factors are fairly close to 1.0. However, for fission products, correction factors are considerably less than 1.0. This result is partially due to the greater spread in the relative differences between computed and measured isotopics for the samples analyzed; however, the fission-product correction factors are most strongly influenced by the large value of $T_{95/95}$ resulting from the limited number of measurements available for fission-product isotopes. The majority of the correction factors are improved (i.e., closer to 1.0) for results based on the 44-group library; this is especially true for mass chain 155 and for ^{147}Sm . However, for several isotopes (most notably ^{151}Sm and ^{152}Sm), the older 27-group library results in better correction factors, indicating better correlation between measured and calculated results.

Isotopic correction factors can be applied to SAS2H-computed isotopic concentrations to obtain a conservative estimate of these concentrations for the purposes of criticality safety calculations. Use of these values, determined at 95%/95% confidence level for **each isotope**, in a criticality calculation for a given spent fuel configuration should result in a conservative upper estimate of k for the configuration with a very high degree of confidence.

CONCLUSIONS

The use of the SCALE code system in spent fuel analyses related to criticality safety under burnup credit assumptions requires the validation of both the depletion and criticality capabilities of the code relative to spent fuel considerations. This paper has addressed a portion of the ongoing efforts at ORNL to validate the capability of the SCALE SAS2H analytical sequence in the prediction of spent fuel isotopic compositions. These efforts have included the introduction of a statistical approach for conservatively estimating isotopic concentrations based on estimated biases and uncertainties in the SAS2H approach.

Results indicate that SAS2H can generally predict the concentrations of most isotopes important in burnup credit spent fuel

Table II. Correction Factors for Conservative Prediction of Spent Fuel Composition.

Isotope	n	T _{95/95,n} ^a	27-Group Library			44-Group Library		
			\bar{x}	s	f	\bar{x}	s	f
⁹⁹ Tc	13	2.671	0.8767	0.106	0.594	0.8732	0.106	0.589
¹³³ Cs	3	7.656	0.9814	0.007	0.928	0.9761	0.009	0.910
¹³⁵ Cs	9	3.031	0.9022	0.027	0.820	0.9471	0.029	0.860
¹⁴³ Nd	3	7.656	0.9909	0.008	0.930	0.9962	0.003	0.976
¹⁴⁵ Nd	3	7.656	0.9961	0.002	0.981	1.0031	0.004	0.975
¹⁴⁷ Sm	3	7.656	1.0349	0.047	0.675	1.0296	0.022	0.864
¹⁴⁹ Sm	3	7.656	1.3972	0.368	0.000 ^c	1.5998	0.320	0.000 ^c
¹⁵⁰ Sm	3	7.656	0.9924	0.048	0.625	1.0165	0.048	0.646
¹⁵¹ Sm	3	7.656	0.7707	0.037	0.487	0.7855	0.075	0.213
¹⁵² Sm	3	7.656	0.8362	0.008	0.775	0.8333	0.035	0.564
¹⁵³ Eu	3	7.656	1.0370	0.057	0.601	0.9542	0.049	0.577
¹⁵⁵ Gd	3	7.656	0.5361	0.085	0.000 ^c	1.3235	0.056	0.893
²³⁴ U	9	3.031	0.9757	0.154	0.509	1.0037	0.160	0.519
²³⁵ U ^b	19	2.423	1.0305	0.035	1.115	1.0208	0.034	1.104
²³⁶ U	19	2.423	0.9936	0.016	0.955	0.9944	0.016	0.956
²³⁸ U	13	2.671	1.0064	0.006	0.990	1.0063	0.006	0.991
²³⁸ Pu	19	2.423	0.9967	0.069	0.830	1.0534	0.075	0.871
²³⁹ Pu ^b	19	2.423	0.9508	0.034	1.033	1.0068	0.035	1.092
²⁴⁰ Pu	19	2.423	1.0615	0.020	1.013	1.0038	0.019	0.957
²⁴¹ Pu ^b	19	2.423	0.9531	0.026	1.016	1.0124	0.028	1.080
²⁴² Pu	15	2.566	1.1160	0.043	1.006	1.0033	0.038	0.911
²⁴¹ Am	9	3.031	1.0800	0.164	0.583	1.1420	0.176	0.609

^a From Ref. 11.

^b Fissile isotope.

^c Factors less than zero are set to zero, since negative concentrations are meaningless.

applications to within $\pm 15\%$. However, improvement is needed, especially in terms of fission-product predictions. Some weaknesses are known to result from limitations in the codes, models, and data used in these SAS2H calculations. Use of the best available cross sections and perhaps the application of methods with better spatial resolution of assembly characteristics should improve the ability of SAS2H to predict the isotopic composition of spent fuel. However, the largest impact on the ability to conservatively estimate spent fuel compositions is the consequence of an inadequate database of spent fuel chemical assay measurements. This is especially true for fission-product nuclides, many of which have only three measurements available from which to estimate bias and uncertainty contributions. Inclusion of additional isotopic measurements in future validation efforts will increase the confidence in the ability of SAS2H to predict

spent fuel characteristics while simultaneously decreasing the excessive conservatism associated with the limited numbers of experimental comparisons. Additional measurements should include a broader range of fuel parameters (e.g., initial enrichments, burnups, and assembly designs). Although not directly applicable in the validation of depletion methods, ongoing experimental work (e.g., CERES [12] and SFSX [13]) will provide data to help in the validation of cross sections for selected actinides and fission products important in burnup credit applications.

Finally, note that the isotopic correction factors provided in this paper were derived for application to low-enriched PWR fuel assemblies, based on chemical assay measurements performed on PWR spent fuel. Furthermore, because many of the nonfissile actinides become fissile actinides in fast, highly enriched systems (e.g., ²³⁴U, ²³⁶U,

and ^{241}Am), the correction factors described herein would be nonconservative under such conditions. Care should be taken in applying these results to any system lying significantly beyond the range of conditions spanned by the experimental data.

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