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Strategies for Applying Isotopic Uncertainties in Burnup Credit

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STRATEGIES FOR APPLYING ISOTOPIC UNCERTAINTIES IN BURNUP CREDIT*

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Abstract

Uncertainties in the predicted isotopic concentrations in spent nuclear fuel represent one of the single largest sources of overall uncertainty in criticality calculations that use burnup credit. The methods used to propagate the uncertainties in the calculated concentrations to the uncertainty in the predicted neutron multiplication factor (k_{eff}) of the system can have a significant effect on the uncertainty in the predicted safety margin and ultimately can affect the potential capacity of spent fuel storage casks. This report surveys several different best estimate strategies for considering the effects of isotopic uncertainties in burnup credit analyses and illustrates the impact of these strategies on the predicted k_{eff} for a prototypical burnup credit cask design. The reactivity margin associated with the more realistic best estimate strategies is discussed in comparison to the reactivity margin associated with conventional bounding methods of uncertainty propagation. The experimental database of isotopic uncertainties used in the study has been significantly expanded to include new high-enrichment and high-burnup spent fuel assay data recently published for burnup credit nuclides. Expanded rare earth fission product measurements that contain the only known publicly available measurement for ^{103}Rh have also been included. This expanded experimental database is reviewed together with the bias and uncertainties obtained using the SCALE code system.

1. INTRODUCTION

Over the past decade, there has been a concerted effort in the United States and other countries to use more accurate and realistic estimates of the reactivity worth of spent nuclear fuel (SNF) in licensing of spent fuel storage and transportation systems by applying burnup credit. Criticality safety analyses have traditionally assumed that the fuel is unirradiated, which has clearly led to considerable safety margins. The process for performing criticality calculations in a burnup credit model requires two distinct steps — the first to predict the spent fuel isotopic concentrations using burnup calculations; the second, to perform a criticality calculation using the isotopic concentrations estimated in the first step. Consideration of the depletion phenomena in the criticality assessment significantly increases the overall complexity of a criticality safety analysis, places increased demands and reliance on computational tools and methods, and necessitates consideration of many additional uncertainties associated with the fuel compositions that are not encountered in analyses that assume the fuel is unirradiated.

The U.S. Nuclear Regulatory Commission (NRC) issued Revision 1 of Interim Staff Guidance 8 (ISG-8) in July 1999, to provide guidance on the application of limited burnup credit in criticality safety analyses for pressurized-water reactor (PWR) spent fuel in transportation and storage casks [1]. This ISG-8 guidance states,

> *"The applicant should ensure that the analysis methodologies used for predicting the actinide compositions and determining the neutron multiplication factor (k-effective) are properly validated. Bias and uncertainties associated with predicting the actinide compositions should be determined from benchmarks of applicable fuel assay measurements."*

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The analyst is ultimately required to assess the impact of the nuclide bias and uncertainty on the predicted neutron multiplication factor (k_{eff}) for the system. Unfortunately, there is currently no guidance or consensus on how the bias and uncertainties should be propagated to the k_{eff} in a burnup credit analysis. The different approaches to considering the nuclide uncertainties in this step of the analysis can have a significant effect on the predicted criticality safety margin and ultimately impact the number and types of spent fuel assemblies that may be considered acceptable for loading in a transport or storage cask.

This report reviews and illustrates several different potential strategies for considering the effects of nuclide uncertainties in burnup credit. These strategies include conventional bounding and best estimate methods to estimate the net effect of nuclide uncertainties. The reactivity margins associated with the different methods are compared for PWR spent fuel loaded in a prototypic burnup credit cask. The nuclide uncertainties used in these studies are based on a SCALE code system analysis of revised and expanded isotopic assay data publicly available in the United States. Recent publication of radiochemical assay measurements for the Japanese Takahama-3 reactor fuel has significantly enhanced the database in terms of both the number of measurements and the enrichment and burnup range of the experimental data. Recently published fission product measurements performed by the Khlopin Radium Institute in Russia for rare earth fission products are also included. The Khlopin measurements include the only known publicly-available measurement for the major fission product absorber ^{103}Rh.

2. METHODS OF UNCERTAINTY PROPAGATION

The different approaches used for treating uncertainties in complex calculational models are generally grouped as either "bounding" methods that use statistically bounding values to conservatively account for individual parameter uncertainty, or "best estimate" methods that use average parameter values and attempt to estimate realistically the effects of parameter uncertainties on the final results by using Monte Carlo (probabilistic) methods or other techniques (e.g., sensitivity analysis). Best estimate methods can provide a more accurate representation of the effects of nuclide uncertainty in a subcritical burnup-credit spent fuel system. However, using a best estimate approach is more complicated because of the additional effort required to determine accurately the uncertainty in the calculated k_{eff} for the system. The available criticality safety margin is underestimated because bounding methods tend to significantly overestimate k_{eff},. Thus, the subcritical margin predicted using best estimate methods would actually be larger than that obtained using a bounding approach. The uncertainty predicted using such realistic methods may be used to derive tolerance intervals to achieve any desired level of statistical confidence. Best estimate methods provide an improved understanding of the uncertainties and can help reduce the subcritical margin needed in burnup credit safety analyses, resulting in fewer casks needing to be transported and decreasing the regulatory burden on licensees while maintaining safety for transporting SNF.

2.1. Bounding Approach

In a conventional bounding approach, the analysis assumptions and input parameters are simultaneously set to their limiting (maximum or minimum) values to produce the most conservative final result. As applied to nuclide uncertainties in burnup credit, this approach applies conservatively bounding values for the predicted concentration of each nuclide. In the bounding model, the calculated concentrations are adjusted in a way that always leads to a more reactive system. In other words, the concentration of fissile nuclides is always increased, while the concentration of absorbing nuclides is always decreased, in order to maximize the k_{eff} of the system. Each nuclide is adjusted for the average bias in the burnup calculation, and a statistically bounding estimate of uncertainty based on the nuclide standard deviation (from calculated-to-experimental ratio) and a multiplier on the uncertainty to achieve the desired level of statistical confidence for the number of samples available. This approach ensures that the estimated uncertainty associated with the prediction of the nuclide inventories will be bounding. Such a conservative approach was proposed by the United States Department of Energy (U.S. DOE) in support of transportation and storage burnup credit [2]. Although this approach assures that the uncertainty in the predicted k_{eff} will be very conservative, simultaneously adjusting all nuclide concentrations to their statistical extremes is not realistic and masks the true importance of the uncertainties on the criticality calculation. However, such an approach is simple, easy to defend, and yields

concentrations that when used in a criticality calculation, will provide an upper bounding estimate of the k_{eff} for the system.

2.2. Best Estimate Approaches

Several best estimate methods can be used to propagate uncertainties from individual parameters contributing to the global k_{eff} uncertainty. Three best estimate methods are discussed and illustrated in this paper:

1. Monte Carlo uncertainty sampling,
2. Sensitivity/Uncertainty method, and
3. a "direct difference" method.

2.2.1. Monte Carlo Sampling Method

The total uncertainty in a computed quantity may be estimated using a technique that involves Monte Carlo (stochastic) sampling of the uncertainty probability distributions of the different parameters used in a calculation. Unlike a bounding calculation performed using a single set of conservative parameter values, the Monte Carlo (MC) approach undertakes multiple calculations with changes to the input parameters that reflect the random uncertainty distributions of the parameters. These multiple calculations yield a distribution of results from which the mean and probability of exceeding a particular value or threshold can be determined statistically.

For burnup credit calculations, the technique involves stochastically varying the nuclide concentrations according to the uncertainty in the predicted concentration of each burnup credit nuclide (i.e., related to the standard deviation). The shape of the probability distributions (typically normal) and uncertainties are established by comparing measured and computed nuclide concentrations in SNF.

The practical implementation of this approach requires automating the Monte Carlo stochastic sampling to determine the needed nuclide concentration sets and apply each set in the specified criticality analysis model. To illustrate this approach, a Monte Carlo sampling method was implemented at Oak Ridge National Laboratory (ORNL) to estimate the effects of nuclide uncertainties in burnup credit analyses using a computer program called KRONOS [3] designed to interact within the SCALE code system. The criticality calculations are repeated automatically, until the mean k_{eff} and probability distribution in k_{eff} from the nuclide uncertainties have converged. The average bias and uncertainty for each nuclide, used to define the sampling distribution, are input directly to the code. KRONOS will perform either a SCALE CSAS1X one-dimensional (1-D) XSDRNPM calculation or a CSAS25 three-dimensional (3-D) KENO V.a criticality calculation for the system. Due to the large number of criticality calculations required by this method to provide statistically reliable results, the code was developed for parallel processing on a distributed network environment to minimize the time to complete an analysis.

2.2.2. Sensitivity/Uncertainty Method

Sensitivity/uncertainty (S/U) analysis techniques have been widely used as a means of quantifying the effect of input data and other parameters on computer model predictions. These methods are used to develop sensitivity coefficients for the system that reflect the change in the calculated response to a change in an input parameter. To a first order, a sensitivity coefficient of 1.0 means that a 1% change in the parameter will cause a 1% change in the result. Combined with uncertainty information, sensitivity techniques can provide a powerful tool to estimate the global system uncertainty.

With this approach the relative change in the k eigenvalue due to a change in the concentration of nuclide N is expressed through first-order accuracy by the linear relationship

$$\frac{dk}{k} = S_N \frac{dN}{N} \quad ,$$

were the proportionality constant S_N is the sensitivity coefficient of k to the nuclide concentration N. This technique provides a straightforward method of predicting the uncertainty in the k_{eff} given the variation in nuclide concentration that is attributed to the nuclide uncertainty. The nuclide uncertainty, expressed as a relative change in the concentration, dN/N, may be obtained for the important burnup credit nuclides by comparing predicted and measured nuclide concentrations in SNF. The uncertainty from multiple nuclides may be combined to provide a measure of total uncertainty. If the uncertainties are assumed to be uncorrelated or random, they may be combined using a root-mean-square (RMS) approach. The total uncertainty was calculated using the expression

$$\frac{dk}{k} = \sqrt{\sum_{i=1}^{n} \left(\frac{dN}{N} \{S_N\} \right)^2} \quad ,$$

where the sum is performed over all n burnup credit nuclides in the criticality analysis. Note that if the uncertainty from each nuclide is combined additively (using the absolute values of the sensitivity coefficients S_N), then the result is equivalent to the uncertainty predicted using the conventional bounding approach.

Sensitivity coefficients used in this work were generated for a generic burnup-credit cask model using SEN35, a prototypic SCALE code sequence that implements sensitivity analysis techniques for 3-D Monte Carlo criticality calculation [4]. The methods used to generate the sensitivity information are based on the widely-used perturbation theory approach [5]. SEN35 calculates forward and adjoint neutron fluxes using an enhanced version of the KENO V.a Monte Carlo criticality code. Once the fluxes are obtained, the SAMS module (**S**ensitivity **A**nalysis **M**odule of **S**CALE) produces flux moments and calculates the sensitivity coefficients from these data and the cross-section data. SAMS also calculates the uncertainty in the sensitivity coefficients due to the Monte Carlo uncertainties. The motivation and principal application of the SEN35 sequence has been the need for modern computational tools that can generate sensitivity data necessary to gauge the applicability of validation experiments used for criticality studies [6]. SEN35 provides sensitivities for the various partial and total cross sections in the criticality calculations. However, the sensitivity coefficient for the nuclide total cross section is equal to that for the nuclide concentration. This allows sensitivity coefficients for each nuclide (at a given concentration) to be obtained directly. To generate burnup-dependent sensitivity coefficients, multiple KENO V.a input files were created for SEN35, with each file containing the burnup-dependent compositions of the burnup credit nuclides in spent fuel. Sensitivity coefficients were generated for a uniform axial burnup and an axial burnup profile for the spent fuel assemblies.

2.2.3. Direct-Difference Method

Another best estimate technique has been explored recently at ORNL. Instead of evaluating the bias and uncertainty for individual nuclides used in a burnup credit calculations, the measured nuclide concentrations from radiochemical assay experiments are applied directly in a criticality calculation for the spent fuel configuration used in the intended burnup credit application. The k_{eff} calculated for the system is then compared to the value predicted using calculated nuclide concentrations for the same set of burnup credit nuclides. The difference (Δk) is a measure of the net bias in the k_{eff} calculation associated with the predicted nuclide concentrations, and the variation of the results obtained using multiple experimental data sets provides a direct measure of the uncertainty. In this paper, this technique is called the direct difference method.

Unlike the other methods, this approach deals with the aggregate effect of the nuclide uncertainties on k_{eff} directly, and does not required an analysis of the bias and uncertainty for any individual nuclide. Rather, the net effect of the uncertainty from all nuclides is determined simultaneously. This technique can be used to

estimate the bias and uncertainty in the neutron multiplication factor for a given set of burnup credit nuclides. The bias and uncertainty in the neutron multiplication factor are obtained from a statistical analysis of the distribution of Δk values obtained using measured and predicted nuclide concentrations. The variance in the Δk results is related to the uncertainty in the predicted nuclide compositions and can be used to estimate the net effect of the reactivity margin to any desired level of statistical confidence. The method does not require any *a prior* assumptions about the shape of the uncertainty distribution of individual nuclides, and does not assume that uncertainty in different nuclides is uncorrelated. In addition, the method may yield trends in the k_{eff} bias with fuel enrichment and/or burnup that may not be evident by analyzing individual nuclides. However, this method requires a comprehensive database of measured isotopic data for a common set of burnup credit nuclides. That is, an experimental data set must contain measurements for all nuclides selected for the burnup credit analysis in order to be used in this method. A sufficient number of measurements is needed to allow statistically reliable observations to be made about the uncertainty and trends in the k_{eff}. As the number of nuclides in a burnup-credit analysis increases, the number of appropriate experiments tends to decrease. Another limitation of the method is that only spent fuel with a uniform axial burnup can be simulated since assay data are not available for the range of axial burnup values required to simulate an axial profile.

3. REVIEW AND ANALYSIS OF EXPERIMENTAL DATA

Several benchmark studies have been published using measured data available in the U.S. to establish the bias and uncertainties in the predicted nuclide concentrations using specific code systems for PWR fuel. As part of this current study, a comprehensive review of publicly-available PWR isotopic assay data was undertaken with a re-analysis of the nuclide bias and uncertainties using the current version of the SCALE code system. The benchmark results provide a common set of nuclide uncertainty values that were applied to the different uncertainty propagation techniques. The data sets selected for this study include all PWR assays from Refs. [7] and [8], and the Yankee Rowe reactor assay data used in Ref. [9]. Details of the reactor descriptions, fuel descriptions, laboratories, and experimental methods can be found in the citations to the original work.

In addition to previous studies, recently published radiochemical assay data for spent fuel from the Japanese PWR Takahama-3 reactor were analyzed and added to the experimental database. These data represent an important addition to the available radiochemical assay data. The Takahama-3 data provide 10 new samples with extensive burnup credit nuclide measurements. These samples have an enrichment of 4.11 wt % ^{235}U and relatively high burnups, significantly extending the enrichment and burnup range of publicly available data. The updated radiochemical assay database includes a total of 56 individual spent fuel assay samples from seven different reactors. Table 1 summarizes the reactors, assembly designs, and basic fuel parameters. Nuclide measurements are available for all of the major burnup credit actinides [10]. Spent fuel assay data for the Mihama reactor, used previously in Ref. [9], were not selected for this study. A review [11] of the Mihama data indicated there was high variability in the measurements for fuel having similar burnups and fuel assembly locations, and erratic behavior was also observed for fuel samples taken from different axial positions of the same rod. Since the enrichment and burnup range of the Mihama measurements did not extend beyond the range already available from other data sets, these data were not added to the present study.

The amount of publicly available experimental fission product data is currently very limited. Measurements for fission products important to burnup credit are available in only a small subset of the fuel samples. The majority of fission product data come from measurements of the Calvert Cliffs Approved Test Material (ATM) samples performed at the Pacific Northwest Laboratory (PNL) Materials Characterization Center (MCC). Subsequent to the original ATM-series fission product measurements, the V. G. Khlopin Radium Institute in Russia performed an independent analysis of the three ATM-104 samples, and one sample from ATM-106. The isotopic measurements included Nd, Sm, Eu, and Gd for all samples. In addition, the ^{103}Rh content of the ATM-106 sample was measured. This measurement represents the only known publicly-available data for ^{103}Rh in spent fuel. The Khlopin results were only recently published [12] in an effort supported by the NRC for burnup-credit validation studies. In this study the Khlopin results were used for all

Nd, Sm, Eu, and Gd isotopes, except ^{145}Nd. Results for the important burnup credit Nd and Sm isotopes were also available for the Takahama-3 samples, significantly augmenting the number of measurements available for these isotopes. To date, no results for ^{95}Mo, ^{101}Ru, or ^{109}Ag for LWR fuel have been published in open literature.

Table 1. Summary of selected PWR spent fuel radiochemical assay data

Reactor	Lattice type	Enrichment (wt %)	Burnup (GWd/t)	Absorbers	No. of samples
Trino Vercellese	WE 15 × 15	3.13	11.5 – 24.5	CR[a]	13
		3.897	12.0		1
Turkey Point	WE 15 × 15	2.556	30.5 – 31.5		5
Obrigheim	CE 14 × 14	3.13	25.9 – 29.5		6
H. B. Robinson-2	WE 15 × 15	2.561	16.0 – 31.7	BPR[b]	4
Yankee Rowe	WE 17 × 18	3.4	16.0 – 36.0	CR	8
Calvert Cliffs	CE 14 × 14	3.038	27.4 – 44.3		3
		2.72	18.7 – 33.2		3
		2.453	31.4 – 46.5	BPR	3
Takahama-3	WE 17 × 17	4.11	14.3 – 47.3	BPR	10
Total		2.56 – 4.11	11.5 – 47.3		56

[a] CR = Assemblies exposed to control rods
[b] BPR = Assemblies with burnable poison rods

The radiochemical assay database was used to benchmark the computational methods of SCALE to provide a measure of the mean bias and uncertainty of each nuclide required for the different uncertainty propagation methods. All nuclide inventory calculations were performed with the SCALE 1-D depletion analysis sequence SAS2H [13] and the SCALE 44-group ENDF/B-V-based cross-section library.

4. UNCERTAINTY ANALYSIS RESULTS

This work deals only with the uncertainties in the predicted spent fuel nuclide compositions used as input to the criticality calculation. Additional uncertainties associated with other depletion phenomena (i.e., spatial variation of the burnup), operating history, and the actual criticality calculation itself (e.g., cross-section uncertainties, etc.) are not considered. The results in this report are intended to illustrate the typical uncertainty margins due to the predicted SNF inventory that can be expected in burnup credit analyses using different uncertainty analysis methods. In these current studies, the uncertainty in the reactivity margin was determined as the 2 standard deviation ($\pm 2\,\sigma$) uncertainty interval for the results. Tolerance factors that account for the additional uncertainty associated with limited sample sizes may also be applied. In this work tolerance factors were not applied for simplicity. The effect of tolerance factors will be very small for most of the actinides that have relatively large amounts of measured data, but would result in larger fission product uncertainties than shown in this current study.

The criticality calculations were performed using a common cask design, based on a conceptual generic rail-type burnup credit cask that would accommodate 32 fuel assemblies [14]. The spent fuel assembly was assumed to be a Westinghouse 17 × 17 OFA design. Burnup credit calculations were performed for a flat axial burnup and for an 18-axial-zone burnup profile derived for assemblies with an average burnup greater than 30 GWd/MTU [2].

4.1. Actinide-Only Burnup Credit

The uncertainty in the calculated k_{eff} for the spent fuel cask due to the uncertainties in the predicted nuclide concentrations using the MC and S/U best estimate methods are compared to the conventional bounding approach in Fig. 1 for actinide-only burnup credit with a uniform (flat) axial burnup distribution. The results for the axially-varying burnup profile are shown in Fig. 2. The uncertainty is expressed in units of reactivity ($\rho = \Delta k/k$). All of the important burnup credit actinides [10] were included in the criticality calculations. In all results presented here, the nuclide uncertainties were based on code validation results using the SCALE 4.4a code system using the updated radiochemical assay database. In the bounding analysis method the burnup-credit nuclide concentrations were simultaneously adjusted for uncertainty by an amount equal to the 2 sigma value determined from the comparisons of measured and calculated concentrations for each nuclide. For the Monte Carlo sampling calculations, the concentration of each nuclide was sampled according to a normal distribution defined by its 1 sigma uncertainty value, and the uncertainty in the k_{eff} value determined from the $\pm$ 2 σ uncertainty in the computed distribution of k_{eff} values calculated by the KRONOS code. For the KRONOS results 100 KENO V.a criticality calculations were performed to determine the k_{eff} value uncertainty associated with each point. For the S/U calculations the effects on the k_{eff} value due to a 2 σ perturbation in the nuclide concentrations was determined from the sensitivity coefficients calculation using SEN35, and propagated to the k_{eff} value using the RMS combination method. These methods were used to quantify the estimated uncertainty in an illustrative example using fuel with an initial enrichment of 3.5 wt % ^{235}U and burnup values from 10 to 60 GWd/MTU.

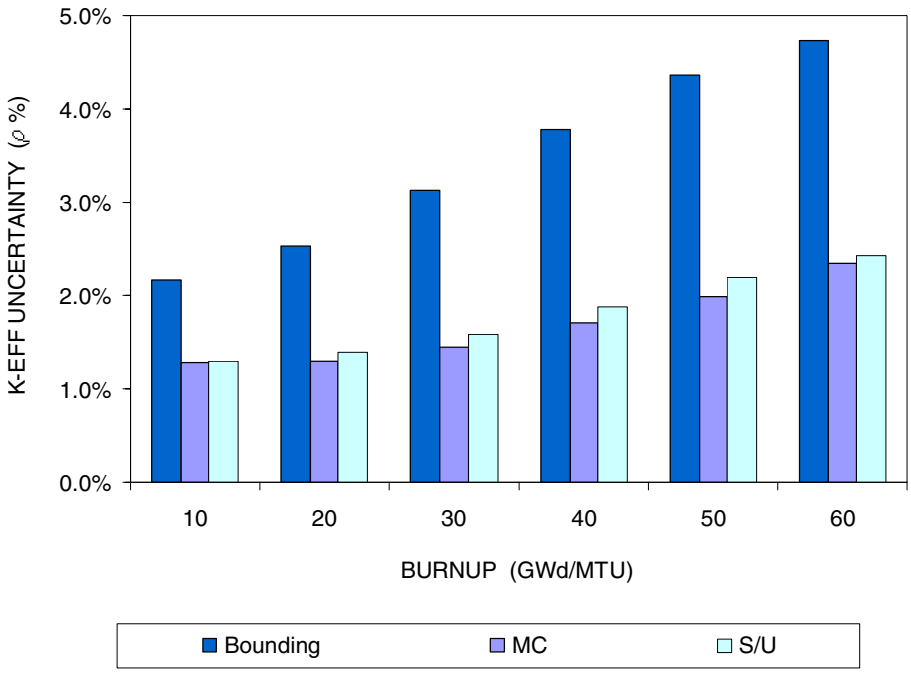

FIG. 1. Estimated uncertainty (2 sigma) in k_{eff} value, in units of reactivity, associated with predicted actinide compositions for a burnup credit cask and flat axial burnup distribution (3.5 wt % ^{235}U).

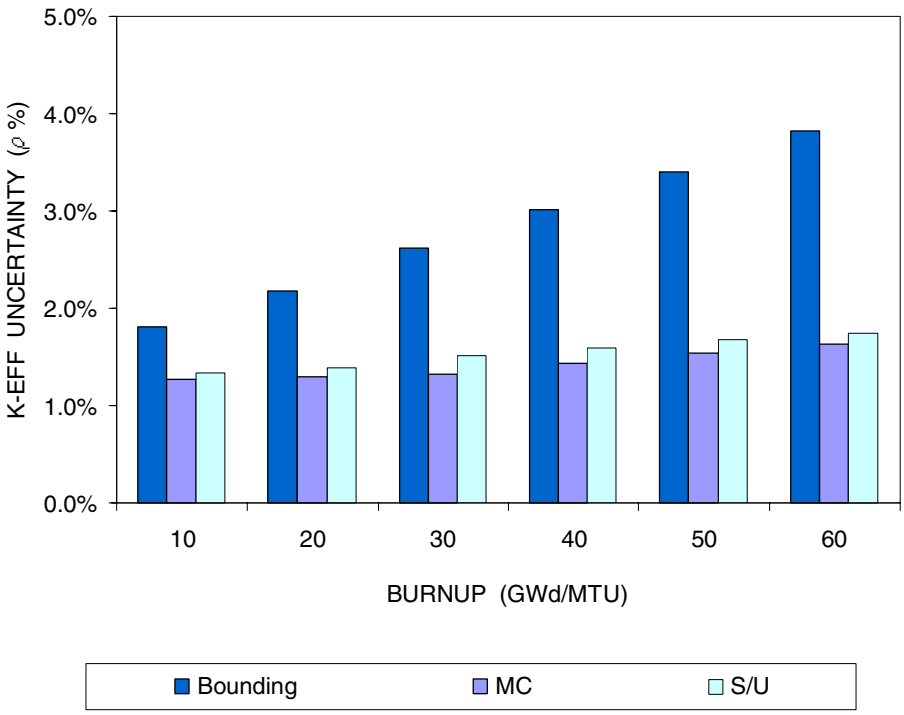

FIG. 2. Estimated uncertainty (2 sigma) in k_{eff} value, in units of reactivity, associated with predicted actinide compositions for a burnup credit cask and axially-varying burnup distribution (3.5 wt % ^{235}U).

The results in Figs. 1 and 2 show that the predicted levels of uncertainty (2 σ) are similar for the two different independent best estimate methods. The uncertainty in the k_{eff} value, expressed as reactivity, is observed to increase with increasing burnup, from about 1.2% at 10 GWd/MTU to 2.3% at 60 GWd/MTU for uniform axial burnup. The uncertainty decreases when an axial burnup profile is used due to the lower burnup at the ends of the assembly, which become increasing important to the reactivity of the fuel in burnup credit. The uncertainty predicted using the bounding approach is seen to be about double the value obtained using the best estimate methods.

Fig. 3 shows the results obtained using the direct difference method for the major uranium and plutonium isotopes ^{235}U, ^{236}U, ^{238}U, ^{239}Pu, ^{240}Pu, and ^{241}Pu plotted as a function of sample burnup. Radiochemical assay data for these actinides were available from all 56 experimental data sets evaluated. Combined, these nuclides represent typically > 90% of the relative reactivity worth for all actinides in spent fuel 5 years after discharge. Additional subsets of burnup credit nuclides have also been evaluated but are not shown here. However, as the number of actinides increases, the number of available measurements typically decreases. Consequently this approach attempts to balance the number of actinides available in the criticality calculation with the number of experiments in order to ensure an adequate number of comparisons are available to allow a reliable statistical interpretation of the results. The criticality calculations performed using this approach assumed a uniform axial burnup profile, since nuclide measurements were available for only a limited set of burnup values. The differences in the calculated k_{eff} value using the predicted nuclide inventories from SCALE and the measured inventories, expressed in units of reactivity[*] ρ, yield information about the bias and uncertainty associated with the predicted nuclide concentrations. Fig. 3 shows the data points for each fuel

[*] ρ % defined as $(k_m - k_c)/k_m \times 100$, where k_m and k_c are the k_{eff} values based on measured and calculated nuclide concentrations.

sample evaluated, and shows the linear regression fit and the ± 2 σ uncertainty interval from the values. The results indicate a small negative bias trend (increasing with burnup) and an average 2 σ uncertainty interval of ± 1.76%. A similar trend in the bias and uncertainty was found when the results are plotted as a function of sample enrichment.

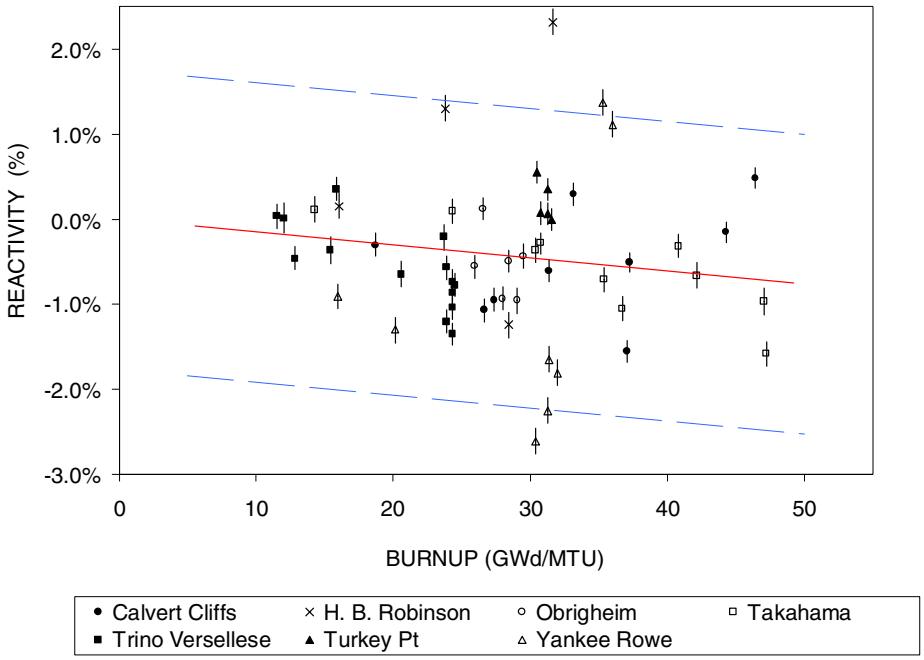

FIG. 3. Estimated reactivity effect (average bias and 2 sigma uncertainty interval) associated with predicted nuclide concentrations for six major actinides, ^{235}U, ^{236}U, ^{238}U, ^{239}Pu, ^{240}Pu, and ^{241}Pu. Results are plotted as a function of sample burnup, for a generic burnup credit cask.

The increase in the uncertainty with increasing burnup found in the Monte Carlo sampling and the S/U results is not evident in the direct difference results. However, there is an important distinction between the direct difference method and the other analysis methods. The direct difference results are based on experimental data with a wide range of enrichments and burnup values. In most cases (but not all) the sample burnup is commensurate with the initial enrichment. That is, as the enrichment increases, so does the discharge burnup. The results, therefore, reflect a proportionality between the enrichment and burnup. However, the Monte Carlo and S/U calculations were performed for variable burnup and a *fixed* initial enrichment (see Figs. 1 and 2). Therefore, the results in the low burnup regime reflects the uncertainty for fuel that has not achieved a typical burnup, while the results in the high-burnup regime reflects fuel that is overburned with respect to the initial enrichment, as compared to typical discharged fuel. The uncertainty determined using the Monte Carlo uncertainty approach for a burnup of 40 GWd/MTU, a value commensurate with the initial enrichment of 3.5 wt % ^{235}U used in the analysis, is about 1.7%. Note that this value is in good agreement with the results derived from the direct difference method of about 1.8% over the range of the experimental data.

4.2. Actinide and Fission Product Burnup Credit

Uncertainty calculations were repeated using both actinide and fission product credit. The quantity of measured radiochemical assay data for the fission products is considerably less that that available for the actinides, and for some major fission products (e.g., ^{95}Mo, ^{109}Ag, and ^{101}Ru) there are no known publicly available measurements. The limited quantity of radiochemical assay data makes a statistical evaluation of fission product nuclide uncertainties difficult and increases the uncertainty associated with the predicted concentrations. The lack of a comprehensive fission product database precludes the use of the direct difference method with fission products. Uncertainties were estimated using the MC and S/U methods using

nuclide uncertainties for ^{99}Tc, ^{103}Rh, ^{133}Cs, ^{143}Nd, ^{145}Nd, ^{147}Sm, ^{150}Sm, ^{151}Sm, ^{152}Sm, ^{151}Eu, ^{153}Eu, and ^{155}Gd. The one ^{103}Rh measurement provides an estimate of the calculational bias, but precludes an estimate of the uncertainty. For this study the uncertainty in ^{103}Rh was assumed to be nominally 30%. Nuclides with no measured data, ^{95}Mo, ^{109}Ag, and ^{101}Ru, were excluded from the criticality analysis. As noted previously, the addition of tolerance factors to the fission product results would have the effect of significantly increasing the estimated nuclide uncertainty for those nuclides with few measurements.

The uncertainty results for the best estimate methods are compared to the results for the bounding method in Fig. 4 for actinide and fission product credit. The bounding uncertainty predicted for the k_{eff} value is considerably larger when fission products are included. For calculations with a uniform (flat) axial burnup, the 2 σ uncertainty interval is about ± 8.4% at 60 GWd/MTU, compared to about ± 4.2% for actinide-only burnup credit. The uncertainties are somewhat less when an axial profile is included. The results using the best estimate methods are again observed to be similar to each other. The increase in the uncertainty when fission products are included is seen to be significantly less using the best estimate methods compared to the bounding approach. The best estimate 2 σ uncertainty interval for a uniform axial burnup was ± 3.0% at 60 GWd/MTU, compared to ± 2.2% when only the actinides are credited. The uncertainty in calculations that use an axial burnup profile are again observed to be smaller than those for the uniform axial burnup cases.

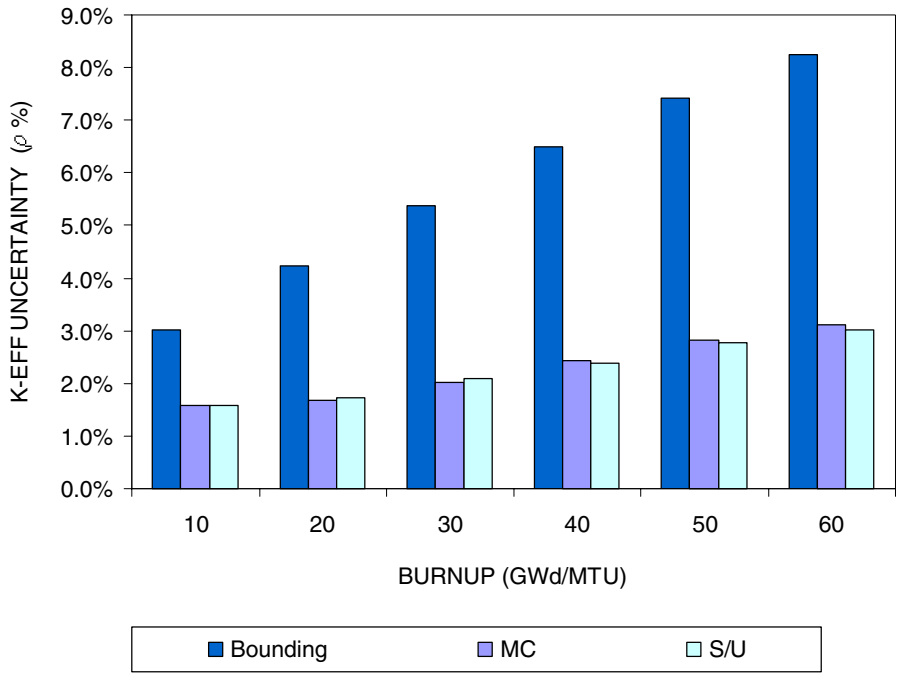

FIG. 4. Estimated uncertainty (2 sigma) in k_{eff} value, in units of reactivity, associated with predicted actinides and fission products compositions for a burnup credit cask and flat axial burnup distribution (3.5 wt % ^{235}U).

5. SUMMARY AND CONCLUSIONS

The results indicate that use of best estimate uncertainty methods will have a significantly reduce the level of uncertainty in the predicted k_{eff} compared to conventional bounding methods for actinide burnup credit calculations. The reduction is observed to be even greater when both actinide and fission product credit is considered. Bounding methods, while clearly conservative and easy to defend, are not realistic and result in higher uncertainties and an underestimation of the true safety margin in a criticality evaluation. The best estimate methods yield more realistic estimates of the effect of nuclide uncertainties, but require considerable more computational effort. An improved understanding of the uncertainties can help reduce the subcritical

margin needed in burnup credit safety analyses, thus resulting in fewer casks needing to be transported and reducing regulatory burden on licensees while maintaining safety for transporting SNF.

The uncertainty values presented in this paper are intended for illustrative purposes only since the results are based on the evaluation of a specific set of experiments using the SCALE code system. Also, the nuclide uncertainties have not included tolerance factors to account for the uncertainty from the sample size. The addition of tolerance factors would lead to larger fission product uncertainties than shown in this work but would not have a significant effect on the actinide results since the number of actinide measurements is relatively large.

The direct difference approach illustrated in this report provides a relatively straightforward and definitive method of predicting the net effect of nuclide uncertainties. The results are also consistent with the other independent best estimate methods. The direct difference method requires no ***a priori*** assumptions about the potential enrichment or burnup dependence of the nuclide uncertainties, correlation between different nuclides, or statistical shape of the nuclide uncertainties. In short, the method requires no evaluation of the individual nuclide uncertainties. In practice, this method could be used to estimate the reactivity associated with the nuclide uncertainties, as predicted using a given code system for a specific SNF configuration, and this reactivity would be incorporated into the subcritical margin. This is significantly different than conventional bounding approaches that have been proposed in the U.S. that apply uncertainties directly to the predicted individual nuclide concentrations.

One of the important findings from this work to burnup credit in the U.S. is that the accuracy of the calculated actinide concentrations for the high burnup Takahama-3 fuel samples is comparable to that observed in previous studies involving lower enrichment and lower burnup samples. The ISG-8 guidance on burnup credit recommends limiting the amount of credit for burnup to 40 GWd/MTU or less, and recommends a loading offset for fuel with an initial enrichment between 4.0 and 5.0 wt %. These restrictions on burnup and enrichment are based largely on the lack of sufficient radiochemical assay data above 40 GWd/MTU and 4.0 wt % (the majority of enrichments were under 3.4 wt %) for code validation at the time the guidance was issued. The recent publication and analysis of the Takahama-3 PWR radiochemical assay data significantly extends the range of the database for code validation. The Takahama-3 results indicate that the nuclide uncertainties for fuel exceeding 4 wt % and 40 GWd/MTU are not significantly different than lower enrichment and burnup fuels analyzed previously. This finding has potential implications for the current ISG-8 guidance and may provide a technical basis to support extending burnup credit beyond the present limits.

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REFERENCES

[1] U.S. Nuclear Regulatory Commission, Spent Fuel Project Office Interim Staff Guidance – 8, Rev. 1 – Limited Burnup Credit, U.S. NRC, July 30, 1999.

[2] U.S. Department of Energy, *Topical Report on Actinide-Only Burnup Credit for PWR Spent Nuclear Fuel Packages*, DOE/RW-0472, Rev. 2, Office of Civilian Radioactive Waste Management, U.S. Department of Energy, 1998.

[3] DeHART, M.D., *A Stochastic Method for Estimating the Effect of Isotopic Uncertainties in Spent Nuclear Fuel*, ORNL/TM-2001/83, UT-Battelle, LLC, Oak Ridge National Laboratory, September 2001.

[4] REARDEN, B.T., CHILDS, R.L., "Prototypical Sensitivity and Uncertainty Analysis Codes for Criticality Safety with the SCALE Code System," in *Transactions of the American Nuclear Society, 2000 International Winter Meeting and Embedded Topical Meetings*, Washington, DC, November 12–16, 2000. *Trans. Am. Nucl. Soc.* **83,** 98–100 (2000).

[5] BROADHEAD, B.L., REARDEN, B.T., "Foundations for Sensitivity-Based Criticality Validation Techniques," in *Transactions of the American Nuclear Society, 2000 International Winter Meeting and Embedded Topical Meetings*, Washington, DC, November 12–16, 2000. *Trans. Am. Nucl. Soc.* **83,** 93-95 (2000).

[6] BROADHEAD, B.L., HOPPER, C.M., PARKS, C.V., *Sensitivity and Uncertainty Analyses Applied to Criticality Safety Validation: Illustrative Applications and Initial Guidance*, NUREG/CR-6655, Vol. 2 (ORNL/TM-13692/V2), U.S. Nuclear Regulatory Commission, Oak Ridge National Laboratory, November 1999.

[7] DeHART, M.D., HERMANN, O.W., *An Extension of the Validation of SCALE (SAS2H) Isotopic Prediction for PWR Spent Fuel*, ORNL/TM-13317, Lockheed Martin Energy Research Corporation, Oak Ridge National Laboratory, September 1996.

[8] HERMANN, O. W., BOWMAN, S. M., BRADY, M. C., PARKS, C. V., *Validation of the SCALE System PWR Spent Fuel Isotopic Composition Analyses*, ORNL/TM-12667, Lockheed Martin Energy Research Corporation, Oak Ridge National Laboratory, March 1995.

[9] RAHIMI, M., FUENTES, E., LANCASTER, D., *Isotopic and Criticality Validation for PWR Actinide-Only Burnup Credit*, DOE/RW-0497, Office of Civilian Radioactive Waste Management, U.S. Department of Energy, May 1997.

[10] PARKS, C.V., DeHART, M.D., WAGNER, J.C., *Review and Prioritization of Technical Issues Related to Burnup Credit for LWR Fuel*, NUREG/CR-6665 (ORNL/TM-1999/303), UT-Battelle, LLC, Oak Ridge National Laboratory, 2001.

[11] HERMANN, O.W., DeHART, M.D., MURPHY, B.D., *Evaluation of Measured LWR Spent Fuel Composition Data for Use in Code Validation*, ORNL/M-6121, Lockheed Martin Energy Research Corp., Oak Ridge National Laboratory, February 1998.

[12] BRADY-RAAP, M.C., TALBERT, R.J., *Compilation of Radiochemical Analyses of Spent Nuclear Fuel Samples*, Pacific Northwest National Laboratory, PNNL-13677, September 2001.

[13] HERMANN, O.W., PARKS, C.V., "SAS2H: A Coupled One-Dimensional Depletion and Shielding Analysis Module," Vol. I, Sect. S2 of *SCALE: A Modular Code System for Performing Standardized Computer Analyses for Licensing Evaluation*, NUREG/CR-0200, Rev. 6 (ORNL/NUREG/CSD-2/R6), Vols. I, II, and III, May 2000. Available from Radiation Safety Information Computational Center at Oak Ridge National Laboratory as CCC-545.

[14] WAGNER, J.C., *Computational Benchmark for Estimation of Reactivity Margin from Fission Products and Minor Actinides in PWR Burnup Credit*, NUREG/CR-6747 (ORNL/TM-2001/306), U.S. Nuclear Regulatory Commission, Oak Ridge National Laboratory, October 2001.