

AN APPROACH FOR VALIDATING ACTINIDE AND FISSION PRODUCT BURNUP CREDIT CRITICALITY SAFETY ANALYSES— ISOTOPIC COMPOSITION PREDICTIONS*

G. Radulescu, I. C. Gauld, G. Ilas, and J. C. Wagner

Oak Ridge National Laboratory
P.O. Box 2008, Bldg. 5700, MS-6170
Oak Ridge, TN, U.S.A. 37831

RadulescuG@ornl.gov; GauldI@ornl.gov; IlasG@ornl.gov; and WagnerJC@ornl.gov

ABSTRACT

The expanded use of burnup credit in the United States for storage and transport casks, particularly in the acceptance of credit for fission products, has been constrained by the availability of experimental data to support code validation. U.S. Nuclear Regulatory Commission (NRC) staff has noted that the rationale for restricting the Interim Staff Guidance on burnup credit for storage and transportation casks (ISG-8) to actinide-only is based largely on the lack of clear, definitive experiments that can be used to estimate the bias and uncertainty for computational analyses associated with using burnup credit. To address the issues of burnup credit criticality validation, the NRC initiated a project with Oak Ridge National Laboratory to (1) develop and establish a technically sound validation approach for commercial spent nuclear fuel (SNF) criticality safety evaluations based on best-available data and methods and (2) apply the approach for representative SNF storage and transport configurations/conditions to demonstrate its use and applicability as well as to provide reference bias results. The purpose of this paper is to describe the isotopic composition (depletion) validation approach and resulting observations and recommendations. Validation of the criticality calculations is addressed in a companion paper at this conference. For isotopic composition validation, the approach is to determine burnup-dependent bias and uncertainty in the effective neutron multiplication factor (k_{eff}) due to bias and uncertainty in isotopic predictions via comparisons of isotopic composition predictions (calculated) and measured isotopic compositions from destructive radiochemical assay and a best-estimate Monte Carlo-based method. This paper (1) provides a detailed description of the burnup credit isotopic validation approach and its technical bases, (2) describes the application of the approach for representative pressurized water reactor and boiling water reactor safety analysis models to demonstrate its use and applicability, and (3) provides reference bias and uncertainty results based on a quality-assurance-controlled prerelease version of the Scale 6.1 code package and ENDF/B-VII nuclear cross section data.

Key Words: criticality, burnup, validation, transportation, spent fuel

1 INTRODUCTION

One of the most significant challenges associated with implementation of burnup credit is the validation of depletion and criticality calculations used in the safety evaluation—in particular, the availability and use of experimental data to support validation. The expanded use of burnup credit in the United States for storage and transport casks, particularly in the acceptance of credit for fission products, has been constrained by both the availability of experimental data to support code validation and a lack of a clear technical basis or approach for use of the data. U.S. Nuclear Regulatory Commission (NRC) staff has noted that the rationale for restricting the Interim Staff Guidance on burnup credit for storage and transportation casks [1] to actinide-only is based largely on the lack of clear, definitive experiments that can be used to estimate the bias and uncertainty for

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computational analyses. For burnup credit in spent fuel pools, an approach based on engineering judgment has been used to address uncertainties in fuel depletion calculations [2]. To address the issues of burnup credit criticality validation, the NRC initiated a project with Oak Ridge National Laboratory to (1) develop and establish a technically sound validation approach for commercial spent nuclear fuel (SNF) criticality safety evaluations based on best-available data and methods and (2) apply the approach for representative SNF storage and transport configurations/conditions to demonstrate its use and applicability as well as to provide reference bias results. This paper provides a detailed description of the burnup credit isotopic validation approach, its technical bases, and reference burnup-dependent bias and uncertainty results obtained for representative safety analysis models using a quality-assurance-controlled prerelease version of the Scale 6.1 code package [3] and the Scale 6.1 238-group cross-section library based on the Evaluated Nuclear Data File/B Version VII.0 (ENDF/B-VII) [4] nuclear data.

The validation of fuel isotopic compositions for criticality safety analyses is based on comparisons of isotopic composition predictions (calculated) and measured isotopic compositions from destructive radiochemical assay (RCA) data. A total of 28 nuclides [5] were considered in the fuel compositions based on their importance to fuel reactivity and on the availability of measurement data. The burnup credit nuclides include the 12 actinide nuclides ^{234}U , ^{235}U , ^{236}U , ^{238}U , ^{237}Np , ^{238}Pu , ^{239}Pu , ^{240}Pu , ^{241}Pu , ^{242}Pu , ^{241}Am , and ^{243}Am and the 16 fission product nuclides ^{95}Mo , ^{99}Tc , ^{101}Ru , ^{103}Rh , ^{109}Ag , ^{133}Cs , ^{143}Nd , ^{145}Nd , ^{147}Sm , ^{149}Sm , ^{150}Sm , ^{151}Sm , ^{152}Sm , ^{151}Eu , ^{153}Eu , and ^{155}Gd . An extensive database of recently compiled spent fuel isotopic data [6, 7] was used to determine the bias and uncertainty associated with calculated fuel concentrations. The data evaluations published in Refs. 6 and 7, based on primary experimental references, include information that enables two-dimensional (2-D) depletion calculation modeling and provide a database to validate computational predictions of spent fuel isotopic compositions important to criticality safety (burnup credit).

The method used to propagate the bias and uncertainty in the calculated nuclide compositions to the effective neutron multiplication factor (k_{eff}) values for spent fuel pool (SFP) storage and transportation cask configurations is based on Monte Carlo uncertainty sampling. This method enables simulation of nuclide composition variations within the range of depletion uncertainty and provides accurate statistical estimates of bias and uncertainty in k_{eff} due to bias and uncertainty in the calculated nuclide compositions. The Monte Carlo uncertainty sampling method was selected because it may be applied to any SNF compositions in the safety analysis models and to axially varying assembly burnup profiles.

Representative analysis models were developed for SFP storage rack and transport/storage cask configurations to provide reference k_{eff} bias and uncertainty values for burnup credit criticality safety analyses. The sensitivity of bias and uncertainty in k_{eff} to a range of parameters important to criticality safety analysis (for example, spent fuel depletion conditions, fuel assembly type, burnup, cooling time, axial burnup representation, pool rack design, soluble boron concentration, and cross-section data library) was also evaluated.

2 ISOTOPIC VALIDATION METHODOLOGY APPROACH

2.1 Isotopic Validation Data

Bias and uncertainty values associated with calculated nuclide concentrations in pressurized water reactor (PWR) SNF were based on direct comparisons to measured nuclide concentrations in 100 representative PWR fuel samples obtained from low-, moderate-, and high-burnup fuel assemblies irradiated in the following nine PWRs: Trino Vercellese, Kernkraftwerk Obrigheim, Turkey Point Unit 3, H.B. Robinson Unit 2, Calvert Cliffs Unit 1, Three Mile Island Unit 1, Takahama Unit 3, Gösgen, and GKN II. The samples cover fuel with initial enrichments varying

from 2.453 to 4.657 wt % ^{235}U and burnup varying from 7 to 60 GWd/MTU. The number of measured nuclides varies depending on the measurement program. The earlier programs generally provide measurement data for the uranium and plutonium isotopes, whereas the more recent programs provide measurement data for up to 50 nuclides, including nuclides important to burnup credit criticality safety, radiological dose, and decay heat generation.

The bias and uncertainty values associated with calculated nuclide concentrations in boiling water reactor (BWR) SNF were based on comparisons to measured nuclide concentrations for 32 BWR fuel samples obtained from fuel assemblies with 8×8 , 7×7 , and 6×6 pin lattices from the Fukushima Daini Unit 2, Cooper, and Gundremmingen-A reactors, respectively. The initial fuel enrichment for the samples varies from 2.54 to 3.91 wt % ^{235}U , and the burnup varies from 14.4 to 44.0 GWd/MTU. Measurement BWR data for the 12 burnup credit actinide nuclides and for eight of the 16 burnup credit fission product nuclides (^{99}Tc , ^{143}Nd , ^{145}Nd , ^{147}Sm , ^{149}Sm , ^{150}Sm , ^{151}Sm , and ^{152}Sm) were available for use in the isotopic validation. In addition, the validation results for the BWR SNF are subject to large uncertainties, primarily associated with modeling uncertainties in local void conditions [7]. Therefore, the BWR assay data are relatively limited in their range of applicability.

2.2 Bias and Uncertainty Associated with the Calculated Nuclide Compositions

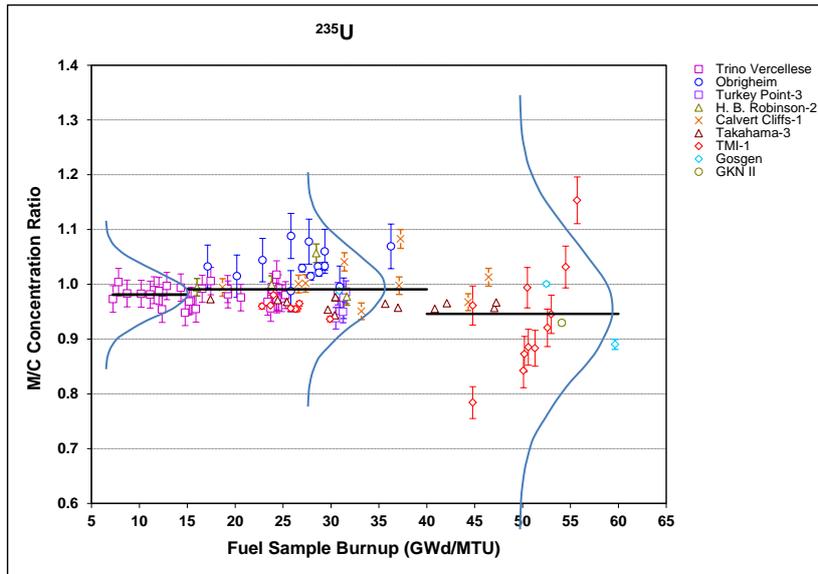
The Monte Carlo estimation method employed in the isotopic validation relies on the assumption of normality of data to make probabilistic inferences from samples to populations. The random variable of interest for simulating fuel composition variations within the range of uncertainty is the measured-to-calculated (M/C) nuclide concentration ratio. Nuclide concentrations in the evaluated fuel samples were calculated using the TRITON 2-D depletion sequence [8] and the 238-group ENDF/B-VII library in Scale 6.1. An evaluation of the statistical characteristics of the M/C concentration values was performed to determine whether or not the data sets may be adequately modeled with normal distributions and to establish the parameters of the distributions. For nuclides with fewer than 10 measurement data points (^{133}Cs and ^{99}Tc for the PWR and BWR fuel, respectively), the M/C concentration values were assumed to conform to a continuous uniform distribution [9]. The statistical evaluations included a trending analysis used to identify relationships between the M/C concentration and sample burnup and initial enrichment, histogram plots used to visualize the shapes of frequency distributions, and the Shapiro-Wilk normality test at the 0.05 level used to identify data sets that do not approach the normal distribution. Identified statistical characteristics were taken into account when establishing the values of bias and uncertainty associated with the calculated nuclide compositions. The mean of the M/C concentration values is referred to as the isotopic bias; the standard deviation of a normal distribution model or the parameter of a uniform distribution model established as described below is referred to as the isotopic uncertainty.

In the case of the PWR isotopic evaluations, the trending analysis identified a dependence of the M/C concentration values on sample burnup for the major actinide nuclides ^{235}U , ^{236}U , ^{238}U , ^{239}Pu , ^{240}Pu , and ^{241}Pu . Correlation between the M/C concentration values and the initial enrichment of the measured PWR fuel samples was not identified for any of the burnup credit nuclides. The approach used to address the observed burnup dependence was to determine the isotopic bias and uncertainty values based on M/C concentration values from measured PWR fuel samples within a certain burnup interval. The number of burnup intervals varied from one to three, depending on the number of measurement data available for each burnup credit nuclide. For nuclides with a large number of measurements, including the actinide nuclides ^{235}U , ^{238}U , ^{239}Pu , ^{240}Pu , ^{241}Pu , and ^{242}Pu , three different sets of isotopic bias and uncertainty values were determined for three burnup intervals, including burnup values lower than 15 GWd/MTU, from 15 to 40 GWd/MTU, and greater than 40 GWd/MTU. Two different sets of isotopic bias and uncertainty values applicable to burnup

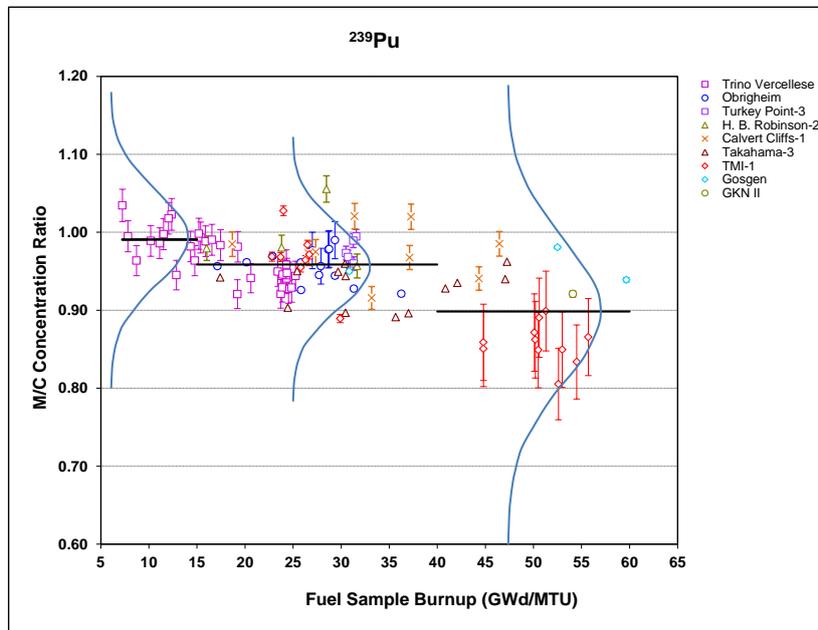
values below and above 40 GWd/MTU were determined for the actinide nuclides ^{234}U , ^{236}U , ^{237}Np , ^{238}Pu , ^{241}Am , and ^{243}Am . A single set of isotopic bias and uncertainty values was determined for each burnup credit fission product nuclide since a smaller number of measurement data were available (e.g., from 7 for ^{133}Cs and up to 44 for ^{143}Nd and ^{145}Nd). By employing this approach, the bias and uncertainty values within each burnup interval were based on sets of M/C concentration values that do not exhibit a dependence on burnup, as illustrated in Figure 1 (a) and (b) for ^{235}U and ^{239}Pu , respectively. In the case of the BWR isotopic data evaluations, the isotopic bias and uncertainty values were based on the entire set of available BWR RCA data because dependencies on fuel sample initial enrichment and final burnup were not identified for any of the burnup credit nuclides with measured concentrations.

Since a limited number of measurement data were available for use in the burnup credit isotopic validation, the parameters of a normal distribution model characterizing uncertainties in nuclide concentrations were established based on tolerance intervals. A statistical tolerance interval defines the limits within which a stated proportion of a population is expected to lie, based on a sample that was measured from that population. The sample mean of the M/C concentration values served as the estimator for the mean of a distribution model; the sample standard deviation of the M/C concentration values adjusted for sample size served as an estimator for the standard deviation of the distribution model. For a normal distribution model, the adjustment (multiplication) factor was the two-sided tolerance-limit factor for a normal distribution, corresponding to the sample size, 95% certainty, and 68.3% of the population, which varied from 1.174 (69 samples) to 1.664 (11 samples). The one-sided tolerance-limit factor for a normal distribution, corresponding to the sample size, 95% certainty, and 95% of the population, was used as an adjustment factor to determine the extent for a uniform distribution model. The one-sided tolerance limit factor values used for six and seven samples were 3.711 and 3.399, respectively. Therefore, greater adjustments were applied to the uncertainty values of nuclides with small sample sizes.

The M/C concentration values for some of the nuclides and burnup intervals did not pass the normality test. Variations in the magnitude of the uncertainty associated with measurement and depletion calculation modeling data across the evaluated fuel samples may affect the M/C concentration values in a nonuniform manner, thus producing anomalies in the frequency distributions of the M/C concentration values. A few outliers among some of the M/C concentration values were observed to produce skewed unimodal frequency distributions. An example of skewed unimodal frequency distribution is the set of M/C concentration values for ^{235}U based on the PWR fuel samples within the burnup interval between 15 and 40 GWd/MTU. However, the distribution models determined as described above bound the tails of the frequency distributions in a manner that is conservative with respect to criticality (e.g., higher ^{235}U concentrations will be sampled using the distribution model as compared to the fit distribution). Therefore, even if not all the sets of the M/C concentration values passed the normality test, those data sets may be adequately modeled with normal distributions to produce nuclide concentrations that are conservative with respect to criticality. As illustrated by the shapes of the normal distribution models shown for ^{235}U and ^{239}Pu in Figure 1 (a) and (b), respectively, the distribution models are broad enough to encompass the M/C concentration values and associated measurement errors.



(a)



(b)

Figure 1. M/C concentration versus fuel sample burnup for (a) ^{235}U and (b) ^{239}Pu .

Note: The error bars in the graph represent the reported one-sigma measurement errors; very small measurement errors (0.1%) are not visible on the graph; the bell-shaped curves shown on the graph suggest the sampling distributions used in the Monte Carlo simulations.

2.3 Monte Carlo Uncertainty Sampling Method

A stochastic Monte Carlo approach [9] was applied to estimate the bias and uncertainty in k_{eff} due to bias and uncertainty in the calculated nuclide compositions. The Monte Carlo uncertainty sampling method was selected because it may be applied to any SNF compositions in the safety analysis models and to axially varying assembly burnup profiles. The Monte Carlo method samples from distribution models developed for nuclide composition uncertainties. The sampled nuclide compositions are subsequently used in a safety analysis model to calculate a series of k_{eff} values from which the values of bias and uncertainty in k_{eff} are determined. This method is computationally intensive because a significant number of simulations are necessary to ensure that the sampled

uncertainty values are representative of the underlying probability distributions and to ensure convergence of the Monte Carlo estimates for the bias and uncertainty in k_{eff} . The direct difference method [10] was also used in this work for comparison purposes.

Random numbers drawn from either the standard normal distribution (i.e., the normal distribution with the distribution mean of zero and standard deviation of unity) or from the uniform distribution of parameters -1 and 1 were used as shown in Eq. (1) to simulate random nuclide composition variations within the range of depletion uncertainty,

$$c_{n,b}^k = \begin{cases} c_{n,b}(\bar{X}_n^b + \sigma_n^b R_n^k|_{NR}), & \text{if } N_n \geq 10 \\ c_{n,b}(\bar{X}_n^b + \sigma_n^b R_n^k|_{UN}), & \text{otherwise} \end{cases}, \quad (1)$$

where

- n = the index of a burnup credit nuclide considered in SNF compositions,
- k = the index of a criticality calculation,
- $c_{n,b}^k$ = concentration of nuclide n in a fuel mixture of burnup b for criticality calculation k adjusted for isotopic bias and uncertainty,
- $c_{n,b}$ = predicted concentration of nuclide n in a fuel mixture of burnup b ,
- \bar{X}_n^b = isotopic bias for nuclide n corresponding to the burnup b of the fuel mixture,
- σ_n^b = isotopic uncertainty for nuclide n corresponding to the burnup b of the fuel mixture,
- $R_n^k|_{NR}$ = random number sampled from the standard normal distribution,
- $R_n^k|_{UN}$ = random number sampled from the uniform distribution ranging from -1 to 1 .

Equation (1) was further constrained so that nonphysical values could not be sampled. Nonphysical values consist of either negative concentration values that may be obtained in the case of nuclides with very large isotopic uncertainty values or ^{235}U and ^{238}U sampled concentrations exceeding the initial concentrations in fresh fuel that may occur for small burnup values. For a statistically significant number of Monte Carlo simulations, the k_{eff} values approach a normal distribution with the mean and standard deviation given by Eqs. (2) and (3), respectively,

$$\bar{k}_{eff-MC} = \sum_{i=1}^{N_C} k_{eff}^i / N_C, \quad (2)$$

$$\sigma_{k_{eff-MC}} = \sqrt{\sum_{i=1}^{N_C} (k_{eff}^i - \bar{k}_{eff-MC})^2 / (N_C - 1)}, \quad (3)$$

where

- \bar{k}_{eff-MC} = the sample mean of the k_{eff} values from the Monte Carlo simulations,
- N_C = the number of realizations of nuclide compositions,
- k_{eff}^i = the k_{eff} value for criticality calculation i in the series of N_C criticality calculations,
- $\sigma_{k_{eff-MC}}$ = the sample standard deviation of the k_{eff} values from the Monte Carlo simulations.

The series shown in Eq. (2) converges to the k_{eff} value obtained by adjusting the predicted nuclide concentrations for isotopic bias only. The bias in k_{eff} was determined as the difference between the k_{eff} value based on the predicted nuclide compositions with no adjustments and the k_{eff} value based on the predicted nuclide compositions adjusted for isotopic bias. The sample standard

deviation of the k_{eff} values defined by Eq. (3) was used to determine the uncertainty in k_{eff} at a 95 percent probability, 95 percent confidence level.

For each representative analysis model and average assembly burnup considered, 500 different SNF compositions were generated in accordance with Eq. (1) and were used in criticality calculations. The results of a sensitivity analysis considering parametric variations in the representative PWR SFP storage rack model were based on 250 Monte Carlo simulations. Plots representing k_{eff} sample mean and standard deviation values versus number of simulated cases showed that the convergence of the statistical estimates is adequately achieved within 250 cases. For example, the sample standard deviation of the k_{eff} values was 0.0085 based on 2000 simulations, varied from 0.0083 to 0.0086 based on 500 simulations, and varied from 0.0082 to 0.0088 based on 250 simulations for the representative PWR SFP storage rack model and an assembly average burnup of 40 GWd/MTU. Using the sensitivity and uncertainty analysis method [10], it was determined that the uncertainties associated with the calculated ^{235}U and ^{239}Pu concentrations have a dominating effect on the uncertainty in k_{eff} , thereby causing a relatively fast convergence of the Monte Carlo estimates.

The bias and uncertainty values for the calculated nuclide concentrations were derived directly from measurement data for fuel samples from assemblies of different design and irradiation conditions that were provided by several independent measurement programs. The Monte Carlo uncertainty sampling procedure inherently assumes that the uncertainties associated with calculated individual nuclide concentrations are independent variables. That is, sampling of each nuclide is independent and uncorrelated. The independence of random variables is an approximation employed in this analysis. Complex multivariate statistical analyses [11, 12], which attempt to establish mathematical relationships between variables considered to be relevant to the problem being studied, require detailed understanding of both the calculational and experimental uncertainties and correlations. The appropriateness of the assumption of independent uncertainties was evaluated by performing confirmatory calculations using the direct difference method [10]. This method applies measured nuclide compositions directly in the criticality models and compares the k_{eff} values obtained using calculated and measured nuclide compositions. Although the method cannot analyze the full range of scenarios possible with the Monte Carlo sampling method, the direct difference calculations require no assumptions regarding either the normality of the distributions or the independent nature of the uncertainty data. The calculations were performed using a complete set of the 28 burnup credit actinide and fission product nuclides. For the experiments that did not measure all nuclides considered in the burnup credit analysis, calculated values were applied for missing data in order to provide a consistent basis to compare the results obtained from different experiments. The results of the direct difference method were very similar to those obtained by applying the Monte Carlo uncertainty method, which indicates that the approximation used in the Monte Carlo uncertainty sampling approach does not introduce an additional uncertainty component.

2.4 Representative Safety Analysis Models

Representative safety analysis models for PWR SFP storage rack, BWR SFP storage rack, and PWR SNF transportation/storage cask configurations were developed to demonstrate the application of the burnup credit isotopic validation approach and to provide reference values for the bias and uncertainty in k_{eff} due to bias and uncertainty in the calculated nuclide compositions. The PWR SFP storage rack model was a laterally infinite array of loaded fuel storage cells reflected on the top and bottom by 30 cm of full-density water. One 0.203 cm (0.080 in.) thick Boral® plate with a ^{10}B areal density of 0.020 g/cm² was modeled between each storage cell to the same axial length as the active fuel. The representative safety analysis model for a PWR SNF cask was a generic high-capacity cask design, referred to as a “GBC-32 cask,” which has been developed in Ref. 13 as a reference

configuration for burnup credit studies. The generic cask, which can accommodate 32 PWR assemblies, uses Boral® panels containing ^{10}B as a fixed neutron poison dispersed uniformly with a ^{10}B areal density of 0.0225 g/cm^2 . The fuel assembly type selected for use in the PWR SFP storage rack and SNF cask models was the Westinghouse 17×17 optimized fuel assembly. This assembly type was selected because it is one of the most reactive assemblies in a cask configuration [13]. Axially, the fuel rods were represented in the model as 18 zones with varying burnup. The burnup values of the axial fuel zones were determined using bounding axial burnup profiles with respect to criticality [14]. Axially varying burnup-dependent fuel compositions were generated using reactor operating parameters for depletion calculations that result in spectrum hardening and increased discharge reactivity [5]. The SNF compositions consisted of either the 12 actinide nuclides (actinide-only compositions) or of the 28 actinide and fission product nuclides. The approach employed for the PWR burnup credit criticality analysis used a range of initial enrichment and assembly average burnup values based on the loading curve constrained to fuel initial enrichment values below 5%. The PWR SFP storage rack model with zero ppm soluble boron and burnup-dependent fuel compositions corresponding to a three-day cooling time had a k_{eff} value of 0.99; the PWR SNF cask model with zero ppm soluble boron and burnup-dependent fuel compositions corresponding to a five-year cooling time had a k_{eff} value of 0.94.

The BWR SFP storage rack was modeled as a laterally and axially infinite array of loaded fuel storage cells. One 0.203 cm thick (0.080 in. thick) Boral® plate with a ^{10}B areal density of 0.020 g/cm^2 was modeled between each storage cell. The axial representation of the assembly burnup was uniform. The model for a BWR fuel assembly was a generic 10×10 -8 assembly design with eight gadolinia (Gd_2O_3) rods and two water rods that displaced eight fuel rods. A 5 wt % ^{235}U initial enrichment of the UO_2 fuel was used to account for the highest anticipated fuel enrichment. The gadolinia content in UO_2 was 3 wt %. The BWR analysis used the fuel composition corresponding to the assembly peak reactivity achieved during irradiation. The BWR model with zero ppm soluble boron and fuel composition corresponding to a burnup value of approximately 11 GWd/MTU and a three-day cooling time had a k_{eff} value of 0.94.

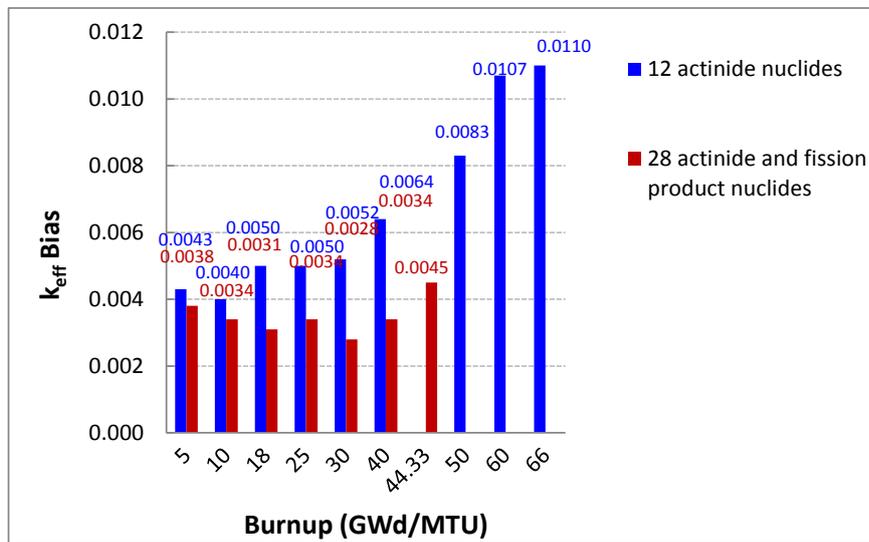
2.5 Parametric Variations in the Representative PWR SFP Rack Model

An analysis was performed to determine the sensitivities of k_{eff} bias and uncertainty to parameters important to criticality safety analyses. Parametric variations from the representative PWR SFP storage rack model include fuel assembly design (Babcock and Wilcox 15×15), fuel irradiation conditions (no WABA rods), pitch size of rack cell (reference value + 1.27 cm), ^{10}B areal density (0 g/cm^2 , 0.018 g/cm^2 , 0.022 g/cm^2), soluble boron concentration (1000 ppm; boron concentrations yielding a target k_{eff} value of 0.94), fuel cooling time (5, 20, and 40 years), axial representation of fuel burnup in the safety analysis model (uniform). The 44-group ENDF/B-V library and the Nordheim Integral Treatment for self-shielding were also used in depletion and criticality calculations to evaluate their impact on k_{eff} bias and uncertainty. The SNF compositions consisted of the 28 burnup credit nuclides; the fuel assembly had average burnup values of 10, 25, and 40 GWd/MTU with the nuclide concentrations being determined to yield the target k_{eff} value of 0.99 for all cases except for the soluble boron cases.

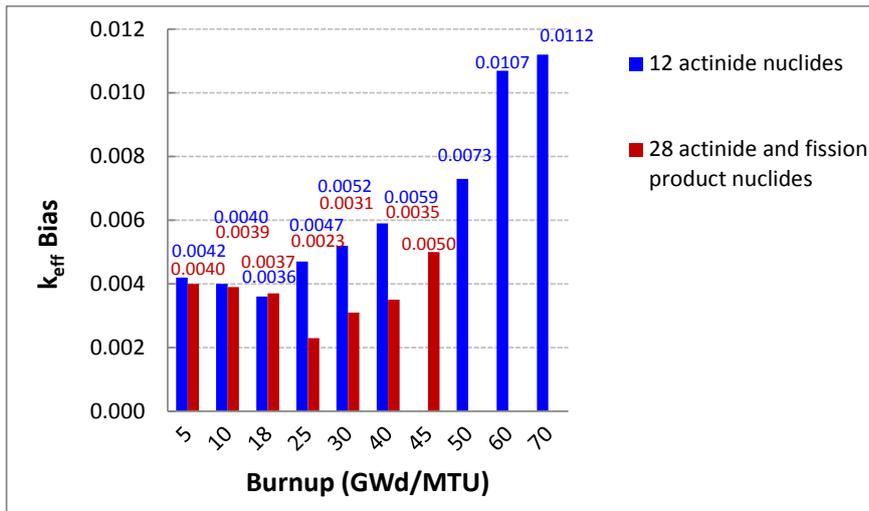
3 RESULTS

Reference values of bias and uncertainty in k_{eff} due to bias and uncertainty in the calculated SNF nuclide compositions for representative safety analysis models as well as the effects of variations in parameters important to criticality safety analysis on k_{eff} bias and uncertainty are presented in this section. The values of uncertainty in k_{eff} presented throughout this paper correspond to a 95 percent probability, 95 percent confidence level.

Similar k_{eff} bias and k_{eff} uncertainty values were calculated for the two representative PWR safety analysis models. The values of bias and uncertainty in k_{eff} for the representative analysis models are shown as a function of assembly average burnup in Figures 2 and 3, respectively. The calculated k_{eff} bias values were all positive and significantly smaller than the calculated k_{eff} uncertainty values. The k_{eff} bias values varied from ~ 0.004 to ~ 0.011 for the actinide-only nuclide compositions and from ~ 0.002 to ~ 0.005 for the actinide and fission product nuclide compositions, depending on assembly average burnup. Typically, positive k_{eff} bias values are not credited in criticality safety analyses. The k_{eff} uncertainty values varied from ~ 0.013 to ~ 0.025 for the actinide-only nuclide compositions and from 0.014 to ~ 0.020 for the actinide and fission product nuclide compositions, depending on assembly average burnup. Higher bias and uncertainty values were obtained for assemblies with average burnup greater than 40 GWd/MTU because the uncertainties associated with the calculated uranium and plutonium nuclide concentrations are based on fewer fuel samples than those for burnup values less than 40 GWd/MTU.

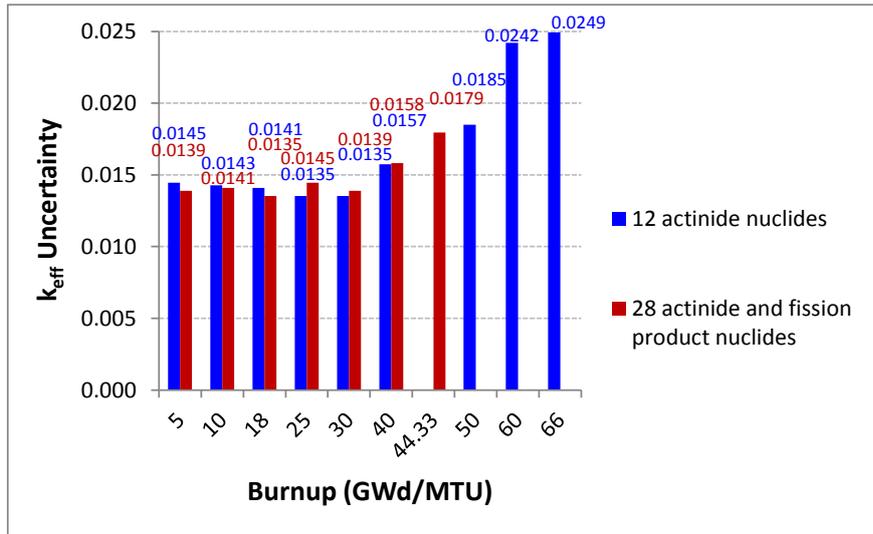


(a)

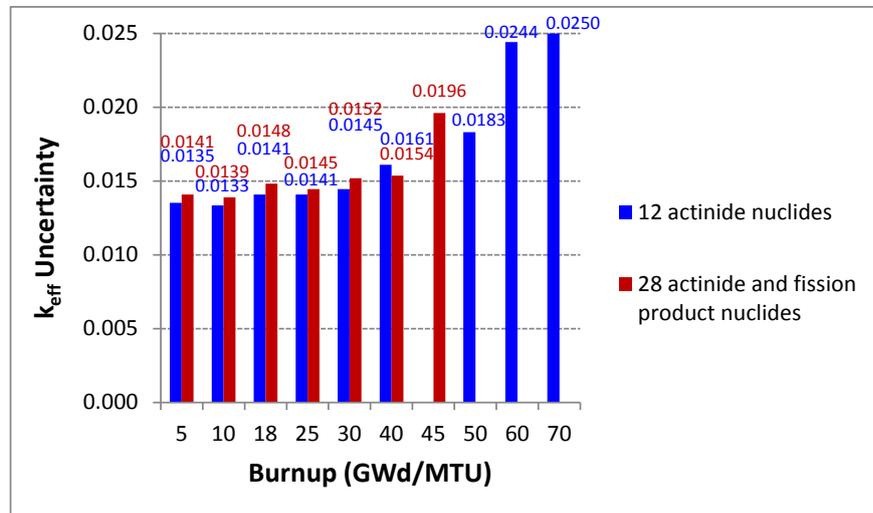


(b)

Figure 2. k_{eff} bias as a function of assembly average burnup for (a) the PWR SFP storage rack model; (b) the PWR SNF cask model.



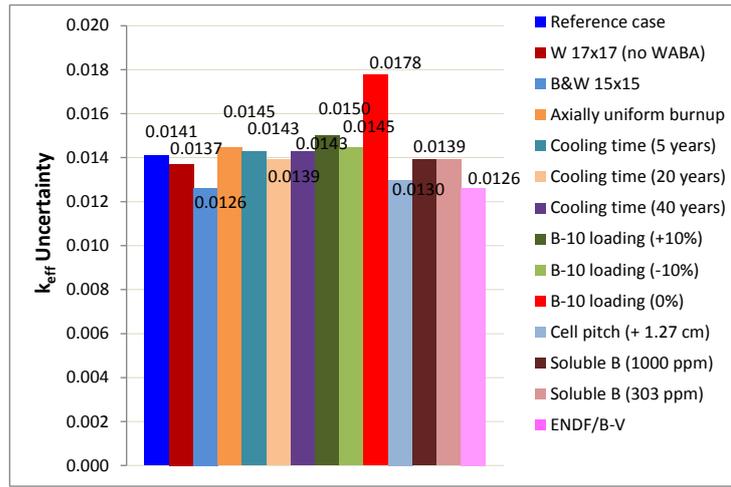
(a)



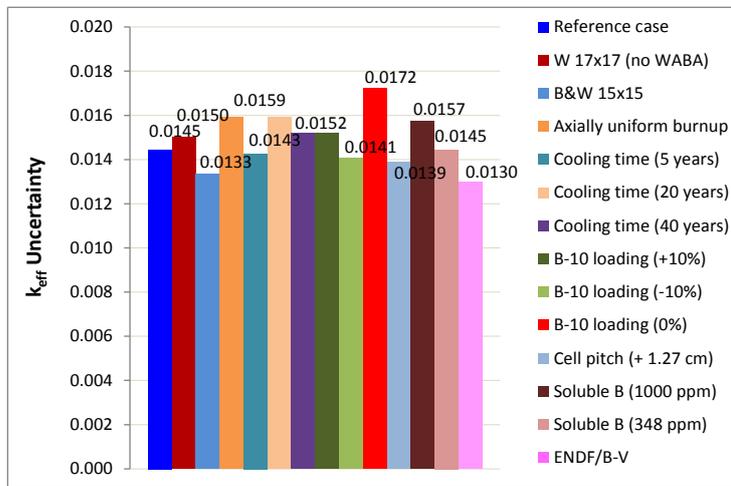
(b)

Figure 3. k_{eff} uncertainty as a function of assembly average burnup for (a) the PWR SFP storage rack model; (b) the PWR SNF cask model. Note: Uncertainty in k_{eff} at a 95 percent probability, 95 percent confidence level.

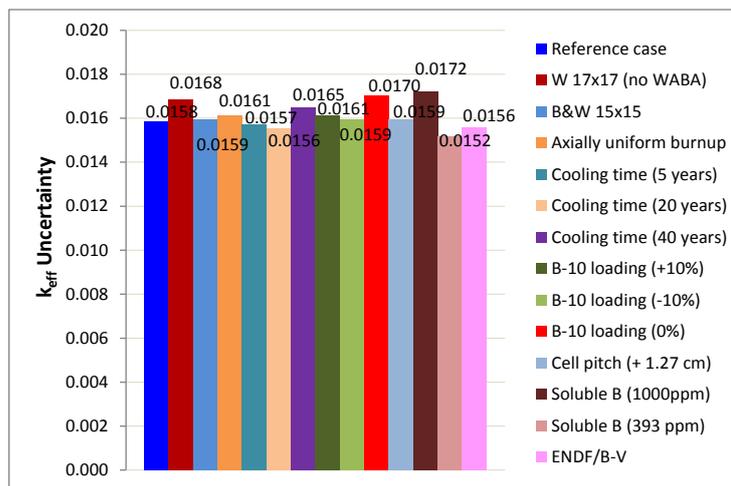
The values of uncertainty in k_{eff} for a range of parametric variations in the representative PWR SFP storage rack model (see Sect. 2.5) are shown in Figure 4, where the representative model is denoted as “reference case.” A bounding k_{eff} uncertainty value of 0.018 was determined for the burnup range 10 to 40 GWd/MTU. The largest variations from the reference case were obtained for the atypical $0 \text{ g/cm}^2 \text{ }^{10}\text{B}$ areal density (10 and 25 GWd/MTU burnup) and for the 1000 ppm soluble boron concentrations (40 GWd/MTU). All the evaluated cases using the Scale 6.1 238-group ENDF/B-VII library produced positive bias values, which typically are not credited in criticality safety analyses. The k_{eff} bias in the case of the Scale 6.1 44-group ENDF/B-V library exhibited negative values varying with assembly average burnup from -0.0001 (10 GWd/MTU) to -0.0040 (40 GWd/MTU). Therefore, there are significant differences between the k_{eff} bias values based on the ENDF/B-VII and on the ENDF/B-V nuclear cross-section data libraries.



(a)



(b)



(c)

Figure 4. Variation of uncertainty in k_{eff} with parameters important to criticality analyses for (a) 10, (b) 25, and (c) 40 GWd/MTU assembly average burnup in the SFP storage rack model. Note: Uncertainty in k_{eff} at a 95 percent probability, 95 percent confidence level.

The k_{eff} bias and k_{eff} uncertainty values for BWR SNF, 0.002 and 0.03, respectively, were based on assay data limited in its range of applicability. The measurement data from relatively old fuel assembly designs used in the isotopic validations is limited in its range of applicability since modern BWR assembly designs are significantly more complex. Measurement data for discharge fuel compositions are also limited in their applicability to nuclide compositions corresponding to assembly peak reactivity, particularly in the case of uranium and plutonium nuclides. No attempt has been made in this study to evaluate the applicability of the experimental assay data, obtained from fuel with typical discharge burnup, to the low burnup range corresponding to peak reactivity. The issue of applicability still remains even if better quality measured nuclide concentrations in discharge fuel compositions may become available. Therefore, the k_{eff} bias and k_{eff} uncertainty values for BWR SNF are provided to demonstrate the use of the methodology only.

4 CONCLUSIONS

This paper presents a technical approach for evaluating the uncertainties in burnup credit criticality calculations due to uncertainties in computed isotopic concentrations in spent fuel. The approach was based directly on the application of experimental data from destructive radiochemical assay of 100 PWR fuel samples and 32 BWR fuel samples to validate the depletion models. The validation approach was demonstrated for representative SNF storage pool and cask configurations/conditions using the Scale 6.1 code system and the Scale 6.1 238-group ENDF/B-VII library. The main characteristics of the isotopic validation technical approach are as follows: (1) bias and uncertainty associated with calculated nuclide compositions were based on comparisons to available measurements of nuclide concentrations obtained by destructive radiochemical assay; (2) 28 nuclides were considered in SNF compositions, including 12 actinide and 16 fission product nuclides, based on nuclide importance to fuel reactivity and on the availability of measurement data; (3) the bias and uncertainty in the calculated nuclide compositions were propagated to the k_{eff} values of the safety analysis models using the Monte Carlo uncertainty sampling method. Although the bias and bias uncertainty values presented in this report were determined using the Scale 6.1 code package and the ENDF/B-VII nuclear cross-section data, the methodology is not dependent on the code or the data.

Uncertainty in k_{eff} is demonstrated to be relatively insensitive to the depletion calculation methodology and nuclear data. The k_{eff} bias may depend on the depletion calculation methodology and on the code/nuclear data library, as shown by the different bias values obtained with the ENDF/B-V and ENDF/B-VII nuclear data. However, both k_{eff} bias and k_{eff} uncertainty may depend on the sets of the RCA data considered in the burnup credit isotopic validation because measurement biases and uncertainties are intrinsic components of the uncertainty methodology. Therefore, a relatively large set of measurement data acquired from different measurement programs is recommended for use in burnup credit isotopic validations to mitigate the impact of measurement uncertainties associated with any one laboratory and to provide a good representation of the spent fuel inventory.

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