

Quantitative Analysis of the Impact of Nuclide Deviation Zoning Strategy on Criticality Calculation Uncertainty under Burnup Credit

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Abstract

Burnup credit reduces the k_{eff} estimate in spent fuel criticality safety assessments by considering changes in nuclide composition during the burnup process; however, uncertainties exist in the sample bias of nuclide compositions, which affect the accuracy of criticality calculation results. To quantify this uncertainty at different burnup depths and improve the accuracy of burnup credit analysis, a nuclide bias partitioning strategy based on linear regression fitting and mean square error minimization was established for determining the optimal burnup interval partitioning of target nuclides. Using the AFA-3G 17 \times 17 fuel assembly as a benchmark problem, the propagation of nuclide bias uncertainty to criticality calculation results was evaluated on the SCALE 6.1 platform combined with ORIGEN and KENO modules through the Monte Carlo sampling method. The results show that this strategy can reduce criticality calculation uncertainty by approximately 40% under low burnup conditions, providing an optimized approach for spent fuel criticality safety evaluation under burnup credit.

Full Text

Quantitative Analysis of the Impact of Nuclide-Bias Partitioning Strategies on Criticality Calculation Uncertainty under Burnup Credit

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Abstract

Burnup credit reduces the estimated effective multiplication factor (k_{eff}) in criticality safety assessments of spent nuclear fuel by accounting for changes in nuclide composition during irradiation. However, uncertainties arising from sample-to-calculation deviations in nuclide inventories degrade the accuracy of criticality calculations. To quantify these uncertainties across different burnup ranges and improve the precision of burnup-credit analyses, this work develops a nuclide-bias partitioning strategy based on linear regression fitting and mean-squared-error minimization to determine optimal burnup interval boundaries for target nuclides. Using an AFA-3G 17 \times 17 PWR fuel assembly as a benchmark, depletion and criticality calculations were carried out on the SCALE 6.1 platform with ORIGEN and KENO modules, and Monte Carlo sampling was employed to propagate nuclide-bias uncertainty into k_{eff} . Results indicate that the proposed partitioning strategy reduces criticality uncertainty by approximately 40% under low-burnup conditions, offering an optimized approach for burnup-credit evaluations in spent-fuel criticality safety assessments.

Keywords: burnup credit; burnup calculation; Monte Carlo method; criticality safety analysis

1. Introduction

With the development of the nuclear power industry, the amount of nuclear fuel used in Chinese nuclear power plants increases annually, with over 14,000 tons of spent fuel expected to be generated by 2025, posing new challenges for spent fuel reprocessing [1]. Compared with traditional criticality safety analysis methods, burnup credit (BUC) reduces the estimated effective multiplication factor (k_{eff}) in criticality safety assessments by accounting for the burnup effects of spent fuel during reactor operation, thereby decreasing excessive safety margins and enhancing the economic efficiency and safety of spent fuel storage and transportation [2]. Burnup credit can significantly reduce the required space for spent fuel storage and transportation and lower operational costs, offering broad application prospects both domestically and internationally.

However, burnup-credit analyses involve depletion and criticality calculation programs that introduce uncertainty factors affecting the results [3]. Depletion calculation programs simulate the evolution of nuclide inventories in spent fuel using nuclear databases and numerical models that differ from actual conditions, introducing additional uncertainties. Meanwhile, criticality calculation programs introduce uncertainties into k_{eff} results through approximations in geometry simplification, mesh coarsening, and cross-section processing when solving geometric models. Furthermore, applicability assessments of depletion calculation codes reveal that calculation accuracy varies across different burnup depths, with limited accuracy in burnup models and cross-section libraries leading to varying degrees of deviation between calculated nuclide concentrations and experimental measurements [4]. In this regard, significant progress has

been achieved in related research both domestically and internationally. The U.S. Nuclear Regulatory Commission (U.S.NRC) [5] has compiled sample values of nuclide bias across different burnup depths based on experimental data and simulation calculations, providing important references for correcting depletion calculations. Oak Ridge National Laboratory (ORNL) [6] conducted burnup-credit analyses for spent fuel storage facilities at different burnup depths, quantifying the impact of burnup depth on criticality calculation results and their uncertainties. The China Institute of Atomic Energy [7] performed theoretical analysis of ^{125}Sb inventory based on experimental chemical analysis data from the TAKAHAMA-3 PWR burnup benchmark, obtaining the variation trends of calculated-to-experimental values with burnup depth and correcting the experimental values. Previous studies have employed partitioning methods to process sample data across burnup depths to evaluate nuclide bias levels at different burnup stages. However, the sample data used for burnup partitioning are not unified across studies, and specific interval division methods still lack scientific justification.

Building upon previous burnup-credit analysis methods, this paper further evaluates the impact of burnup interval boundary selection on criticality calculation results and their uncertainties, proposing a burnup-partitioning screening strategy based on linear regression analysis. The burnup-credit application level in this study is APU-1. The methodology first establishes piecewise functions based on the mean nuclide bias within each sub-interval to accurately assess the relationship between nuclide bias and uncertainty as a function of sample burnup. Subsequently, by evaluating the goodness-of-fit between the six actinide nuclide bias sample points of interest at the APU-1 application level and the piecewise functions, the optimal burnup partitioning strategy is identified. The study establishes a fuel assembly analysis model based on depletion and criticality calculation programs and employs Monte Carlo nuclide-concentration sampling to quantify the uncertainty propagated from depletion calculations to criticality calculations. Through burnup-credit analysis of benchmark problems under different burnup partitioning strategies, this paper provides theoretical support for burnup partitioning of nuclide bias.

2.1 Nuclide Bias in Burnup Calculation

Burnup credit is applied in criticality safety analysis of spent fuel by calculating the effective multiplication factor based on actual nuclide compositions, thereby reducing excessive conservatism inherent in traditional methods that ignore burnup effects [8]. To ensure safety while maximizing the economic benefits of spent fuel storage and transportation systems using burnup credit, it is necessary to evaluate various uncertainties affecting criticality calculation results under burnup-credit analysis. Criticality calculation uncertainties primarily originate from the following aspects [9]: (1) statistical or convergence uncertainties in criticality calculations; (2) uncertainties due to material composition and manufacturing tolerances; (3) uncertainties from geometric or material ap-

proximations in computational models; (4) biases and uncertainties introduced into criticality results due to depletion calculation uncertainties; (5) uncertainties in recorded burnup values; and (6) uncertainties associated with specific calculation methods and nuclear cross-section databases.

In depletion calculations, burnup-credit analysis focuses on nuclides with significant impact on criticality (primarily uranium, plutonium, other actinides, and selected fission products). Different nuclear databases or cross-section libraries introduce varying degrees of uncertainty in depletion and criticality calculations. Commonly used depletion codes are affected by model approximations and cross-section library accuracy when simulating fuel burnup history and nuclide evolution, leading to biases that require correction through nuclide bias. Nuclide bias is typically defined as the ratio of measured to calculated nuclide concentrations (M/C) [10], denoted as:

$$\text{Bias}_{i,n} = \frac{M_{i,n}}{C_{i,n}}$$

where i represents a specific nuclide and n denotes a particular fuel sample. For a given nuclide, a set of samples $\{\text{Bias}_{i,n}\}$ represents the bias distribution under different burnup conditions or across different measurement samples. The sample mean and standard deviation of nuclide bias are defined as:

$$\bar{X}_n = \frac{1}{N} \sum_{i=1}^N \text{Bias}_{i,n}$$
$$S_n = \sqrt{\frac{1}{N-1} \sum_{i=1}^N (\text{Bias}_{i,n} - \bar{X}_n)^2}$$

where N is the number of evaluated fuel samples, \bar{X}_n represents the average M/C ratio for the nuclide within the specific sample set, and the uncertainty S_n measures the sample dispersion for subsequent uncertainty analysis.

Due to the limited number of samples available for each nuclide, tolerance intervals are employed to estimate the confidence range of nuclide bias [11]. If the sample size is N , the data can be assumed to follow a normal distribution when the sample size is sufficiently large, and a two-sided tolerance limit factor $f_{b,g}/\sqrt{N}$ must be introduced to ensure a confidence level of $1 - g$ and coverage b . The tolerance interval for the mean nuclide bias is defined as:

$$\bar{X}_n \pm f_{b,g} \cdot \frac{S_n}{\sqrt{N}}$$

where the tolerance coefficient $f_{b,g}$ is calculated based on small-sample statistics with required coverage b and confidence level g . Accounting for the tolerance factor, the corrected standard deviation is:

$$s_n = f_{b,g} \cdot S_n$$

where s_n is termed the nuclide bias uncertainty and serves as an input parameter for nuclide concentration sampling in criticality calculations.

2.2 Relationship between Nuclide Bias and Burnup Depth

Establishing nuclide bias sample sets requires support from existing spent fuel composition measurement data and depletion program calculations [12]. The resulting nuclide bias samples are widely distributed across different burnup depths, while the bias and its uncertainty are closely related to the burnup depth of the samples. Greater burnup depth corresponds to longer neutron irradiation time in the reactor, leading to more complex nuclear reactions including transmutation, fission, and absorption. Depletion calculation programs may neglect minor burnup chains, thereby affecting the bias for different nuclide compositions [13].

[Figure 1: see original paper] shows the linear regression statistics for ^{235}U composition calculation bias. Under high-burnup conditions, the calculated ^{235}U values are generally higher than experimental measurements, consistent with conservative assumptions in criticality safety analysis. In the low-burnup stage, both the ^{235}U composition calculation bias and its uncertainty are relatively small, whereas they increase significantly in the high-burnup stage. Therefore, samples at different burnup depths should not be evaluated using uniform nuclide composition calculation bias and uncertainty. Currently, the number of samples in domestic and international experimental benchmark databases is insufficient to support credible statistical analysis of nuclide composition calculation bias and uncertainty at each individual burnup point. To address the issue of varying nuclide composition calculation bias and uncertainty across different burnup depths, this study introduces a partitioning approach to more accurately perform BUC analysis for samples at different burnup stages. Considering the variation patterns of nuclide bias and uncertainty during burnup evolution and the limited statistical sample size, this research adopts a three-interval partitioning approach (low, medium, and high burnup) based on NRC reports [5] to balance physical representativeness and statistical reliability. This partitioning method effectively reflects the statistical characteristics of nuclide bias across different burnup ranges while ensuring sufficient sample numbers in each interval. The partitioning method fits the relationship between nuclide bias and burnup depth as piecewise constant functions within different intervals:

$$\text{Bias}(BU) = \begin{cases} \bar{X}_1 & \text{if } BU \leq x_1 \\ \bar{X}_2 & \text{if } x_1 < BU \leq x_2 \\ \bar{X}_3 & \text{if } BU > x_2 \end{cases}$$

where x_1 and x_2 are burnup depth boundary values that divide the burnup range into low, medium, and high stages. N is the total number of nuclide bias samples, n_1 is the sample index with the highest burnup depth in the low-burnup interval, and n_2 is the sample index with the highest burnup depth in the medium-burnup interval.

After partitioning, the mean and standard deviation of nuclide composition bias are statistically analyzed for each nuclide within each burnup interval based on Equation (6), yielding the distribution characteristics of each nuclide bias in different burnup ranges. The burnup depth partitioning method determines the appropriate burnup interval for different nuclides within a sample based on its burnup depth and selects the corresponding mean and standard deviation parameters of nuclide bias within that interval. These statistical parameters are then used together with nuclide concentration values calculated by depletion programs as inputs for Monte Carlo random sampling to generate nuclide concentration samples for criticality calculations, thereby quantifying the propagation of sample nuclide composition uncertainty to criticality calculation uncertainty at different burnup depths in criticality safety analysis. However, traditional burnup interval partitioning often relies on industry conventions and applies uniform interval divisions to all nuclides of interest, lacking theoretical justification and statistical optimization tailored to the bias distribution characteristics of individual nuclides, making it difficult to accommodate inconsistent bias trends among different nuclides during burnup evolution.

2.3 Nuclide Bias Partitioning Strategy

To more accurately quantify the impact on criticality calculation uncertainty, it is necessary to partition burnup intervals separately for different nuclides according to their specific bias variation characteristics during burnup. Therefore, this study proposes a nuclide-specific partitioning strategy based on nuclide bias sample characteristics, which determines optimal boundary points (x_1 , x_2) by fitting the relationship between nuclide bias and burnup depth, enabling a complete workflow from partitioning to burnup-credit calculations. The optimal boundary points for each nuclide's burnup partitioning are determined by the mean squared error (MSE) between the nuclide bias sample points and the fitted piecewise function at the boundary points.

This study assumes that the M/C bias samples for each nuclide follow a piecewise constant distribution as shown in Equation (6) within burnup depth intervals. The optimal boundary points are determined by minimizing the MSE between sample biases and interval means within each region.

The MSE calculation is performed as shown in Equation (7). Based on burnup depth, samples are first partitioned into different regions. The mean value \bar{X}_k within each interval is calculated, followed by computing the squared difference between each sample and its interval mean. These squared differences are accumulated and then divided by the total number of samples N to obtain the mean squared error for the entire sample set under three-interval partitioning. An automated program was developed to independently optimize the partitioning strategy for different nuclides based on existing nuclide bias sample data. The program discretizes the burnup depth range with a step size of 1 GWd/tU and, while ensuring sufficient sample numbers in each interval, traverses all possible boundary point combinations to calculate the corresponding MSE values, selecting the combination with the minimum MSE as the optimal boundary. After obtaining the nuclide bias and uncertainty under the optimal boundary points, these parameters serve as inputs for Monte Carlo sampling to determine nuclide concentrations in burnup-credit analysis, followed by criticality calculations on the generated nuclide composition samples. The specific workflow of the automated program is shown in Figure 2 [Figure 2: see original paper].

3.1 Benchmark Problem and Model Selection

Based on the methodology described above, this study aims to demonstrate the optimized burnup-credit criticality calculation results using the nuclide bias partitioning strategy and analyze its effectiveness in reducing uncertainty and improving calculation accuracy. The AFA-3G 17 \times 17 fuel assembly [14] was selected as the benchmark problem for burnup-credit analysis, with spent fuel storage racks in the criticality calculation model used to store this assembly type. The specific parameters of the fuel assembly and fuel rods are listed in Table 1 .

Depletion and criticality calculations were performed using the SCALE 6.1 platform [15] with two main program modules: (1) Depletion calculation: The ORIGEN module was used to simulate fuel burnup history in the reactor and calculate the evolution of nuclide concentrations at various burnup depths. (2) Criticality calculation: The KENO module based on the Monte Carlo method was employed to evaluate the criticality of fuel assemblies and analyze the uncertainty introduced by nuclide concentration bias in the calculations. The fuel rod layout for the AFA-3G benchmark problem is shown in Figure 3 [Figure 3: see original paper].

To account for the axial power distribution effects in the AFA-3G fuel assembly, this study constructed a three-dimensional criticality model using the STARBUCS burnup-credit analysis module in SCALE. Referencing the axial burnup envelope distribution determined by Parish [16] from the PWR spent fuel distribution curve database, this envelope distribution divides the active region into 18 axial segments. Table 2 provides the corresponding envelope distributions for different burnup ranges.

The nuclide bias sample set used in this study includes experimental data from the international spent fuel composition database SFCOMPO2.0 [17] and calculation results from the domestic nuclide composition calculation program Bamboo-SFuel. Both Bamboo-SFuel and ORIGEN use the same ENDF/B-VII.0 nuclear data cross-section library [18] in depletion calculations. To verify the consistency between the two programs' results, benchmark comparisons were performed using the ORIGEN module, showing that the maximum relative deviation of nuclide concentration calculations for six major actinide nuclides remained within 3%. Therefore, nuclide composition bias quantified based on Bamboo-SFuel calculation results can be applied to burnup-credit analysis using SCALE 6.1 to correct its calculated values. The 122 PWR spent fuel assembly calculation benchmarks selected as the nuclide bias sample set are listed in Table 3, covering initial enrichment ranges from 2.453 wt% to 4.66 wt% and burnup depths from 6.9 GWd/tU to 75 GWd/tU, satisfying the initial enrichment and burnup depth requirements for most spent fuel assemblies at discharge.

This study employs the Monte Carlo sampling method [19] to obtain nuclide composition inputs for criticality calculations and quantify the propagation of nuclide bias uncertainty to criticality results. The specific procedure is: (1) Determine the mean and corrected standard deviation for each target nuclide based on its nuclide bias sample data at different burnup depths; (2) Assume the target nuclide bias follows a normal distribution based on the above statistical parameters and perform Monte Carlo random sampling to generate a large number of nuclide concentration samples; (3) Use the sampled nuclide concentration distributions as input parameters for Monte Carlo-based criticality calculations, then statistically analyze the keff distribution characteristics to evaluate criticality calculation uncertainty under burnup credit.

3.2 Uncertainty Quantification Results

At the APU-1 burnup-credit application level, this study considers the net reduction of fissile isotopes and neutron absorption by actinide nuclides in criticality safety analysis results [22]. To verify the impact of the proposed partitioning strategy screening method on burnup-credit analysis, the automated program was used to calculate MSE values for partitioning strategies of each nuclide of interest. Due to the limited number of nuclide bias samples used in this study, and referencing previous research methods for statistically processing nuclide bias across different burnup depth ranges [5], boundary point x_1 was traversed within the range of 12-28 GWd/tU and boundary point x_2 within 30-50 GWd/tU to ensure that each burnup interval after partitioning contained sufficient nuclide bias samples (10 and benchmark problem types (3) and that each interval was sufficiently wide (10 GWd/tU). Non-compliant boundary point combinations were excluded to avoid introducing additional errors from improper interval selection. The nuclide partitioning strategy screening results are shown in Figure 4 [Figure 4: see original paper].

After screening boundary points for different nuclides, the partitioning strategy with the minimum MSE—representing the best fit to nuclide bias samples—was obtained. Table 4 presents the relevant parameters of nuclide sample bias after optimization by the partitioning strategy.

Compared with direct analysis of nuclide bias across the entire burnup depth range without partitioning, the burnup partitioning strategy fully accounts for differences in interval division among different nuclide bias samples and the relationship between nuclide bias and its uncertainty as functions of burnup depth. Taking nuclides ^{235}U , ^{239}Pu , and ^{241}Pu as examples, their bias and uncertainty values with and without burnup partitioning are shown in Figures 5 [Figure 5: see original paper]–10 [Figure 10: see original paper].

Based on the mean nuclide bias and uncertainty values for each nuclide across the three burnup intervals, this study used the Monte Carlo sampling method to obtain nuclide concentration sampling results at different burnup points from 10 GWd/tU to 70 GWd/tU for the benchmark problem. During sampling, the optimal burnup partition boundaries for each nuclide were first used to determine its burnup partition (low, medium, or high burnup stage) at each burnup point. After processing with the burnup partitioning strategy, the partition information for the benchmark problem at different burnup points was obtained, as shown in Table 5. Subsequently, the mean and standard deviation of nuclide bias in the corresponding interval were used as random sampling parameters to generate corrected nuclide concentration samples via Monte Carlo sampling, as expressed in Equation (8):

$$C'_{n,b} = C_{n,b} \times (X_{n,b} + s_{n,b} \cdot U_n)$$

where $C'_{n,b}$ is the corrected nuclide concentration obtained through Monte Carlo sampling; $C_{n,b}$ is the calculated value of the n -th nuclide at burnup point b defined by the Monte Carlo sampling; U_n is a random value sampled from the standard normal distribution; and $X_{n,b}$ and $s_{n,b}$ are the bias and uncertainty of the n -th nuclide composition at burnup point b , respectively, both determined by the statistical parameters of the burnup interval to which the nuclide belongs after partitioning at burnup point b .

Each nuclide was sampled 500 times, and the results were used as input parameters in the KENO module input file to obtain the sample mean k_{eff} and sample standard deviation $s_{k_{\text{eff}}}$ of the criticality calculation results. Following the double 95% principle, a one-sided tolerance factor for 500 samples with 95% coverage and 95% confidence level was selected. The impact of nuclide composition uncertainty on criticality calculations is presented in Table 6.

According to the criticality calculation results, except for a difference of nearly 800 pcm at the 30 GWd/tU burnup depth, the k_{eff} calculation results before and after optimization are essentially consistent at other burnup points. Based on the partitioning strategy processing results in Table 5, it can be observed

that at the 30 GWd/tU burnup depth, the ^{235}U nuclide, which contributes most significantly to criticality reactivity and uncertainty, uses statistical parameters from the medium-burnup interval, where the ^{235}U nuclide bias is the maximum among the three partitions and larger than the value used in the non-partitioned treatment, introducing additional reactivity and causing the keff result to increase compared with the non-partitioned case. At 40 GWd/tU and 50 GWd/tU burnup depths, ^{235}U also falls in this interval, but since the fissile nuclide ^{239}Pu is in the interval with minimum bias, the keff increase is less significant than at 30 GWd/tU. At low burnup depths, the uncertainties before partitioning strategy application are 2844 pcm and 2614 pcm, respectively, which decrease to 1561 pcm and 1496 pcm after application, demonstrating a significant reduction of approximately 40–45%. This indicates that the partitioning strategy can more accurately reflect nuclide bias and its uncertainty at low burnup, enabling more economical criticality safety assessments. At higher burnup depths, the criticality calculation uncertainty fluctuates after partitioning but remains within $\pm 10\%$. Across the full burnup depth range, the optimized criticality calculation uncertainty shows an increasing trend with burnup depth, which aligns better with theoretical expectations and actual conditions [23].

This study proposes a nuclide bias partitioning strategy based on mean squared error minimization that divides burnup depth into low, medium, and high intervals according to differences in nuclide sample bias and uncertainty across burnup stages. By independently optimizing partition boundaries for each nuclide through an automated program and propagating the obtained nuclide concentration uncertainties to criticality calculations using Monte Carlo sampling, the study successfully achieves refined processing of nuclide composition correction under burnup credit. The burnup partitioning strategy ensures that the bias mean and uncertainty parameters used for correction at different burnup depths better reflect the statistical characteristics of the respective burnup intervals, avoiding inappropriate influences from samples across different intervals on calculation results at specific burnup points. This method primarily addresses the problem of uncertainty overestimation at low burnup stages in traditional burnup-credit analysis caused by mixing high-burnup-depth bias samples, thereby improving safety margins for low-burnup sample analysis under burnup credit. For higher burnup stages, the partitioning strategy excludes the influence of low-burnup nuclide bias samples, resulting in slight increases in criticality calculation uncertainty for some samples. However, compared with non-partitioned treatment, the additional uncertainty introduced is limited, and the keff and uncertainty results obtained through partitioning maintain the correct statistical trend with burnup depth variation. The partitioning method proposed in this paper can further compress criticality margins at low burnup depths while ensuring criticality safety, thereby improving the economic efficiency of spent fuel storage and transportation systems.

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