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Validating Actinides and Fission Products for Burnup Credit Criticality Safety Analyses – Nuclide Compositions Prediction with Extended Validation Basis

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ABSTRACT

Criticality safety evaluations accounting for burnup credit require validation of the computational tools and associated nuclear data used to perform the fuel depletion and criticality calculations. The NUREG/CR-7108 report published in 2012 documents an approach for establishing the bias and bias uncertainty in the calculated neutron multiplication factor (k_{eff}) that is associated to calculated spent nuclear fuel nuclide compositions, using as validation basis radiochemical assay (RCA) measurement data for nuclide concentrations.

Studies documented in the current report build upon the NUREG/CR-7108 approach. The bias and bias uncertainty in calculated k_{eff} for a representative analysis model that includes pressurized water reactor (PWR) spent fuel is determined by applying a stochastic sampling method. The validation basis presented herein is improved compared to that in NUREG/CR-7108 to fill data gaps at high burnups, with practical impact on burnup credit for modern discharged fuel. This extended validation basis includes recent RCA measurements performed at Oak Ridge National Laboratory under an experimental program jointly funded by the Nuclear Regulatory Commission and the Department of Energy. The improvement in the validation basis led to an overall decrease in the k_{eff} bias uncertainty for the considered analysis model compared to the corresponding values in ISG-8 Rev.3.

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EXECUTIVE SUMMARY

Criticality safety evaluations accounting for burnup credit require validation of the computational tools and associated nuclear data that are used to perform the fuel depletion and criticality calculations involved in these evaluations. An approach for establishing the bias and bias uncertainty in the calculated neutron multiplication factor (k_{eff}) for a criticality safety model, resulting from bias and bias uncertainty in calculated nuclide concentrations in spent nuclear fuel, was documented by Oak Ridge National Laboratory (ORNL) in the NUREG/CR-7108 report published in 2012. The approach was independent of the depletion and criticality computational methods being used and of the choice of the safety analysis model. The validation basis for the approach consisted of measured nuclide concentrations in spent nuclear fuel samples that were obtained from radiochemical assay (RCA) experiments.

The approach documented in the current report builds upon the approach documented in NUREG/CR-7108. The bias and bias uncertainty in calculated k_{eff} is determined for a representative safety analysis model of a generic burnup credit (GBC)-32 cask that includes pressurized water reactor (PWR) spent fuel assemblies. The approach applied herein is based on a new and standardized implementation of the stochastic sampling method presented in NUREG/CR-7108. This implementation employs the capabilities in the Sampler uncertainty quantification tool, which was introduced in the SCALE modeling and simulation suite for nuclear safety analysis and design in 2016.

The approach for propagating bias and bias uncertainty in calculated nuclide concentrations to bias and bias uncertainty in calculated k_{eff} for a criticality safety model is based on these primary steps: (1) develop a model for the bias and bias uncertainty in calculated nuclide concentrations for a given set of nuclides over a burnup range of interest based on measurement data for these nuclides, (2) randomly vary the nuclide concentrations within the range of their estimated uncertainties for the fuel compositions of the criticality model, (3) calculate k_{eff} for the criticality safety model using the randomly varied fuel compositions and repeat the process multiple times, and (4) estimate the bias and bias uncertainty in calculated k_{eff} for the criticality safety model from the distribution of k_{eff} values.

All analyses presented in this report are performed using the computational tools available in the 6.2.4 release of SCALE and nuclear cross sections based on Evaluated Nuclear Data File (ENDF)/B-VII.1. Fuel compositions for the criticality safety model include 28 nuclides important for burnup credit: 12 actinides and 16 fission products.

The loading curve presented in NUREG/CR-7108 for the GBC-32 cask model was reassessed to verify its applicability when using updated computational tools in SCALE 6.2.4 and 252-group ENDF/B-VII.1 cross section libraries. Based on the observed results, a new loading curve was generated and served for updating the cask reference model before using it to determine the bias and bias uncertainty in k_{eff} associated to calculated nuclide concentrations. The updated loading curve corresponds to fuel compositions that include all 28 burnup credit nuclides and was generated based on a new methodology.

The RCA validation basis is significantly extended in the current report for high burnups. It includes measurement data for 129 PWR fuel samples that cover a burnup range of 7–80 GWd/MTU, with 30 of these samples having burnups greater than 40 GWd/MTU. The validation basis considered in NUREG-7108 included 100 PWR fuel samples with burnups between 7 and 60 GWd/MTU. The current validation basis includes high-burnup RCA

measurements from international experimental programs and from recent experiments performed at ORNL for nine PWR spent fuel samples. These recent measurements were obtained as part of an ongoing experimental program jointly funded by the US Nuclear Regulatory Commission (NRC) and the US Department of Energy (DOE). Bias and bias uncertainty values in calculated k_{eff} for the considered criticality safety model are presented in this report for assembly-average burnups ranging between 5 and 70 GWd/MTU, with both actinide and fission product nuclides being credited. The k_{eff} bias varies between 0.0010 and 0.0033, and the k_{eff} bias uncertainty varies between 0.0136 and 0.0187. The k_{eff} bias uncertainty values for burnups greater than 30 GWd/MTU and fuel compositions that include actinide and fission product burnup credit nuclide are smaller than the corresponding values shown in NUREG/CR-7108. Part of this decrease is caused by a reduction in the bias uncertainty for the calculated nuclide concentrations resulting from improved statistics (increased number of measurements for burnup credit nuclides) and the addition of high-quality measurement data for impactful actinides and fission products.

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ABBREVIATIONS AND ACRONYMS

ANL	Argonne National Laboratory
ATM	approved testing material
CEA	Commissariat à l'Énergie Atomique
CSN	Consego de Securidad Nuclear
DOE	US Department of Energy
ENDF	Evaluated Nuclear Data File
ENRESA	Empresa Nacional de Residuo Radioactivo, SA (Spain)
ENUSA	Empresa Nacional del Uranio, SA (Spain)
GBC	generic burnup credit cask
GE-VNC	General Electric Vallecitos Nuclear Center
GKN II	Gemeinschaftskernkraftwerk Unit II in Neckarwestheim/Neckar
GWd/MTU	gigawatt-day per metric ton of uranium (unit of fuel burnup)
IAEA	International Atomic Energy Agency
IRCh	Institute for Radiochemistry at Karlsruhe
ISG	interim staff guidance
ITU	European Institute for Transuranium Elements
JAERI	Japan Atomic Energy Research Institute (now Japan Atomic Energy Agency)
JRC	Joint Research Center, European Commission
k_{eff}	effective neutron multiplication factor
KRI	Khlopin Radium Institute
LWR	light-water reactor
M/C	measured-to-calculated
NEA	Nuclear Energy Agency
NRC	US Nuclear Regulatory Commission
OFA	optimized fuel assembly
ORNL	Oak Ridge National Laboratory
PNNL	Pacific Northwest National Laboratory
PSI	Paul Scherrer Institute
PWR	pressurized water reactor
RCA	radiochemical assay
SCK·CEN	Studiecentrum voor Kernenergie – Centre d'étude de l'Énergie Nucléaire
$T_{1/2}$	half-life
TMI	Three Mile Island
W	Westinghouse
WABA	wet annular burnable absorber
WAK	Karlsruhe Reprocessing Plant
YMP	Yucca Mountain Project

1 INTRODUCTION

Technical challenges associated with the use of burnup credit for the transport and storage of spent nuclear fuel have been the focus of numerous investigations performed in the United States and other countries for decades. Advances in developing new approaches to address these challenges or to improve the existing ones have been possible as a result of advances in the supporting modeling and simulation tools and the availability of new relevant experimental data. Criticality safety evaluations accounting for burnup credit require validation of the computational methods, computational tools and associated nuclear data used to perform fuel depletion and criticality calculations needed for the evaluations. Current US Nuclear Regulatory Commission (NRC) guidance for calculation of burnup credit in criticality safety analyses of pressurized water reactor (PWR) spent fuel for transportation and storage casks is based on the Interim Staff Guidance (ISG)-8, Rev 3 [1] that was released in 2012. The guidance in ISG-8 Rev. 3 has since been incorporated into the "Standard Review Plan for Spent Fuel Dry Storage Systems and Facilities" (NUREG-2215) [2] and "Standard Review Plan for Transportation Packages for Spent Fuel and Radioactive Material" (NUREG-2216) [3] reports, which were released in 2020. The technical basis supporting this burnup credit guidance includes an approach developed by Oak Ridge National Laboratory (ORNL) for establishing the bias and bias uncertainty in calculated neutron multiplication factor (k_{eff}) for transportation and storage casks resulting from bias and bias uncertainty in calculated nuclide concentrations in spent nuclear fuel. This approach was documented in detail in NUREG/CR-7108 in 2012 [4] and other related publications [5].

Bias and bias uncertainty in calculated nuclide concentrations for 28 actinide and fission product nuclides important for burnup credit [1] in PWR spent fuel were determined in NUREG/CR-7108 through validation based on radiochemical assay (RCA) data for 100 fuel samples with burnups between 7 and 60 gigawatt-day per metric ton of uranium (GWd/MTU) and initial fuel enrichments between 2.453 and 4.657%. The bias and bias uncertainty in calculated k_{eff} for a representative safety analysis model of a generic burnup credit (GBC)-32 cask [6] with PWR spent fuel assemblies, with fuel compositions including both actinide and fission product burnup credit nuclides, were reported in NUREG/CR-7108 [4] for assembly-average burnups ranging between 5 and 45 GWd/MTU. All underlying analyses were performed using computational tools in version 6.1 of SCALE [7] and nuclear cross sections based on Evaluated Nuclear Data File (ENDF)/B-VII.0 [8].

Studies documented in the current report build upon and complement the previous investigations [4]. A new and standardized implementation is provided for the stochastic sampling method presented in NUREG-7108 [4]. This implementation is based on capabilities in the Sampler [9] uncertainty quantification tool which was introduced in the SCALE code package in 2016. All analyses in this report were performed using computational tools in version 6.2.4 of SCALE and nuclear cross sections based on ENDF/B-VII.1 [10].

The RCA validation basis for PWR spent nuclear fuel is extended in the current report to higher burnups than previously [4] available and covers a 7–80 GWd/MTU range. This extended validation basis includes recent RCA measurements performed at ORNL for nine PWR spent fuel samples. These measurements were performed as part of an ongoing experimental program jointly funded by the NRC and the US Department of Energy [11,12]. In addition, nine high burnup spent fuel samples from fuel irradiated in the Gosgen PWR in Switzerland and the Vandellos PWR in Spain are included. Data for these nine samples were acquired through two international experimental programs: the MALIBU program [13] coordinated initially by

Belgonucleaire and later by Studiecentrum voor Kernenergie – Centre d'étude de l'Énergie Nucléaire (SCK-CEN) in Belgium; and the program [14,15] coordinated by the Spanish safety council for nuclear activities Consejo de Seguridad Nuclear (CSN) in collaboration with Spanish fuel vendor Empresa Nacional del Uranio, SA (ENUSA) and Empresa Nacional de Residuo Radioactivo SA (ENRESA), the organization responsible for waste management in Spain. ORNL participated to these international programs through funding from the NRC.

Extension of the validation basis from 60 GWd/MTU [4] to 80 GWd/MTU in the current report has practical impact on criticality safety with burnup credit for modern discharged fuel. The median of the assembly-average burnup characterizing all PWR spent fuel assemblies discharged in US between 2003 and 2013 (i.e., 35,256 assemblies), as documented in NUREG/CR-7227 [16], is 47.1 GWd/MTU. Whereas the maximum assembly-average burnup for these PWR fuel assemblies is 69.7 GWd/MTU [16], which would correspond to a maximum axial burnup of approximately 77 GWd/MTU.

The information presented in this report emphasizes new data and does not repeat details that can be found in NUREG/CR-7108. However, an overview of data and concepts previously discussed [4] is included herein as necessary to help the reader understand the context and relevance of the differences between previous and current findings.

An overview of the terminology used throughout this report and a brief description of the considered criticality safety analysis model are provided in Section 2, along with a description of the method used to determine the bias and bias uncertainty in the calculated k_{eff} of the safety analysis model that is associated to calculated nuclide concentrations for that model. Section 3 presents a new approach for determining loading curves for the considered cask criticality model. Section 4 provides a brief description of the computational tools and associated nuclear data used to perform the analyses presented in this report. Section 5 describes the extended RCA validation basis used to determine the bias and bias uncertainty in calculated nuclide concentrations, summarizes the computational approach used, and presents the results obtained. Section 6 presents the results obtained for the bias and bias uncertainty in calculated k_{eff} for the considered criticality model. Concluding remarks are provided in Section 7. Details concerning the Sampler-based implementation of the stochastic sampling method, verification, and results, are presented in Appendix A.

2 OVERVIEW OF MODELS AND METHODS

2.1 Terminology

Criticality safety analyses are required to demonstrate that a proposed spent nuclear fuel storage or transportation configuration meets applicable requirements. These analyses include development of applicable criticality safety models and calculations to show that the proposed configuration will meet the maximum k_{eff} limits specified in the applicable requirements and guidance [1-3].

The criteria for establishing subcriticality with credit for the reactivity decrease based on fuel burnup are expressed in Eq. (1) [4,17],

$$k_p + \Delta k_p + \beta_i + \Delta k_i + \beta + \Delta k_\beta + \Delta k_x + \Delta k_m \leq k_{limit}, \quad (1)$$

where

k_p is the calculated multiplication factor of the model for the evaluated system;

Δk_p is an allowance for
- statistical or convergence uncertainties, or both, in the determination of k_p,
- material and fabrication tolerances, and
- uncertainties caused by geometric or material representation limitations of the models used in the determination of k_p;

β_i is the bias in k_p resulting from depletion code bias in calculated nuclides concentrations;

Δk_i is the bias uncertainty in k_p resulting from depletion code bias uncertainty in the calculated nuclides concentrations;

β is the bias that results from using a particular calculation method and nuclear cross section data to calculate the benchmark criticality experiments;

Δk_β is the criticality bias uncertainty, which includes
- statistical or convergence uncertainties, or both, in the computation of β,
- uncertainties in the benchmark criticality experiments,
- uncertainty in the bias resulting from application of the linear least-squares fitting technique to the critical experiment results, and
- tolerance interval multiplier to yield a single-sided 95% probability and 95% confidence level;

Δk_x is a supplement to β and Δk_β that may be included to provide an allowance for the bias and uncertainty from nuclide cross section data that might not be adequately accounted for in the benchmark criticality experiments used for calculating β;

Δk_m is a margin for unknown uncertainties deemed to be adequate to ensure subcriticality of the physical system being modeled (this term is typically referred to as an *administrative margin*); and

k_{limit} is the upper limit on the k_{eff} value for which the system is considered.

Determination of the term $\beta_i + \Delta k_i$ is addressed in NUREG/CR-7108 [4] and is revisited in this report. The terminology applied [4] is also used throughout this report for consistency, as follows:

- **k_{eff} bias** represents the average change in the calculated k_{eff} of the considered safety model resulting from bias and bias uncertainty in calculated nuclide concentrations in fuel for that model;

- **k_{eff} bias uncertainty** represents the variance of the change in calculated k_{eff} of the considered safety model, at a 95% probability and 95% confidence level, which results from bias and bias uncertainty in calculated nuclide concentrations for that model.

A bias that reduces the calculated value of k_{eff} is not considered in Eq. (1), consistent with the approach in NUREG/CR-7108 [4] and NUREG/CR-7109 [17].

2.2 Criticality Safety Model

The representative safety analysis model for a PWR spent nuclear fuel cask that serves as basis for k_{eff} bias and bias uncertainty determination in the current report is the GBC-32 cask model [6], the same configuration used as a reference model in NUREG/CR-7108 [4], NUREG-7109 [17], and other burnup credit studies [18,19]. This cask configuration includes 32 PWR fuel assemblies and neutron absorber panels containing a fixed absorber uniformly dispersed in the panels' material. The 3D cask model represents half of the cask configuration, taking advantage of the symmetry. An illustration of this 3D model, as rendered with SCALE, is presented in Figure 2-1. This illustration shows (a) a 2D cross section at the axial midline of the model to emphasize the included fuel assemblies, and (b) a cutaway view of the 3D model with a quarter of the model removed to illustrate the different axial layers with different fuel compositions within the fuel assembly. Each color in Figure 2-1 indicates a different material composition in the model.

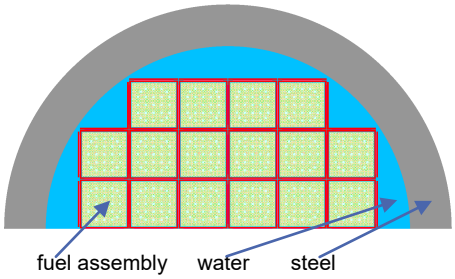
All PWR assemblies in the cask model are identical with respect to geometry and material compositions. The PWR assembly is a Westinghouse (W) 17×17 optimized fuel assembly (OFA) as used in previous reports [4,18]. A 2D cross section of the assembly model is presented in Figure 2-2 showing the assembly layout (a) as used in the assembly depletion simulations, with wet annular burnable absorber (WABA) rods present, in which the burnable absorber material is boron carbide contained in an alumina matrix, and (b) as used in the cask criticality model (no WABA present). Within the cask model, the fuel rods of this assembly are represented using 18 axial zones with the same axial length but different fuel material compositions, reflecting 18 different fuel burnups.

For a given assembly-average burnup, the burnup values of these axial zones in the fuel rods are determined by applying previously used axial burnup profiles [4,18] that were shown to be conservative [18] with respect to criticality. These axial burnup profiles are illustrated in Figure 2-3. These profiles were generated [18] based on data from PWR cycles of operation up to around 1995, for assemblies with much lower assembly-average burnups than those operated in modern PWRs. The axial profile shown in Figure 2-3 for burnups greater than 30 GWd/MTU was developed based on data for 1225 assemblies, out of which: 40% had burnups between 30 and 35 GWd/MTU, 71% had burnups between 30 and 40 GWd/MTU, and 90% of them had burnups between 30 and 45 GWd/MTU. However, it was shown that the use of these profiles for assemblies with assembly burnups higher than 45 GWd/MTU would be conservative

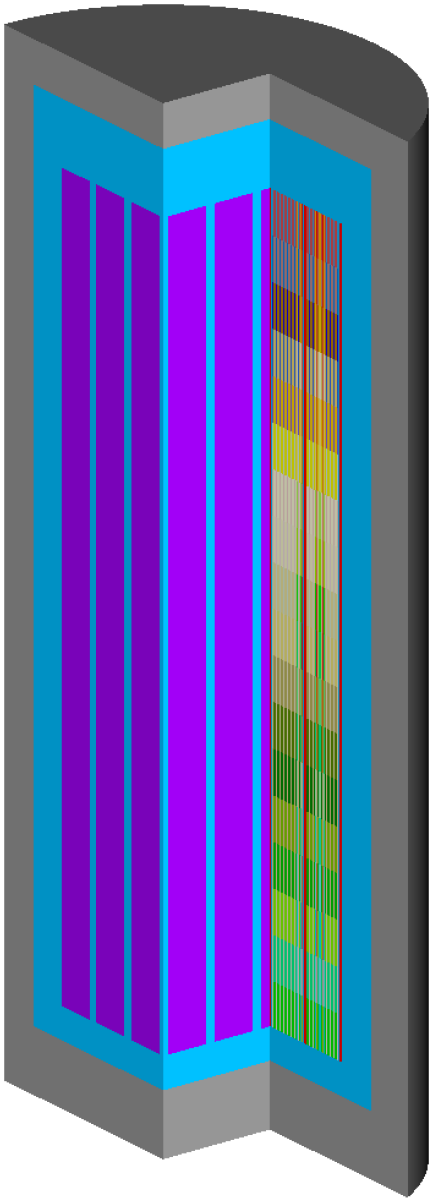
with respect to k_{eff} [18]. Therefore, applying the profile from Figure 2-3 to assemblies with average burnups greater than the burnups used as a basis for deriving this profile will be conservative.

For a given assembly-average burnup, burnup-dependent nuclide concentrations in the fuel rods of the PWR assembly were determined in NUREG/CR-7108 [4] such as the k_{eff} value for the cask model was 0.94, as previously recommended for cask criticality safety analyses [2,3,20]. For validation purposes, loading curves were developed [4] for the cask model. These loading curves were updated for the current report using a new methodology as described in Section 3.

Depletion calculations with conservative reactor operating parameters that increase discharge reactivity [4,21] for spent PWR fuel were performed to generate axially varying burnup-dependent nuclide concentrations for the spent fuel in the considered PWR assembly. The depletion calculations for achieving a given assembly-average burnup are followed by decay simulations for a cooling time of 5 years from the time that the assembly is discharged from the reactor. The resulting fuel compositions are applied in the GBC-32 cask model.

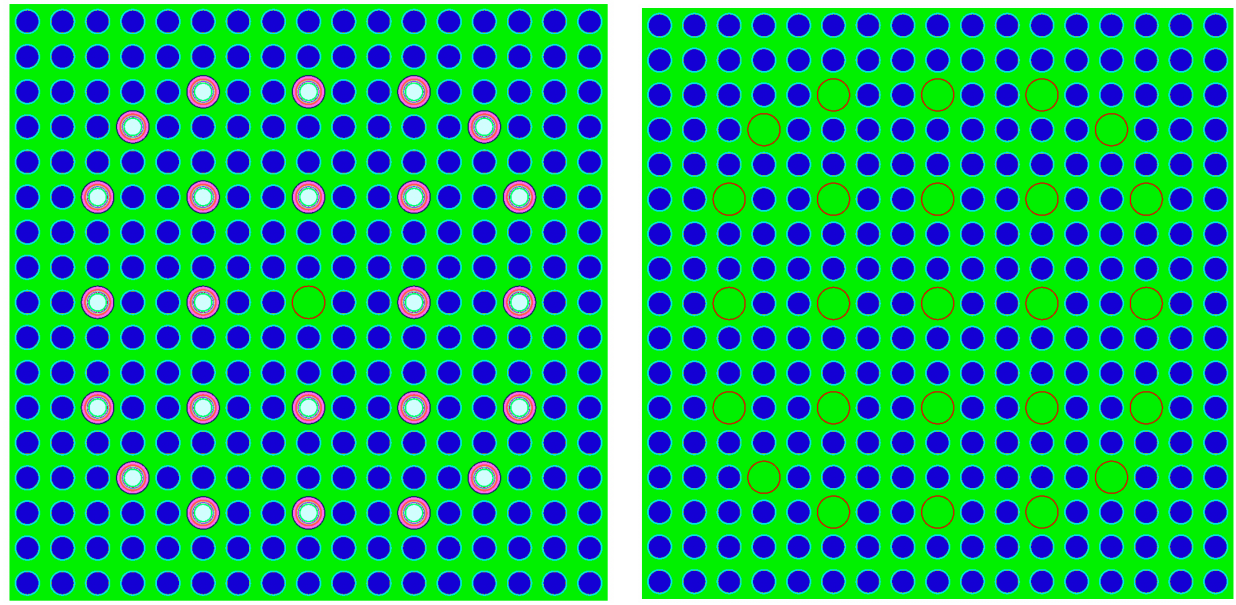


(a) 2D cross section at the axial midline



(b) Cutaway view of the 3D model with a quarter of the model removed (18 axial fuel materials)

Figure 2-1 Ilustration of the SCALE Model for the GBC-32 Cask



WABA rod; fuel rod; guide tube.

(a) As in assembly depletion model (with WABA rods)

(b) As in cask criticality model (no WABA present)

Figure 2-2 Illustration of the PWR Assembly Configuration (2D Cross Section)

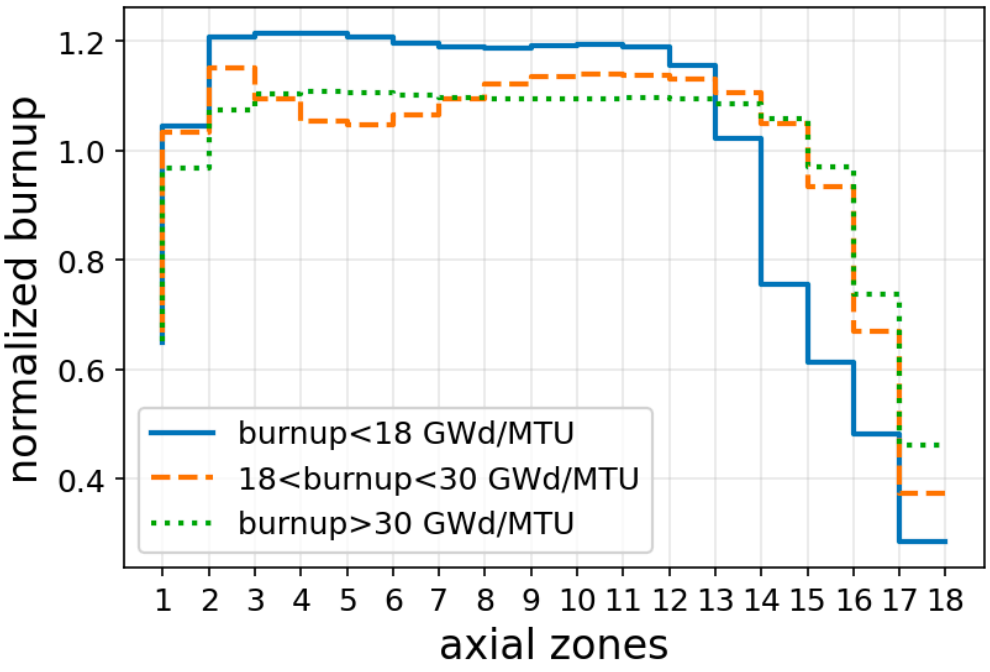


Figure 2-3 Illustration of Axial Burnup Profiles for the PWR Assembly

2.3 Method for Stochastic Uncertainty Sampling

Uncertainty in calculated nuclide concentrations in spent nuclear fuel is one of the greatest sources of uncertainty in criticality calculations that account for burnup credit. Different methods are used to propagate the uncertainty in calculated nuclide concentrations to the uncertainty in k_{eff} for a system of interest. Some of these methods are presented in detail in NUREG-6811 [19]. The stochastic uncertainty sampling method was deemed as enabling a realistic estimate of the effects of nuclides uncertainties by simulating the probability distributions of the nuclide concentrations [19,22]. The foundation of the stochastic uncertainty sampling method was earlier introduced in ORNL/TM-2001/83 [22]. This method was implemented in a computer code named KRONOS [22], which was designed to automate the setting up, execution, and parsing of the output for the large number of criticality calculations involved. Updated versions of this code were applied for the work presented in NUREG-6811 [19] and NUREG/CR-7108 [4]. Appendix A describes the new implementation of the method used in this effort, using the Sampler uncertainty quantification tool available in SCALE 6.2.4.

For completeness, the principles of the stochastic method for uncertainty sampling are briefly summarized herein. Details can be found in related reports [4,19,22]. The objective of propagating the bias and bias uncertainty in calculated nuclide concentrations to bias and bias uncertainty in the k_{eff} of a criticality model can be achieved by applying the following four steps:

A. Establish the expected bias and bias uncertainty in calculated nuclide concentrations by validating the depletion method used to calculate these concentrations based on measurement data for the burnup credit nuclides.

B. Randomly sample the calculated nuclide concentrations within the range of their estimated uncertainties and probability distributions based on data obtained in step A.

C. Calculate k_{eff} for the criticality model using the values of nuclide concentrations from B, and repeat steps B and C multiple times for multiple random realizations.

D. Analyze the distribution of k_{eff} values obtained from C; this distribution provides variations in k_{eff} that correspond to random variations in expected values for calculated nuclide concentrations. Determine bias and bias uncertainty in k_{eff}.

These steps are described further in the sections below.

2.3.1 Step A. Establish bias and bias uncertainty in calculated nuclide concentrations

Validation of the depletion method used to calculate nuclide concentrations for the criticality model can be performed by comparing measured nuclide concentrations to corresponding calculated values determined using the considered depletion method. In this context, *depletion method* refers to the computational tool and the associated nuclear data used to simulate the fuel depletion during irradiation. Given a set of N_i measured concentrations for a burnup credit nuclide i, an average measured-to-calculated (M/C) nuclide concentration ratio $\overline{X}_i$ is calculated as

$$\overline{X}_i = \frac{1}{N_i}\sum_{j=1}^{N_i} X_i^j = \frac{1}{N_i}\sum_{j=1}^{N_i} \frac{M_i^j}{C_i^j}, \quad (2)$$

where

i = index of burnup credit nuclide in the set of 28 burnup credit nuclides,

j = index of a measured concentration for nuclide i in the set of N_i measured concentrations for that nuclide,

M_i^j = measured concentration of nuclide i that corresponds to measurement (i.e., index) j in the set of N_i measured concentrations for that nuclide,

C_i^j = calculated concentration of nuclide i that corresponds to measurement (i.e., index) j in the set of N_i measured concentrations for that nuclide, and

X_i^j = M/C nuclide concentration ratio of nuclide i that corresponds to measurement (index) j in the set of N_i measured concentrations for that nuclide.

The uncertainty in the prediction of $\overline{X}_i$ is the standard deviation s_i associated with $\overline{X}_i$, calculated as

$$s_i = \sqrt{\frac{\sum_{j=1}^{N_i}\left(X_i^j - \overline{X}_i\right)^2}{N_i - 1}}. \quad (3)$$

$\overline{X}_i$ and s_i are unique to each nuclide and can vary as a function of burnup. The value $\overline{X}_i$ represents the factor that, when multiplied by a calculated nuclide concentration C_i for nuclide i, will correct for the average bias in the predicted nuclide concentration, resulting in an improved (best) estimate for that concentration as $C_i\overline{X}_i$.

2.3.2 Step B. Randomly sample calculated nuclide concentrations in the considered criticality model

The method assumes a normal distribution of the expected nuclide concentration corresponding to a calculated concentration C_i. A verification of the normality assumption was performed in NUREG-7108, which indicated that the use of a normal distribution in place of the actual distribution is slightly conservative (i.e., led to a greater value of k_{eff} bias uncertainty). For application in Step C, to randomly sample the nuclide concentrations within the range of their estimated uncertainties, s_i is adjusted using tolerance factors assigned to bound the limit of $\overline{X}_i$ at a given confidence level. This adjustment accounts for uncertainties associated with the number of X_i^j values, or equivalently, the number of available measurements N_i used as the basis for validation for that nuclide and is expressed in Eq. (4). The resulting σ_i serves as the uncertainty estimate [4] for nuclide i applied in Step C. Factor tf_2^i represents a two-sided tolerance-limit factor for the normal distribution to bound 68.3% of the population at a 95% confidence level and is applied to adjust s_i if at least 10 measurements [4] are available for nuclide i. If less than 10 measurements are available for that nuclide, then the adjustment factor tf_1^i is used, which represents the one-sided tolerance limit factor for the normal distribution to bound 95% of the population at a 95% confidence level. The magnitude of a tolerance factor increases as N_i decreases; for nuclides with a small number of measurements, a tolerance interval for uncertainty is significantly larger than a confidence interval. For example, the value of tf_2^i is 1.7255 for N_i = 10, and it decreases to 1.1392 for N_i = 100 [4].

$$\sigma_i = \begin{cases} s_i . tf_2^i & if\ N_i \geq 10 \\ s_i . tf_1^i & if\ N_i < 10 \end{cases} \quad (4)$$

These tolerance factors are applied to account for the size of the statistical sample (i.e., number of measurements for a given nuclide) and to capture potential uncertainties associated with the inability to establish a normal distribution for nuclides with a low number of measurements available. This adjustment will increase the uncertainty in the calculated nuclide concentration.

The model used for generating a distribution of expected concentrations for a given nuclide *i* in a fuel material with burnup *Bu* is described by Eq. (5),

$$C_{i,Bu}^m = C_{i,Bu}\left(\overline{X}_{i,Bu} + \sigma_{i,Bu} R_i^m\right), \quad (5)$$

where

i = index of burnup credit nuclide,

m = index of the randomly sampled concentration value for nuclide *i*,

$C_{i,Bu}$ = predicted concentration of nuclide *i* in a fuel mixture with burnup *Bu*,

$\overline{X}_{i,Bu}$= average M/C nuclide concentration ratio of nuclide *i* that corresponds to burnup *Bu*,

$\sigma_{i,Bu}$= uncertainty in $\overline{X}_{i,Bu}$ corresponding to nuclide *i* and burnup *Bu*,

R_i^m= random number selected from the standard normal distribution, and

$C_{i,Bu}^m$= sampled value of the calculated nuclide concentration of nuclide *i* for a fuel mixture of burnup *Bu*, from the expected range of values that correspond to $\overline{X}_{i,Bu}$ and $\sigma_{i,Bu}$.

The same random number R_i^m is applied to nuclide *i* for all fuel mixtures (i.e., all values of Bu) in the fuel assembly.

2.3.3 Step C. Calculate instances of k_{eff} for the criticality model

The reference criticality model for determining the bias and bias uncertainty in calculated k_{eff} consists of the cask model described in Section 2.2, in which all fuel assemblies have the same assembly-average burnup and the same axial burnup profile. Eighteen axially varying fuel mixtures are included in the assembly model, with each fuel mixture corresponding to a different burnup. The nuclide concentrations for these fuel mixtures, in the context of the stochastic method description, are the $C_{i,Bu}$ values in Eq. (5), and *i* varies from 1 to 28.

A perturbed instance of the reference criticality model (*m=1*) is obtained by replacing the nominal nuclide concentrations values $C_{i,Bu}$ with perturbed values $C_{i,Bu}^1$ calculated per Eq. (5). The nuclide concentrations are simultaneously sampled within the expected probability distribution for each nuclide. The stochastic sampling of the nuclide concentrations for a given nuclide *i* is consistent across all fuel mixtures in the model; in other words, the same random number R_i^1 is applied in Eq. (5) to determine the nuclide concentration $C_{i,Bu}^1$for nuclide *i* in each of the fuel mixtures.

The k_{eff} for the perturbed criticality model (*m=1*) is calculated, and the process is repeated *M* times from *m=1* to *m=M* by randomly sampling the nuclides concentrations for the reference criticality model to obtain *M* modified instances of this model. The k_{eff} is calculated for each perturbed model, and the result is a distribution of the k_{eff} values that reflect the effect of the random variations of the calculated nuclide concentrations. Selection of the M value to ensure that the resulting k_{eff} distribution is meaningful is discussed in Appendix A.2.

2.3.4 Step D. Estimate bias and bias uncertainty in calculated k_{eff} of the criticality model

The mean and the standard deviation of the distribution of k_{eff} values resulting from step C are calculated as

$$\overline{k}_{\text{eff}} = \frac{1}{M}\sum_{m=1}^{M} k_{\text{eff}}^{m} \text{ , and} \tag{6}$$

$$\sigma_{k_{\text{eff}}} = \sqrt{\frac{\sum_{m=1}^{M}\left(k_{\text{eff}}^{m} - \overline{k}_{\text{eff}}\right)^{2}}{M-1}}, \tag{7}$$

where

$\overline{k}_{\text{eff}}$ = mean of the calculated k_{eff} values,

M = number of the calculated k_{eff} values (i.e., number of perturbed cases),

$\sigma_{k_{\text{eff}}}$ = standard deviation of the calculated k_{eff} values, and

k_{eff}^{m} = k_{eff} value for simulation *m* in the series of *M* simulations.

The k_{eff} values from a statistically significant number of criticality calculations were shown [4] to approach a normal distribution with the mean and standard deviation given by Eqs. (6) and (7). Convergence was considered achieved when the values for $\overline{k}_{\text{eff}}$ and $\sigma_{k_{\text{eff}}}$ changed insignificantly (e.g., within ±0.0005) with an additional simulation.

The bias in k_{eff} and the bias uncertainty in k_{eff} are calculated as

$$k_{\text{eff}}\, bias = k_{\text{eff}}^{REF} - \overline{k}_{\text{eff}}\, and \tag{8}$$

$$k_{\text{eff}}\, bias\ uncertainty = \sigma_{k_{\text{eff}}}\, tf_{1}^{M}, \tag{9}$$

where

k_{eff}^{REF} = value of k_{eff} calculated for the reference criticality model (i.e., model using the calculated nuclide concentrations without any adjustment for bias or uncertainty), and

tf_1^M = one-sided tolerance-limit factor for the normal distribution corresponding to the number of calculated k_{eff} values (M), at a 95% probability, 95% confidence level.

The bias and bias uncertainty in calculated k_{eff} that result from bias and bias uncertainty in calculated nuclide concentrations—$\beta_i+\Delta k_i$ per terminology in Eq. (1)—are determined [4] as

$$\beta_i + \mathrm{D}k_i = \begin{cases} \left(k_{eff}^{REF} - \overline{k}_{eff}\right) + \sigma_{k_{eff}}\, tf_1^M & if\ \overline{k}_{eff} > k_{eff}^{REF} \\ \sigma_{k_{eff}}\, tf_1^M & if\ \overline{k}_{eff} \le k_{eff}^{REF} \end{cases} . \qquad (10)$$

An illustration of the main steps involved in the stochastic uncertainty sampling method is shown in Figure 2-4.

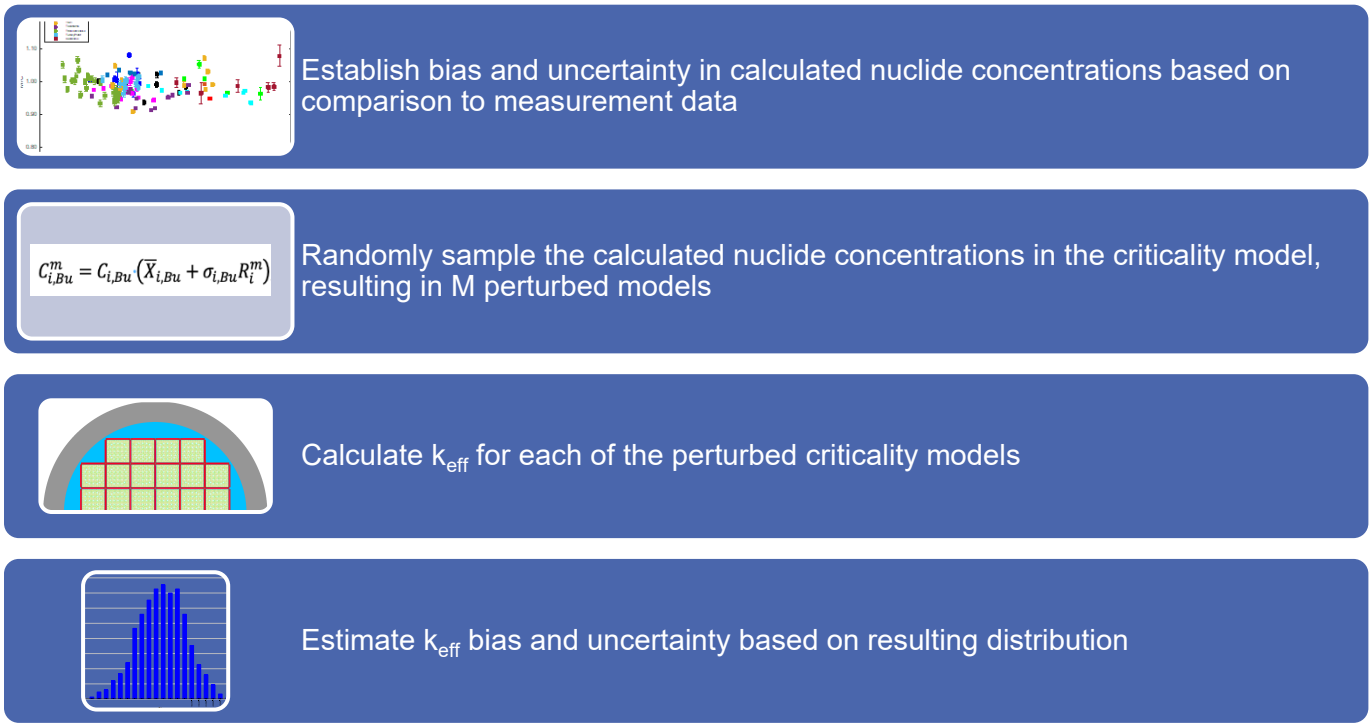


Figure 2-4 Illustration of Steps in Stochastic Uncertainty Sampling Method

3 LOADING CURVES

The loading curve for the GBC-32 cask model presented in NUREG/CR-7108 [4] was reassessed to verify its applicability when using the cask model with computational tools in SCALE 6.2.4 and 252-group ENDF/B-VII.1 cross section libraries. Based on the observed results, a new loading curve was generated to update the cask reference model that serves to determine the bias and bias uncertainty in k_{eff}. The updated loading curve corresponds to fuel compositions that include all 28 burnup credit nuclides (12 actinides and 16 fission products). The updated curve is generated based on the new methodology described briefly in this section. A loading curve considering actinides only was not an object of investigation, because all criticality calculations presented here consider both actinides and fission products. A new approach [23] based on the ORIGAMI computational tool in SCALE 6.2.4 is used to generate loading curves and is briefly summarized in section 3.3. Previously, loading curves were generated [4] based on the STARBUCS sequence in SCALE 6.1.3.

3.1 Burnup-Dependent Fuel Compositions for the Cask Model

The CSAS6 cask models referenced herein have the same geometry as the CSAS5 models used in NUREG/CR-7108. The enrichment and burnup-dependent fuel compositions for a particular CSAS6 model, which correspond to a given assembly's initial enrichment, average burnup, and axial profile, are determined using the following computational steps:

- Generate burnup-dependent ORIGEN libraries for the W 17×17 OFA assembly model for different values of the initial fuel enrichment by running TRITON with 252-group ENDF/B-VII.1 cross sections. These ORIGEN libraries are generated here for enrichments ranging from 1.5 to 8% and a burnup range of 0 to 90 GWd/MTU. The assembly depletion model has the same geometry and uses conservative reactor operating parameters that increase discharge reactivity [4,21], as previously used, with WABAs inserted into the assembly guide tubes throughout the irradiation history.

- Apply the burnup-dependent ORIGEN libraries as input to ORIGAMI and perform depletion and decay calculations for a specific burnup and a specific enrichment. For a given assembly's initial enrichment, average burnup, and axial profile, 18 ORIGAMI calculations are performed, with each calculation corresponding to a given axial burnup in the fuel assembly. Each calculation provides a fuel composition that corresponds to that axial burnup.

- Apply the 18 burnup-dependent fuel compositions from ORIGAMI as input in the cask CSAS6 criticality model.

A Python script was developed to automate the process of generating and running the ORIGAMI inputs and to apply the resulting fuel compositions in the CSAS6 input file.

3.2 Effect of Cross Section Libraries on the Loading Curve

Application of the loading curve developed in NUREG/CR-7108 [4] to the current CSAS6 cask model that uses fuel compositions generated as described in Section 5.1 resulted in k_{eff} values smaller than the 0.94 target value for a reference model. The differences between the target value and the CSAS6 k_{eff} values were in the 0.0024 to 0.0043 range. The CSAS6 k_{eff} values are illustrated in Figure 3-1, along with the k_{eff} values obtained for the CSAS5 models as applied in

NUREG/CR-7108. Figure 3-1 indicates on the x-axis the burnup in GWd/MTU and enrichment that correspond to each of the shown k_{eff} values. The fuel compositions in the CSAS5 models applied in NUREG/CR-7108 were generated using SCALE 6.1.3 and burnup-dependent ORIGEN libraries created using the 238-group ENDF/B-VII.0 cross sections with the TRITON depletion model of the fuel assembly [4]. No significant differences in k_{eff} are expected because a different criticality sequence within a given SCALE release was used (CSAS5 vs. CSAS6) with consistent input. For a consistent input, the k_{eff} values obtained with CSAS5 and CSAS6 are similar within the statistical uncertainty. However, some differences are expected in k_{eff} because of the change in cross sections between ENDF/B-VII.0 and ENDF/B-VII.1. This resulted in differences in the predicted concentrations for burnup credit nuclides.

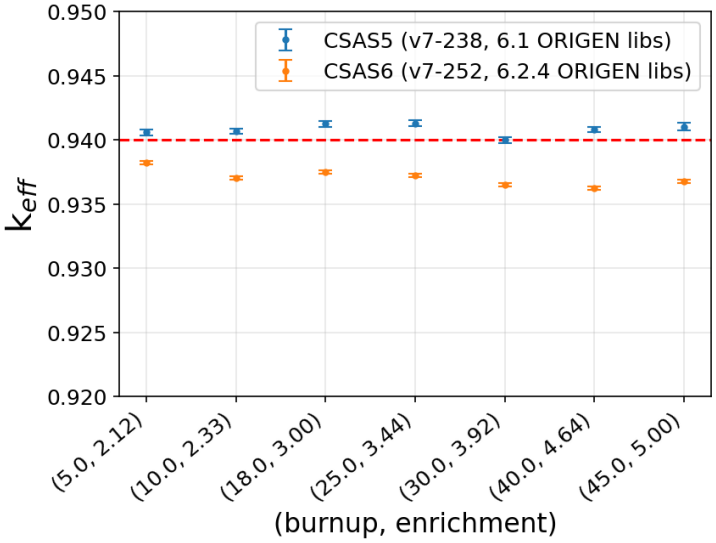


Figure 3-1 k_{eff} Obtained Using the Old Loading Curve [23]

3.3 New Approach for Loading Curve Generation

The new methodology [23] for generating a loading curve for the cask model is consistent with the approach used to generate burnup-dependent fuel compositions for that model. For a given assembly-average burnup and axial profile, the initial enrichment that would ensure reaching the target value of 0.94 for k_{eff} is determined iteratively. The enrichment that corresponds to the previous loading curve [4] for a given assembly-average burnup and axial profile serves as an initial guess and is used to perform a CSAS6 initial run to calculate an initial value for k_{eff}. The initial guess for enrichment and an assumed enrichment sensitivity coefficient (relative to k_{eff}) are applied to predict a new enrichment. Fuel compositions are generated for this new enrichment using ORIGAMI and are applied in the CSAS6 criticality model. The updated CSAS6 model is rerun, resulting in a new k_{eff} value. The process is repeated with the updated sensitivity coefficient based on the new value of k_{eff} until the target of 0.94 for k_{eff} is reached. The new methodology for loading curve generation is implemented through a Python script.

The loading curve is extended above 45 GWd/MTU burnup and 5% enrichment, which were the limits considered in NUREG/CR-7108. The extension led to enrichments greater than the 5% enrichment limit of current commercial fuel. Note that the current RCA validation basis covers burnups up to 80 GWd/MTU and initial enrichments up to 5%. The new loading curve,

generated for a target k_{eff} of 0.94, is illustrated in Figure 3-2. The space above the curve—points defined by assembly average burnup and initial enrichment—corresponds to k_{eff} values smaller than 0.94 and is acceptable, whereas the space under the curve is not acceptable. The horizontal segments of the loading curve are caused by the changes in the axial fuel burnup profiles (see Figure 2-3) at 18 and 30 GWd/MTU.

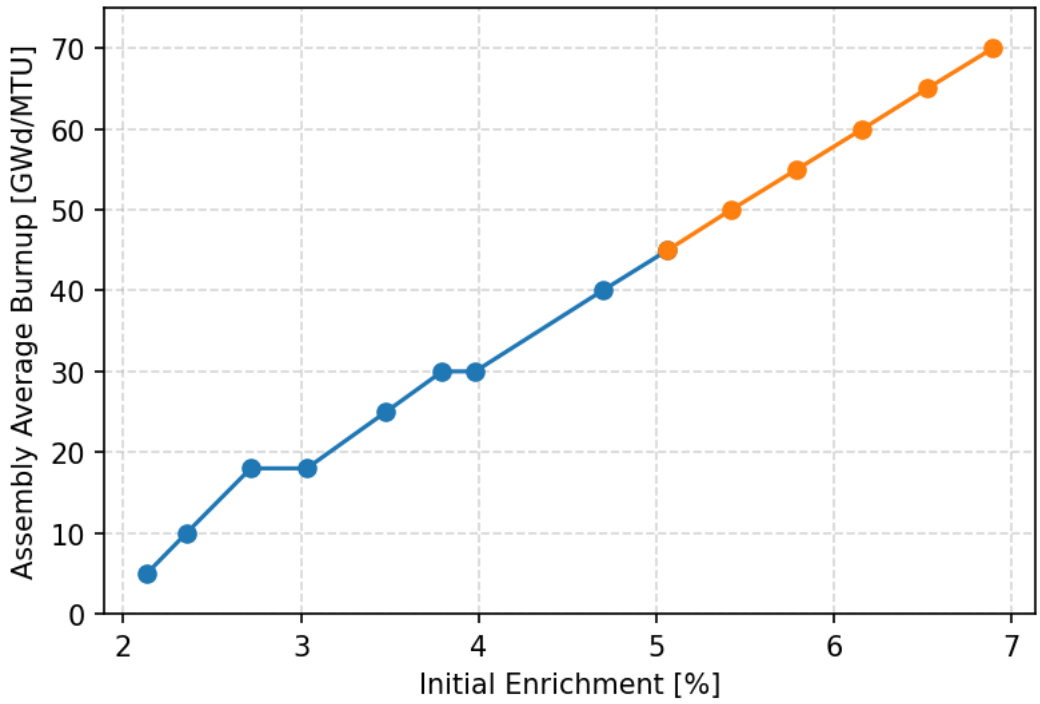


Figure 3-2 Loading Curve for the GBC-32 Cask (12 Actinides and 16 Fission Products)

The burnup and enrichment values for the new loading curve are listed in Table 3-1, along with the enrichment values for the old loading curve developed in NUREG-7108. For a given assembly burnup, the difference between the new and previous enrichment value varies in the range 0.2 to 0.6%, with the difference increasing with increasing burnup.

Table 3-1 Comparison of New and Old Loading Curve Enrichments

Assembly burnup (GWd/MTU)	New enrichment (%)	Old enrichment (%)
5	2.12	2.13
10	2.33	2.36
18	3.00	3.03
25	3.44	3.48
30	3.92	3.98
40	4.64	4.70
45	5.00	5.05

4 COMPUTATIONAL TOOLS AND ASSOCIATED NUCLEAR DATA

The computational tools and nuclear data used for simulations presented in this report are briefly described herein. All these tools are available in version 6.2.4 of the SCALE nuclear analysis modeling and simulation suite for nuclear safety analysis and design [7]. SCALE (https://www.ornl.gov/scale) is developed and maintained by ORNL under contract with the NRC, DOE, and the National Nuclear Security Administration (NNSA) to perform reactor physics, criticality safety, radiation shielding, and spent fuel characterization for nuclear facilities and transportation/storage package designs.

Validation of capabilities and associated nuclear data in the SCALE code system has been a continuous effort at ORNL since these capabilities were first developed. A comprehensive validation report of SCALE 6.2.4 was recently published. The validation report addresses, among other applications, SCALE's capabilities for nuclide inventory characterization and nuclear criticality safety [24–26].

The criticality simulations are performed with the 3D Monte Carlo criticality codes KENO-V.a and KENO-VI under the CSAS5 and CSAS6 criticality safety analysis sequences in SCALE 6.2.4 [27,28]. The results for bias and bias uncertainties in k_{eff} in this report are obtained with KENO-VI and 252-group ENDF/B-VII.1 cross sections. However, KENO-V.a is also used herein for verification purposes and for consistent comparisons with the criticality simulations in NUREG/CR-7108, which employed KENO-V.a with 238-group ENDF/B-VII.0 cross sections.

The fuel depletion and decay simulations for obtaining the burnup-dependent nuclide concentrations applied in the criticality models include the use of the TRITON [7,29], ORIGAMI [7,30], and ORIGEN [7,31] capabilities in SCALE 6.2.4. Depletion and decay simulations for the considered W17×17 OFA assembly are performed using the 2D depletion capability under the TRITON depletion sequence to generate burnup-dependent ORIGEN libraries for this assembly. In this case, TRITON iteratively couples the 2D neutron transport solver NEWT [29] and the ORIGEN nuclear transmutation and decay code. The burnup-dependent ORIGEN library is generated during the TRITON simulation and includes the burnup-dependent nuclear data (cross sections as function of burnup, fission yields, and decay data) that enable burnup-dependent ORIGEN standalone simulations. ORIGEN libraries are generated as a function of burnup and initial fuel enrichment. The generated ORIGEN libraries serve as input to ORIGAMI for performing fast depletion simulations with ORIGEN for the burnups and initial fuel enrichments of interest. ORIGAMI also serves under the new approach for generating loading curves for the PWR cask model as described in Section 3. Note that in NUREG/CR-7108, the STARBUCS sequence in SCALE 6.1.3 was used to determine burnup-dependent nuclide concentrations for PWR criticality calculations and to generate loading curves for the PWR analysis models [4].

The Sampler uncertainty quantification tool [7,9] in SCALE 6.2.4 supports the new and standardized implementation of the stochastic uncertainty sampling method for k_{eff} bias and bias uncertainty estimation. Sampler, which was released in SCALE in 2016, is a sequence that can propagate uncertainties in nuclear data or input parameters to estimate the resulting uncertainty in a SCALE calculated response. These uncertainties are propagated through all computational steps in the simulation, providing a fully coupled analysis that accounts for all correlations inherent to the calculation. In the current work, Sampler is used to perturb the input nuclide concentrations in the criticality model's fuel materials to determine the effect on the calculated k_{eff}. Details about this implementation are provided in Appendix A.

5 NUCLIDE INVENTORY VALIDATION DATA AND RESULTS

The nuclide inventory validation basis applied in the current work for determining the bias and bias uncertainty in code-predicted nuclide concentrations for burnup credit nuclides in PWR spent nuclear fuel is significantly improved. The validation basis now includes RCA measurement data for 129 PWR spent fuel samples covering a burnup range up to 80 GWd/MTU.

Measurement data for 9 of the 129 samples became available recently [11,12]. Under NRC and DOE support, ORNL is conducting experiments to measure important actinides and fission products in 16 PWR spent fuel samples from fuel irradiated in the North Anna PWR. These samples were selected from 6 of the 25 sibling rods that have served for post-irradiation examinations performed by ORNL and Pacific Northwest National Laboratory (PNNL) to study the effects of long-term storage and transportation for light-water reactor (LWR) high-burnup fuel under the DOE Nuclear Energy High Burnup Spent Fuel Data Project [32]. Measurement data are currently available for 9 of the 16 North Anna samples, and the experimental effort is ongoing.

In the future, the RCA validation basis for PWR spent fuel will be further improved with the addition of newer high-quality RCA measurements, as they become available. Under the ongoing ORNL experimental program [11] mentioned previously, measurements are being conducted on the seven other high burnup fuel samples. Additionally, measurement data will become available in the future from the REGAL international experimental program [33] led by SCK-CEN. ORNL participates in this program through funding from the NRC. The first phase of REGAL includes measurements on samples from two UO_2 fuel rods with an estimated rod-average burnup of approximately 50 GWd/MTU and from one gadolinia fuel rod with an estimated burnup of 12 GWd/MTU.

5.1 Nuclide Inventory Validation Data

Table 5-1 lists the main characteristics of the fuel rods from which the 129 considered samples were selected, including assembly lattice, fuel enrichment, and burnup range. These fuel rods were irradiated in 13 PWRs operated in six countries: Germany, Italy, Japan, Spain, Switzerland, and the United States.

The samples that were recently added to the validation basis include the following:

- 6 Vandellos samples [34],
- 5 Three Mile Island (TMI)-1 samples measured at ORNL in 2011 [35],
- 3 H.B. Robinson samples [36], 8 Turkey Point 3 samples [36], and 6 Ohi samples [36], for which measurement reports became available through the SFCOMPO database [37],
- 3 Gosgen samples measured under the MALIBU program [13],
- 9 North Anna samples measured at ORNL in 2022 [11,12].

Eleven of the TMI-1 samples from fuel rod H6 of assembly NJ05YU, which were considered in NUREG/CR-7108 [4], are not included herein. The experimental data for these samples

exhibited higher biases and variability compared to other measurement data for fuel from the same assembly or from other experimental programs. These were primarily associated with the lower precision of the experiment, large discrepancies for the Pu measurements, and large uncertainty of 7% in the estimated sample burnup [38,39]. The measurements for these 11 samples were applied to code validation at the time because they included fission product measurements that provided an opportunity to evaluate uncertainty for nuclides with very few measurements. Notably, the 5 TMI-1 samples that were added in this effort, which were measured at ORNL in 2011 [35], originated from the same fuel assembly as the former 11 TMI-1 samples—2 are from fuel rod H6 and 3 from fuel rod D5—and had estimated burnups in the same 45–55 GWd/MTU burnup range.

5.1.1 Fuel sample characteristics

The samples listed in Table 5-1 span a wide range of PWR assembly designs, including 14 × 14, 15 × 15, 17 × 17 and 18 × 18 fuel rod lattices; fuel enrichments ranging from 1.6874 to 4.657%; and sample burnups ranging from 7.2 to 78.3 GWd/MTU. Approximately one quarter (i.e., 31) of the 129 considered samples have burnups greater than 40 GWd/MTU. The distribution of burnup as a function of enrichment for the 129 considered samples is illustrated in Figure 5-1; the colors indicate the fuels' origins by reactor name. The histogram of the sample burnups is presented in Figure 5-2.

Table 5-1 Summary of PWR RCA Data Used for Validation Data [39]

Reactor	Country	Measurement laboratory [a]	Experimental program	Assembly lattice	Enrichment (%)	No. of samples / no. fuel rods	Burnup (GWd/MTU)
Calvert Cliffs-1	US	PNL, KRI	ATM-103, ATM-104, ATM-106 [b]	14 × 14	2.453, 2.72, 3.038	9/3	18.7–46.5
GKN II	Germany	SCK·CEN	REBUS [c]	18 × 18	3.8	1/1	54.1
Gösgen	Switzerland	SCK·CEN, ITU	ARIANE, MALIBU [c]	15 × 15	3.5, 4.1, 4.3	6/3	29.1–70.4
H. B. Robinson-2	US	PNL, LANL	ATM-101	15 × 15	2.561	7/3	16.0–31.7
North Anna	US	ORNL	NRC/DOE/ORNL	17 × 17	3.6, 4.0, 4.2, 4.5	9/5	33.2–67.3
Obrigheim	Germany	JRC Ispra, ITU, IRCh, WAK	EUR, ICE	14 × 14	2.83, 3.00, 3.13	15/7	17.1–30.9
Ohi-1	Japan	JAERI	JAERI	17 × 17	3.2	1/1	52.4
Ohi-2	Japan	JAERI	JAERI	17 × 17	1.6874, 3.2	5/3	21.5–38.5
Takahama-3	Japan	JAERI	JAERI	17 × 17	2.63, 4.11	13/3	17.4–47.3
TMI-1	US	GE-VNC, ORNL	DOE YMP [d], ORNL	15 × 15	4.013, 4.657	13/5	22.8–55.0
Trino Vercellese	Italy	JRC Ispra, Karlsruhe	EUR	15 × 15	2.719, 3.13, 3.897	31/10	7.2–25.3
Turkey Point-3	US	Battelle-Columbus	NWTS	15 × 15	2.556	13/7	19.9–31.6
Vandellos II	Spain	Studsvik	CSN/ENUSA [e]	17 × 17	4.5	6/2	43.5–78.3

[a]ANL = Argonne National Laboratory; GE-VNC = General Electric Vallecitos Nuclear Center; PNNL = Pacific Northwest National Laboratory; KRI = Khlopin Radium Institute; JAERI = Japan Atomic Energy Research Institute (now Japan Atomic Energy Agency); JRC = Joint Research Center, European Commission; ITU = European Institute for Transuranium Elements; IRCh = Institute for Radiochemistry at Karlsruhe; WAK = Karlsruhe Reprocessing Plant; IAEA = International Atomic Energy Agency; SCK·CEN = Studiecentrum voor Kernenergie – Centre d'étude de l'Énergie Nucléaire; PSI = Paul Scherrer Institute; CEA = Commissariat à l'Énergie Atomique;
[b]ATM = approved testing material
[c]International experimental programs initially managed by Belgonucleaire, then by SCK·CEN (Belgium)
[d]DOE YMP = US Department of Energy Yucca Mountain Project
[e]CSN = Consego de Securidad Nuclear; ENUSA = Empresa Nacional del Uranio, SA (Spain)

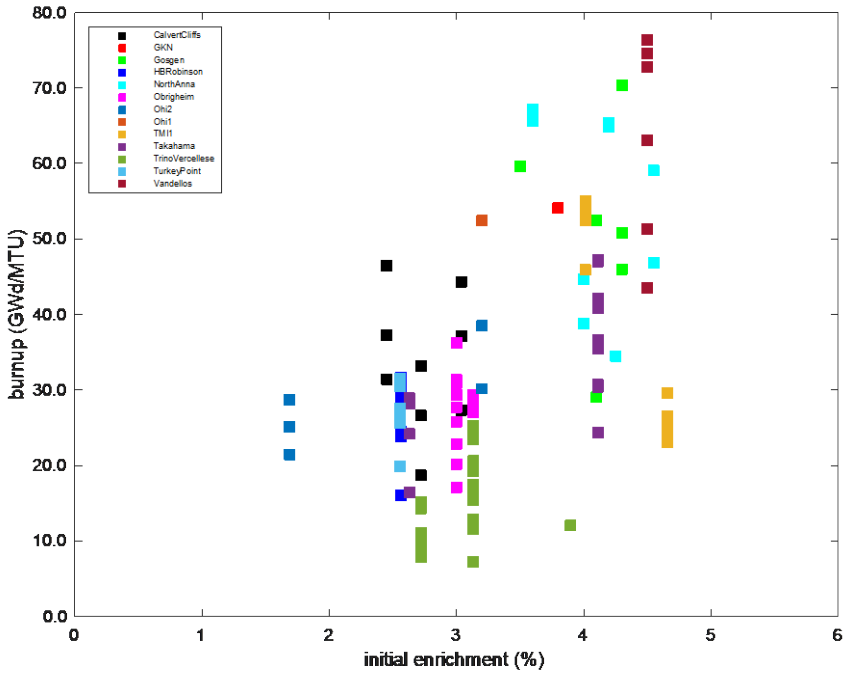


Figure 5-1 Burnup vs. Enrichment for the Measured Fuel Samples

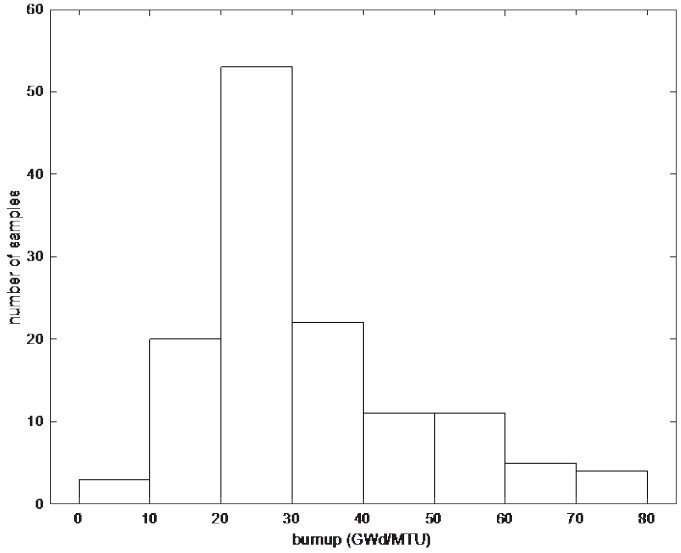


Figure 5-2 Burnup Distribution Histogram for the Measured Fuel Samples

5.1.2 Number of measurements for burnup credit nuclides

Burnup credit nuclides [1,2] consist of 12 actinides and 16 fission products with large fission cross sections and/or large neutron absorption cross sections, and which are stable or have relatively long half-lives. The numbers of measurements available for these nuclides in this validation are shown in Table 5-2, along with the half-lives of the nuclides and the burnup ranges of the measured samples. The number of measurements per nuclide and the corresponding burnup range for the data used in NUREG/CR-7108 [4] are also included for comparison.

Nine of the 12 actinides listed in Table 5-2 (^{234}U, ^{235}U, ^{238}U, ^{238}Pu, ^{239}Pu, ^{240}Pu, ^{241}Pu, ^{242}Pu, and ^{241}Am) account for about 95% of the reactivity's worth of the actinides and close to 70% of the total reactivity's worth of all nuclides in typical spent fuel. Whereas 6 nuclides (^{143}Nd, ^{149}Sm, ^{103}Rh, ^{151}Sm, ^{133}Cs, and ^{155}Gd) of the listed 16 fission products account for approximately 75% of the fission product worth and 20% of the total worth in typical spent nuclear fuel [40].

Uranium and plutonium nuclide measurements are available for most of the considered samples. In the past, application of fission product burnup credit was negatively impacted [19] by the limited availability of RCA measurement data to allow adequate validation for these nuclides. The validation basis presented herein is expanded overall for the fission products, particularly for the top six contributors, with ^{133}Cs being the nuclide with the largest relative increase in the number of measurements.

Table 5-2 Number of RCA Measurements per Nuclide

Nuclide	Nuclide half-life	Number of measurements [herein]	Burnup range (GWd/MTU) [herein]	No. of measurements NUREG/CR-7108 [4]	Burnup range (GWd/MTU) NUREG/CR-7108 [4]
^{234}U	2.45×10^{5} yr	86	7.2–70.3	63	7.2–59.7
^{235}U	7.04×10^{8} yr	128	7.2–78.3	100	7.2–59.7
^{236}U	2.34×10^{7} yr	113	12.9–78.3	85	12.9–59.7
^{238}U	4.47×10^{9} yr	123	7.2–70.3	100	7.2–59.7
^{237}Np	2.14×10^{6} yr	49	16.0–77.0	44	16.0–59.7
^{238}Pu	87.71 yr	112	12.9–78.3	85	12.9–59.7
^{239}Pu	2.41×10^{4} yr	127	7.2–78.3	100	7.2–59.7
^{240}Pu	6.56×10^{3} yr	127	7.2–78.3	100	7.2–59.7
^{241}Pu	14.29 yr	127	7.2–78.3	100	7.2–59.7
^{242}Pu	3.75×10^{5} yr	126	7.2–78.3	99	7.2–59.7
^{241}Am	433 yr	58	16.4–78.3	47	17.1–59.7
^{243}Am	7,370 yr	65	16.4–78.3	48	17.1–59.7
^{95}Mo	stable	20	29.1–70.3	15	31.1–59.7
^{99}Tc	2.11×10^{5} yr	33	16.0–70.3	25	16.0–59.7
^{101}Ru	stable	15	29.1–70.3	15	31.1–59.7
^{103}Rh	stable	21	29.1–70.3	16	31.1–59.7
^{109}Ag	stable	9	45.9–59.7	14	44.8–59.7
^{133}Cs	stable	26	27.4–77.0	7	27.4–59.7
^{143}Nd	stable	62	16.0–78.3	44	16.0–59.7
^{145}Nd	stable	62	16.0–78.3	44	16.0–59.7
^{147}Sm	1.06×10^{11} yr	42	23.0–78.3	32	23.7–59.7
^{149}Sm	stable	38	23.0–77.0	28	23.7–59.7
^{150}Sm	stable	44	23.0–78.3.	32	23.7–59.7
^{151}Sm	90 yr	43	23.0–78.3	32	23.7–59.7
^{152}Sm	stable	44	23.0–78.3	32	23.7–59.7
^{151}Eu	stable	26	23.0–70.3	21	23.7–59.7
^{153}Eu	stable	38	23.0–78.3.	27	23.7–59.7
^{155}Gd	stable	38	21.5–78.3	27	23.7–59.7

5.2 Bias and Uncertainty in Calculated Nuclide Concentration

Individual TRITON depletion models are developed for each measured fuel sample to simulate the sample irradiation and decay history. These models are similar to those applied in previous validation studies that were performed with different SCALE versions and cross section data [41-43]. They are 2D models representing quarter, half, or full assembly configurations, depending on the applicable symmetry and the ease of modeling. All TRITON simulations presented herein are performed using SCALE version 6.2.4 with 252-group ENDF/B-VII.1 cross sections. Details on TRITON computational models, measurement data, and results obtained for subsets of measurement data used in this effort can be found in other SCALE validation studies [26,41-43].

The calculated nuclide concentrations are compared with the corresponding measurement data to determine the mean M/C nuclide concentration ratio and the associated standard deviation per Eqs. (2) and (3). The mean represents the bias, and the bias uncertainty is determined per Eq. (4). The bias and bias uncertainty in the calculated nuclide concentrations for the burnup credit nuclides are presented in Table 5-3 for actinide nuclides and in Table 5-4 for fission product nuclides. The biases for different nuclides are known to be strongly correlated because of the inherent associated transmutation and decay physics [19,22]. However, variations around the bias for a given nuclide are assumed to be independent of variations for the other nuclides in the stochastic uncertainty sampling method. By sampling the nuclide concentrations independently, this approach inherently assumes that the uncertainties in individual nuclide concentrations are independent [4,19,22]. This assumption was previously reviewed [4] by evaluating the degree of correlations among nuclide uncertainties and their effect on the k_{eff} bias uncertainty for the cask analysis model. It was shown that the k_{eff} bias uncertainty was dominated by ^{235}U and ^{239}Pu, which correlation was statistically insignificant, whereas correlations for all other nuclides had relatively small contributions to k_{eff} bias uncertainty.

Table 5-3 shows the bias and bias uncertainty in calculated actinide nuclides concentrations over two burnup ranges: $\leq$ 40 GWd/MTU and 40–80 GWd/ MTU. Compared to measurement data, ^{235}U is on average underpredicted by approximately 1.2% for lower burnups and overpredicted by 1.2% for higher burnups: the associated bias uncertainty is 3.8% for lower burnups and 8.3% for higher burnups. The variation with burnup of ^{235}U content transitions from linear decrease at lower burnups to exponential decrease at higher burnups; a small uncertainty in the sample burnup at higher burnups will affect the calculated nuclide content to a greater extent than at lower burnups. For ^{239}Pu, one of the top actinides impacting reactivity, the content is underpredicted on average by close to 1.5% for both burnup ranges considered, and the associated uncertainty is less than 4.5%.

Notably, the bias uncertainties of major actinides ^{235}U and ^{239}Pu, other plutonium, and americium isotopes are smaller in the current analysis than the corresponding values in NUREG/CR-7108 [4] for burnups greater than 40 GWd/ MTU. This reduction results primarily from: improved statistics from increasing the number of measurements for a given nuclide; replacement of the problematic TMI-1 data with newer measurements of the same fuel; and recent high-quality, low-uncertainty measurements for high burnup fuel. These changes led to less variability of bias values. Detailed comparisons are presented in Section 5.3.

Previous validation studies [26,42,43] indicate that on average, the prediction of uranium isotopes does not significantly change when using SCALE 6.1/ENDF/B-VII.0 vs. SCALE 6.2.4/ENDF/B-VII.1. A significant improvement was noted for bias of plutonium isotopes (smaller bias) when using SCALE 6.2.4/ENDF/B-VII.1, particularly for ^{239}Pu and ^{238}Pu; consequently,

changes in plutonium nuclides impacted the calculation of nuclide contents in the higher actinide americium. Part of the change in bias and bias uncertainty for plutonium isotopes in this work is the result of the change in nuclear cross sections used in the simulations, from ENDF/B-VII.0 in NUREG/CR-7108 to ENDF/B-VII.1. The observed differences are not impacted as significantly by the different SCALE version used, given that the underlying computational methods were not changed significantly between the two SCALE versions. The improvement in the bias for plutonium isotopes was attributed mostly to the significant change in the (n,γ) cross section for ^{238}Pu between ENDF-B/VII.0 and ENDF-B/VII.1 [26,43].

Table 5-3 Nuclide Bias and Bias Uncertainty for Actinides

Burnup range / Nuclide	< 40 GWd/t			40 GWd/t - 80 GWd/t		
	No. of meas.	Bias	Bias uncertainty	No. of meas.	Bias	Bias uncertainty
^{234}U	61	0.9136	0.1496	25	0.9276	0.1001
^{235}U	98	0.9883	0.0383	30	1.0121	0.0829
^{236}U	83	1.0194	0.0407	30	0.9841	0.0274
^{238}U	98	1.0010	0.0044	25	1.0029	0.0079
^{237}Np	28	1.0634	0.1456	21	1.0828	0.1778
^{238}Pu	83	1.0538	0.1012	29	0.9765	0.0848
^{239}Pu	98	0.9855	0.0371	29	0.9864	0.0452
^{240}Pu	98	1.0020	0.0370	29	0.9727	0.0337
^{241}Pu	98	1.0334	0.0614	29	1.0101	0.0424
^{242}Pu	97	1.0504	0.0826	29	1.0035	0.0691
^{241}Am	28	0.9442	0.1997	30	0.9493	0.2054
^{243}Am	36	1.1024	0.2246	29	1.0542	0.1511

The variation of the bias with burnup is illustrated in Figures 5-3 and 5-4 for ^{235}U and ^{239}Pu. In these figures, the y-axis shows the M/C nuclide concentration ratio corresponding to each of the measured samples, with 128 values for ^{235}U and 127 values for ^{239}Pu. These two major actinides were measured in all samples; however, ^{239}Pu final data for two North Anna fuel samples were not available by the time of this analysis, while ^{235}U measured in one Vandellos fuel sample was not recommended due to experimental issues [34]. The different colors in Figures 5-3 and 5-4 indicate the origin of the measured fuel by reactor name. The error bars account for the reported measurement uncertainty and are shown only for qualitative purposes. They are not used in any calculations in this work. The reported measurement uncertainties differ greatly among measurement laboratories and cannot be used consistently, due to inconsistencies in the uncertainty analysis techniques used to report them [44].

Table 5-4 presents the bias and bias uncertainty in calculated fission product nuclide concentrations over the entire burnup range for which measurement data are available for

validation. When compared to NUREG/CR-7108, notable changes in bias and bias uncertainty are observed for ^{133}Cs and ^{155}Gd, as presented in the next section. These changes are largely caused by the increased number of measurements used for validation, particularly the addition of the recent measurements on North Anna spent fuel [11,12]. The bias variation with burnup is illustrated in Figure 5-5 for ^{133}Cs.

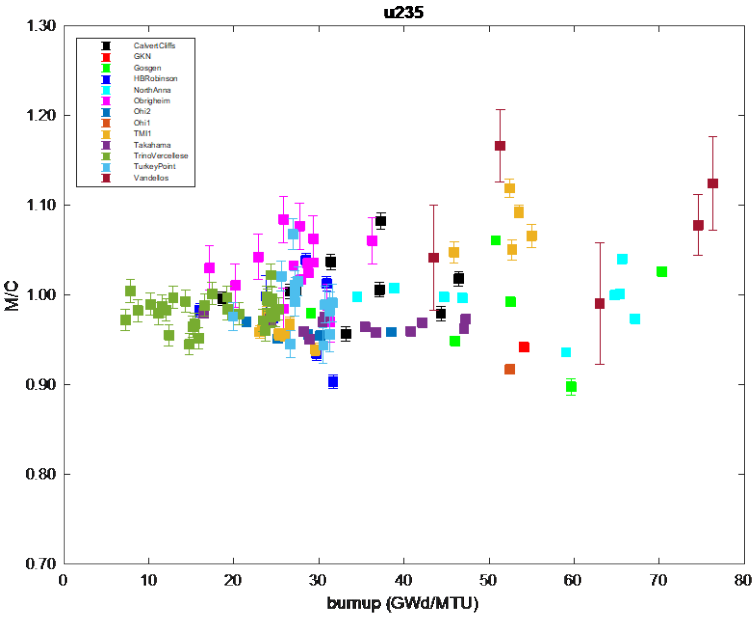


Figure 5-3 M/C Ratio vs. Burnup for ^{235}U

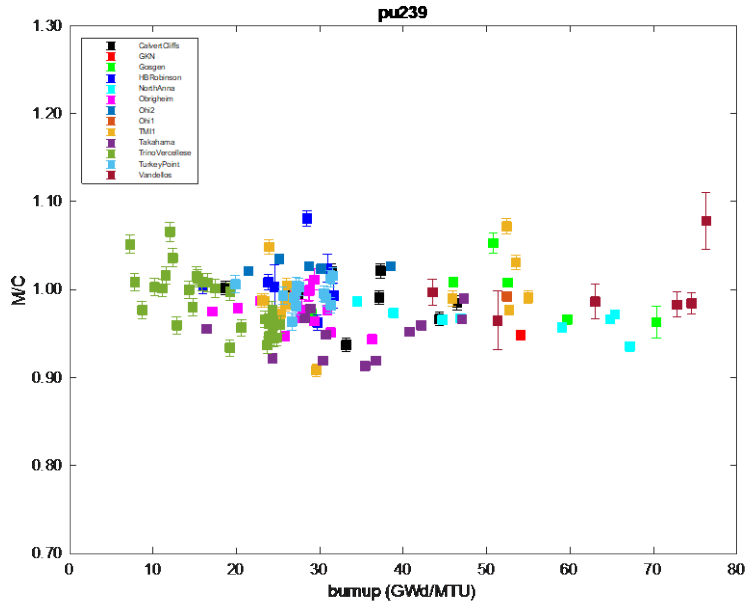


Figure 5-4 M/C Ratio vs. Burnup for ^{239}Pu

Table 5-4 Nuclide Bias and Bias Uncertainty for Fission Products

Burnup range	< 80 GWd/t		
Nuclide	No. of meas.	Bias	Bias uncertainty
^{95}Mo	20	1.0353	0.1522
^{99}Tc	33	0.9877	0.2343
^{101}Ru	15	0.9468	0.1314
^{103}Rh	21	0.9524	0.1607
^{109}Ag	9	0.8146	0.6870
^{133}Cs	26	0.9732	0.0234
^{143}Nd	62	0.9906	0.0288
^{145}Nd	62	0.9969	0.0186
^{147}Sm	42	0.9943	0.0370
^{149}Sm	38	0.9715	0.1238
^{150}Sm	44	0.9812	0.0386
^{151}Sm	43	1.0360	0.0658
^{152}Sm	44	0.9974	0.0395
^{151}Eu	26	1.0964	0.4285
^{153}Eu	38	1.0212	0.1034
^{155}Gd	38	1.0471	0.2243

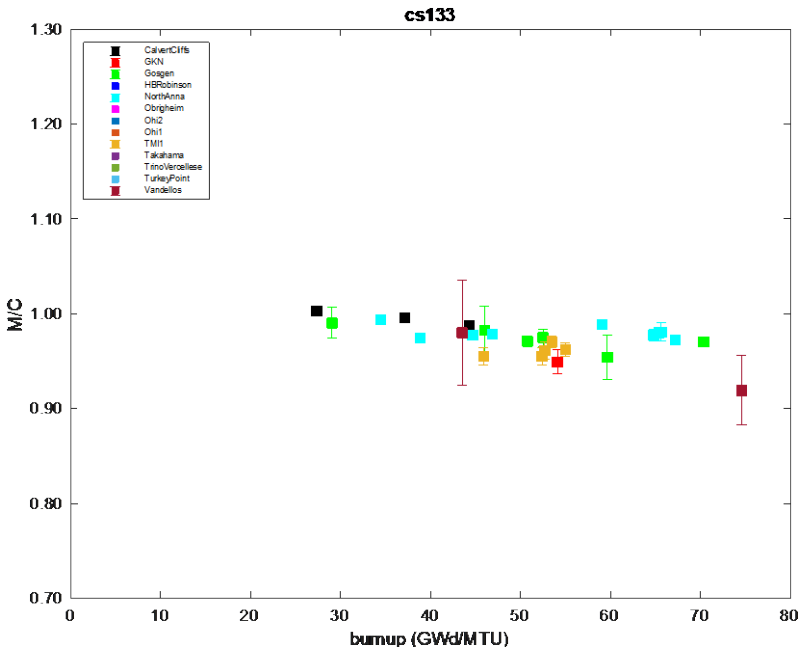


Figure 5-5 M/C Ratio vs. Burnup for ^{133}Cs

5.3 Comparison to Bias and Uncertainty in Calculated Nuclide Concentrations from NUREG/CR-7108

Bias and uncertainty in calculated actinides concentrations corresponding to the current validation basis, compared to the previous one [4], are generally not significantly changed for burnups smaller than 40 GWd/MTU. The measurement data available for validation are practically the same for low burnups. Significant changes are observed for bias uncertainty in some actinides at burnups greater than 40 GWd/MTU: these are caused by the removal of the lower-precision TMI-1 measurement data and addition of new data, primarily from the Vandellos and North Anna samples. For example, the relative bias uncertainty in ^{235}U is on average reduced from approximately 11% [4] to 8.3% herein. Whereas, for ^{239}Pu the relative bias uncertainty decreases from the previous value of 7.3% to 4.5% in current work. Comparisons for all burnup credit actinides are presented in Table 5-5 and illustrated in Figure 5-6. The bias calculated herein is closer to 1.0 (i.e., better prediction) than the previous values for most actinides; relative bias improvement is greater than 5% for ^{235}U, ^{238,239,240,242}Pu, ^{237}Np and ^{243}Am. Except for ^{234,236,238}U, and ^{242}Pu, a reduction in the relative bias uncertainty of more than 2% is observed for all other actinides.

Table 5-5 Bias and Bias Uncertainty for Actinides for Burnups > 40 GWd/MTU Compared to Previous Data

Nuclide	This report		NUREG/CR-7108		Difference[a]	
	Bias	Bias uncertainty	Bias	Bias uncertainty	Bias	Bias uncertainty
^{234}U	0.9276	0.1001	0.9114	0.1077	0.0162	-0.0076
^{235}U	1.0121	0.0829	0.9459	0.1096	0.0662	-0.0267
^{236}U	0.9841	0.0274	0.9862	0.0303	-0.0021	-0.0029
^{238}U	1.0029	0.0079	1.0020	0.0021	0.0009	0.0058
^{237}Np	1.0828	0.1778	1.0011	0.1072	0.0817	0.0706
^{238}Pu	0.9765	0.0848	1.1375	0.2331	-0.1610	-0.1483
^{239}Pu	0.9864	0.0452	0.8984	0.0727	0.0880	-0.0275
^{240}Pu	0.9727	0.0337	0.8981	0.0810	0.0746	-0.0473
^{241}Pu	1.0101	0.0424	0.9833	0.0839	0.0268	-0.0415
^{242}Pu	1.0035	0.0691	1.0636	0.0852	-0.0601	-0.0161
^{241}Am	0.9493	0.2054	0.9947	0.3224	-0.0454	-0.1170
^{243}Am	1.0542	0.1511	0.9216	0.2124	0.1326	-0.0613

[a] Values in this report compared to those in NUREG/CR-7108.

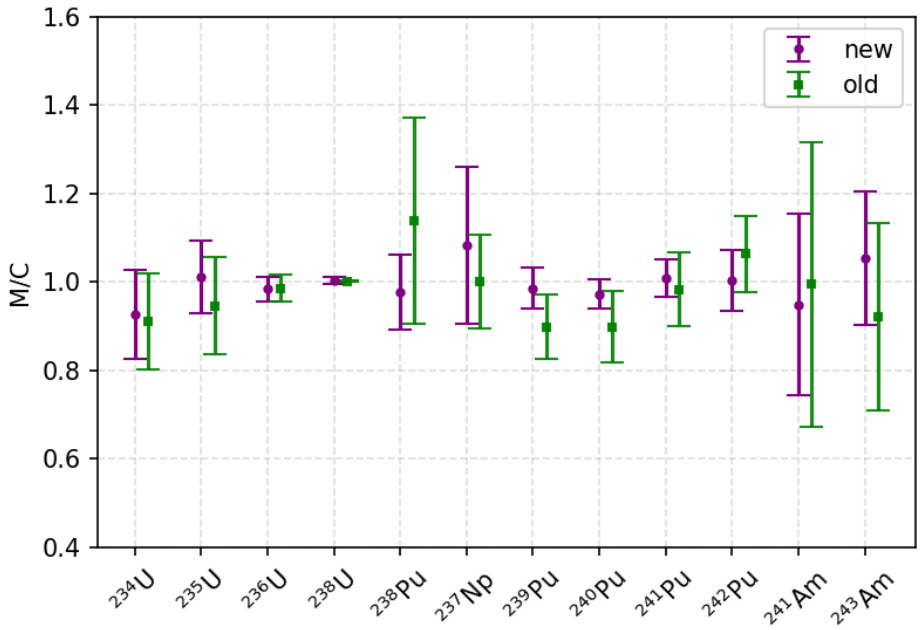


Figure 5-6 Bias and Bias Uncertainty for Actinides Compared to Previous Data for Burnups > 40 GWd/MTU

For fission products, notable reductions are observed between the previous and the current values of bias and bias uncertainty for ^{155}Gd, one of the six top fission products contributing to reactivity. For ^{155}Gd, the relative bias and bias uncertainty changed from 26% and 34% previously to 5% and 22% in this work. Comparisons for all burnup credit fission products are presented in Table 5-6 and illustrated in Figure 5-7. Better prediction (i.e., bias closer to 1.0) and a relative bias change greater than 5% are observed for ^{99}Tc, ^{103}Rh, ^{109}Ag, ^{147}Sm and ^{151}Eu. The bias uncertainty decreases for 9 of the 16 fission products; for the other 7 fission products, it increases, with increases greater than 5% in the relative bias uncertainty being observed for ^{95}Mo, ^{103}Rh, ^{109}Ag and ^{153}Eu. For these four fission products, the bias and uncertainty changes are primarily due to the change in the individual M/C values used as a basis, given the addition of new samples and removal of the 11 TMI-1 samples. The largest bias uncertainty increase that is observed for ^{109}Ag is primarily due to the smaller number of measurements in the current validation set than in the previous one, and this increase is associated with the application of a larger tolerance factor than that applied previously [4] to account for the limited number of measurements.

Table 5-6 Bias and Bias Uncertainty for Fission Products Compared to Previous Data

Nuclide	This report		NUREG/CR-7108		Difference[a]	
	Bias	Bias uncertainty	Bias	Bias uncertainty	Bias	Bias uncertainty
^{95}Mo	1.0353	0.1522	1.0002	0.0745	0.0351	0.0777
^{99}Tc	0.9877	0.2343	0.9400	0.2030	0.0477	0.0313
^{101}Ru	0.9468	0.1314	0.9726	0.1152	-0.0258	0.0162
^{103}Rh	0.9524	0.1607	0.9021	0.0894	0.0503	0.0713
^{109}Ag	0.8146	0.6870	0.5546	0.2694	0.2600	0.4176
^{133}Cs	0.9732	0.0234	0.9810	0.0680	-0.0078	-0.0446
^{143}Nd	0.9906	0.0288	0.9779	0.0526	0.0127	-0.0238
^{145}Nd	0.9969	0.0186	0.9978	0.0291	-0.0009	-0.0105
^{147}Sm	0.9943	0.0370	0.9379	0.0967	0.0564	-0.0597
^{149}Sm	0.9715	0.1238	0.9634	0.0995	0.0081	0.0243
^{150}Sm	0.9812	0.0386	0.9656	0.0663	0.0156	-0.0277
^{151}Sm	1.0360	0.0658	0.9961	0.0782	0.0399	-0.0124
^{152}Sm	0.9974	0.0395	0.9736	0.0427	0.0238	-0.0032
^{151}Eu	1.0964	0.4285	1.4721	0.7644	-0.3757	-0.3359
^{153}Eu	1.0212	0.1034	0.9967	0.0480	0.0245	0.0554
^{155}Gd	1.0471	0.2243	1.2556	0.3391	-0.2085	-0.1148

[a] Values in this report compared to those in NUREG/CR-7108.

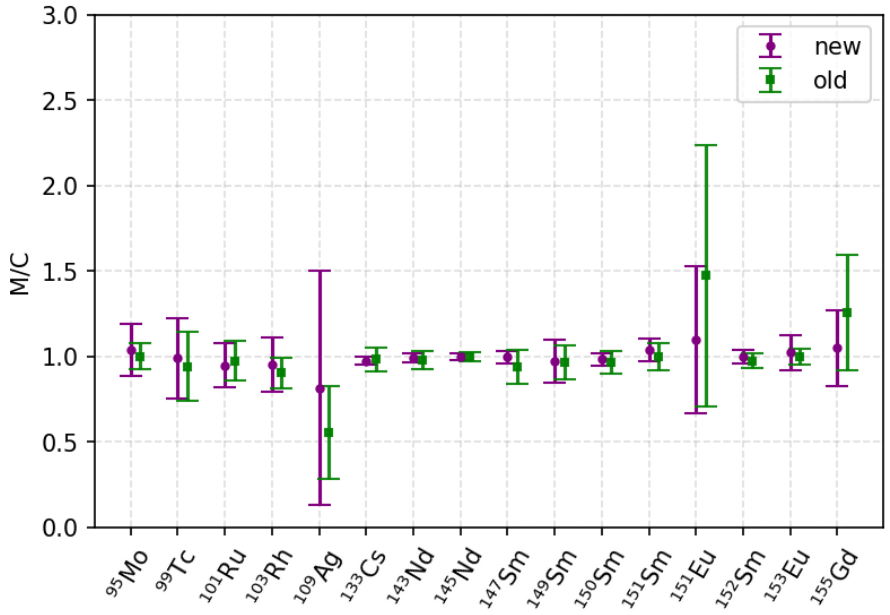


Figure 5-7 Bias and Bias Uncertainty for Fission Products Compared to Previous Data

6 BIAS AND BIAS UNCERTAINTY IN CALCULATED k_{eff}

The Sampler uncertainty quantification tool in SCALE 6.2.4 is used for the new and standardized implementation of the stochastic uncertainty sampling method described in Section 2.3 to determine the bias and bias uncertainty in calculated k_{eff} for the cask criticality model. Details on the Sampler implementation are presented in Appendix A.

6.1 k_{eff} Bias and Bias Uncertainty Calculated with SCALE 6.2.4/ ENDF/B-VII.1

Bias and bias uncertainty in calculated k_{eff} for the cask model as a function of the assembly average burnup are shown in Table 6-1. These values are obtained using a CSAS6 model of the cask with 252-group ENDF/B-VII.1 cross sections, the stochastic method for uncertainty sampling presented in section 2.3, and the bias and bias uncertainty in calculated actinides and fission product concentrations from Tables 5-3 and 5-4. Two sets of values are applied in the stochastic sampling method for the bias and bias uncertainty of burnup-dependent actinide concentrations in fuel materials of the CSAS6 model: one set for burnups ≤ 40 GWd/MTU, and another set for burnups greater than 40 GWd/MTU. A single set of values is used for fission product concentrations over the entire burnup range up to 80 GWd/MTU. The k_{eff} bias and bias uncertainty values shown in Table 6-1 correspond to 1,000 Sampler samples (i.e., CSAS6 calculations) for each assembly burnup. However, as presented in Appendix A.2, which discusses the convergence of the results as a function of the number of random samples used, similar bias and uncertainty values are obtained with 300 Sampler samples.

Table 6-1 Bias and Bias Uncertainty for k_{eff} of the Cask Model

Assembly burnup (GWd/MTU)	Initial enrichment (%)	k_{eff} bias	k_{eff} bias uncertainty	k_{eff} bias + k_{eff} bias uncertainty
5	2.13	0.0033	0.0165	0.0198
10	2.36	0.0032	0.0155	0.0187
18	3.03	0.0028	0.0142	0.0169
25	3.48	0.0023	0.0138	0.0161
30	3.98	0.0020	0.0136	0.0159
40	4.70	0.0017	0.0139	0.0157
45	5.06	0.0010	0.0152	0.0162
50	5.42	0.0011	0.0149	0.0160
55	5.79	0[a]	0.0187	0.0187
60	6.16	0	0.0184	0.0184
55	5.79	0	0.0182	0.0182
70	6.16	0	0.0180	0.0180

[a] A bias that reduces the calculated value of k_{eff} is not considered [2] and is set to 0; actual values are -0.0005, -0.0007, -0.0006, and -0.0003 for burnups of 55, 60, 65, and 70 GWd/MTU, respectively.

The variations of k_{eff} bias, k_{eff} bias uncertainty, and the sum of k_{eff} bias and bias uncertainty as a function of the assembly average burnup are illustrated in Figures 6-1 through 6-3.

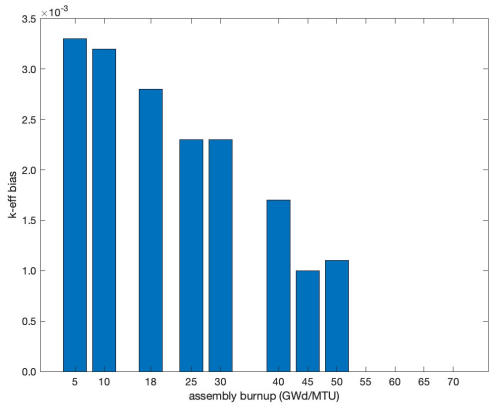


Figure 6-1 k_{eff} Bias for the Cask Model

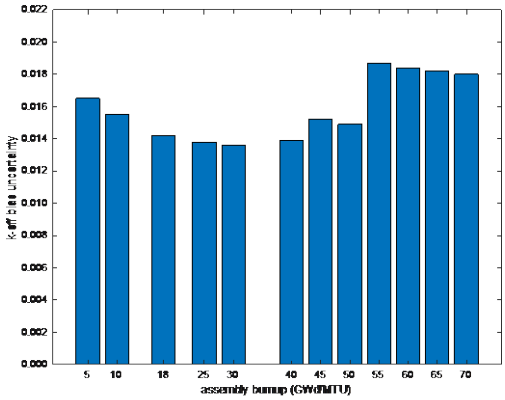


Figure 6-2 k_{eff} Bias Uncertainty for the Cask Model

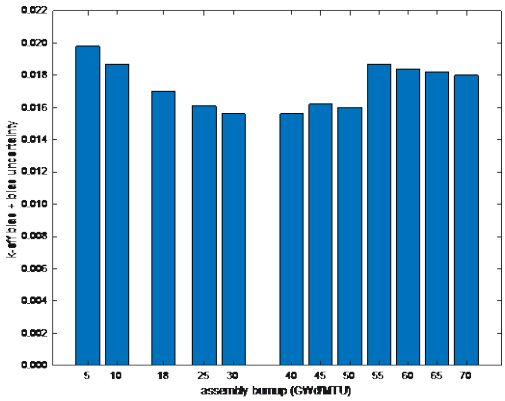


Figure 6-3 k_{eff} Bias + Bias Uncertainty for the Cask Model

The results presented in Table 6-1 and illustrated in Figures 6.1 through 6.3 indicate the following:

- Bias varies in the range of 0.0033–0.0010, and it decreases with increasing burnup for burnups ranging from 5 to 50 GWd/MTU; bias is positive and conservatively set to 0 for burnups ranging from 55 to 70 GWd/MTU.

- Bias uncertainty varies in the range of 0.0136–0.0187 over burnups ranging from 5 to 70 GWd/MTU; the largest values plateau at around 0.018-0.019 for burnups in the 55–70 GWd/MTU range.

- Bias + bias uncertainty values and variation with burnup are driven by the bias uncertainty values, which are much greater than the bias values.

- Bias + bias uncertainty values vary in the 0.0157 to 0.0198 range for burnups ranging from 5 to 70 GWd/MTU; the largest value occurs at 5 GWd/MTU, where the bias is maximum.

6.2 k_{eff} Bias Uncertainty Comparison with NUREG/CR-7108 and ISG-8, Rev 3

Comparison of k_{eff} bias uncertainty as a function of assembly-average burnup, which is based on data shown in Table 6-1 and corresponding values available in ISG-8 Rev.3 [1], is illustrated in Figure 6-4. These ISG-8 Rev.3 values are identical with those reported in NUREG/CR-7108 [4] for burnups $\leq$ 45 GWd/MTU for the same cask analysis model; for burnups above 45 GWd/MTU, the values included are those calculated for a spent fuel pool analysis model. The NUREG-7108 does not report data for fuel compositions including both actinide and fission product burnup credit nuclides beyond 45 GWd/MTU. When compared to the previous values, the bias uncertainty estimated in this report is significantly smaller at high burnups. The absolute difference in bias uncertainty between the current and previous values is approximately 0.002 for burnups ranging up to 40 GWd/MTU. This difference increases above 0.002 with increasing burnup for burnups greater than 40 GWd/MTU: it varies from 0.005 at 45 GWd/MTU to 0.012 at 60 GWd/MTU.

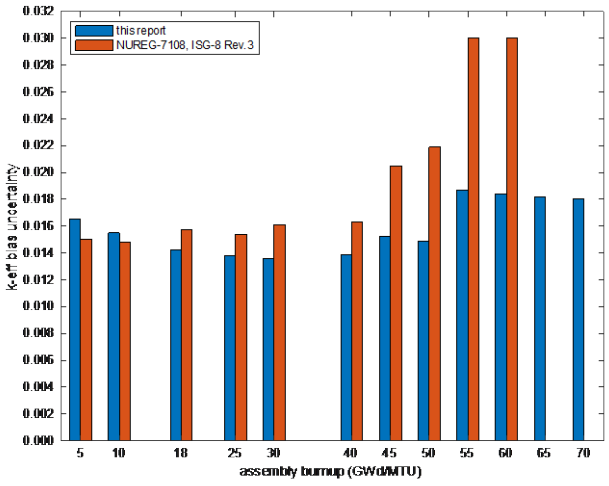


Figure 6-4 Comparison of k_{eff} Bias Uncertainty for the Cask Model

7 CONCLUSIONS

The studies documented in this report build upon and complement the approach presented in NUREG/CR-7108 to estimate k_{eff} bias and bias uncertainty in burnup credit criticality safety analyses that are associated with calculated nuclide concentrations of spent nuclear fuel. This approach includes three main steps: (1) develop a model for the bias and bias uncertainty in calculated nuclide concentrations for a given set of nuclides and a burnup range of interest based on a comparison of measured and corresponding calculated data for these nuclides; (2) based on the bias and bias uncertainty in the calculated nuclide concentrations, sample the probability distribution of the nuclide concentrations in the fuel compositions of the criticality model within the range of their estimated uncertainties; and (3) using the stochastically sampled fuel compositions within the criticality safety model, calculate the corresponding k_{eff}, and estimate the k_{eff} bias and bias uncertainty based on the resulting distribution of k_{eff} values.

The approach applied herein is based on a new and standardized implementation of the stochastic sampling method that was used previously. This new implementation takes advantage of the capabilities in the Sampler uncertainty quantification tool, which was introduced in the SCALE modeling and simulation suite in 2016. All results presented herein correspond to burnup credit criticality safety calculations that consider 28 nuclides—12 actinides and 16 fission products—in the spent fuel compositions of the criticality safety model. All analyses described in this report were performed using computational tools in the 6.2.4 release of SCALE and ENDF/B-VII.1 cross sections. The analyses documented in NUREG/CR-7108 were performed using SCALE 6.1.3, with nuclear cross sections based on ENDF/B-VII.0.

The loading curve developed for the GBC-32 cask model in NUREG/CR-7108 was reassessed to verify its applicability when using the criticality calculation sequences in SCALE 6.2.4 with ENDF/B-VII.1 cross sections. It was observed that the use of this loading curve with ENDF/B-VII.1 cross sections led to k_{eff} values slightly lower than the 0.94 target value, by approximately 0.0024 to 0.0043. To ensure that the reference models used to determine k_{eff} bias and bias uncertainty in this study met the k_{eff} target value, a new loading curve was generated. The updated loading curve was derived using a new methodology that employs the ORIGAMI fast depletion computational tool in SCALE 6.2.4.

The validation basis for estimating the bias and bias uncertainty in calculated concentrations of burnup credit nuclides was significantly improved in this work. It includes measurement data for 129 spent fuel samples and extends the previous burnup range of up to 60 GWd/MTU in NUREG-7108 to 80 GWd/MTU herein. The current validation basis includes high-burnup measurement data from international experimental programs and from recent RCA measurements conducted at ORNL. These recent RCA measurements were performed under an ongoing experimental program jointly funded by the NRC and DOE.

The significant improvement of the validation basis, along with the increase in the number of measurements, helped fill data gaps for burnup credit nuclides, especially at high burnups. As a result, the uncertainty in the calculated nuclide concentration for most of these nuclides was reduced. For example, the relative uncertainty for the major actinide ^{239}Pu was reduced for burnups greater than 40 GWd/MTU by approximately half, from 7.3% to 4.5%, partly due to increase in the number of measurements and partly due to change of cross sections. This has practical impact on criticality safety with burnup credit for modern discharged fuel characterized by higher burnups.

The improvement in the validation basis led to an overall decrease in the k_{eff} bias uncertainty for the considered cask criticality model compared to the corresponding values shown in ISG-8 Rev.3. The k_{eff} bias uncertainty in the current report varies in the 0.0136 to 0.0187 range over burnups from 5 to 70 GWd/MTU. The bias uncertainty for burnups less than 40 GWd/MTU compared to the previous values is approximately 0.002 smaller. The reduction in k_{eff} bias uncertainty for burnups greater than 40 GWd/MTU varies from 0.005 at 45 GWd/MTU to 0.012 at 60 GWd/MTU compared to ISG-8 Rev.3. Part of this decrease resulted from a reduction in the bias uncertainty for the calculated nuclide concentrations. This reduction is a result of an increased number of measurements for burnup credit nuclides, the effect of nuclear data cross sections, and the addition of high-quality measurement data for impactful actinides and fission products.

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APPENDIX A
STOCHASTIC UNCERTAINTY SAMPLING METHOD – IMPLEMENTATION USING SAMPLER

A.1 Sampler Template Input for k_{eff} Stochastic Uncertainty Sampling

```
=sampler

read parameters
 n_samples=1000
 library="v7-252"
 perturb_xs=no
 perturb_geometry=yes
 run_cases=yes
 force_run=no
end parameters

read case [PWR_cask]

sequence=csas6
gbc-32
v7-252
read comp
'burnup=  55.0 gwd/mtu; enrichment=5.79 %
' #{variable id}placeholder for each burnup credit
' nuclide in each of the 18 fuel materials
'node[01]
 o-16     101  0  4.68370E-02 293.0 end
 u-234    101  0  #{u2341}    293.0 end
 u-235    101  0  #{u2351}    293.0 end
 u-236    101  0  #{u2361}    293.0 end
 u-238    101  0  #{u2381}    293.0 end
 np-237   101  0  #{np2371}   293.0 end
 pu-238   101  0  #{pu2381}   293.0 end
 pu-239   101  0  #{pu2391}   293.0 end
 pu-240   101  0  #{pu2401}   293.0 end
 pu-241   101  0  #{pu2411}   293.0 end
 pu-242   101  0  #{pu2421}   293.0 end
 am-241   101  0  #{am2411}   293.0 end
 am-243   101  0  #{am2431}   293.0 end
 mo-95    101  0  #{mo951}    293.0 end
 tc-99    101  0  #{tc991}    293.0 end
 ru-101   101  0  #{ru1011}   293.0 end
 rh-103   101  0  #{rh1031}   293.0 end
 ag-109   101  0  #{ag1091}   293.0 end
 cs-133   101  0  #{cs1331}   293.0 end
 nd-143   101  0  #{nd1431}   293.0 end
 nd-145   101  0  #{nd1451}   293.0 end
 sm-147   101  0  #{sm1471}   293.0 end
 sm-149   101  0  #{sm1491}   293.0 end
 sm-150   101  0  #{sm1501}   293.0 end
 sm-151   101  0  #{sm1511}   293.0 end
 sm-152   101  0  #{sm1521}   293.0 end
 eu-151   101  0  #{eu1511}   293.0 end
 eu-153   101  0  #{eu1531}   293.0 end
```

```
 gd-155   101  0  #{gd1551}   293.0 end

…….
'node[02]
 o-16     102  0  4.68350E-02 293.0 end
 u-234    102  0  #{u2342}    293.0 end
 u-235    102  0  #{u2352}    293.0 end
 u-236    102  0  #{u2362}    293.0 end
 u-238    102  0  #{u2382}    293.0 end
 np-237   102  0  #{np2372}   293.0 end
 pu-238   102  0  #{pu2382}   293.0 end
 pu-239   102  0  #{pu2392}   293.0 end
 pu-240   102  0  #{pu2402}   293.0 end
 pu-241   102  0  #{pu2412}   293.0 end
 pu-242   102  0  #{pu2422}   293.0 end
 am-241   102  0  #{am2412}   293.0 end
 am-243   102  0  #{am2432}   293.0 end
 mo-95    102  0  #{mo952}    293.0 end
 tc-99    102  0  #{tc992}    293.0 end
 ru-101   102  0  #{ru1012}   293.0 end
 rh-103   102  0  #{rh1032}   293.0 end
 ag-109   102  0  #{ag1092}   293.0 end
 cs-133   102  0  #{cs1332}   293.0 end
 nd-143   102  0  #{nd1432}   293.0 end
 nd-145   102  0  #{nd1452}   293.0 end
 sm-147   102  0  #{sm1472}   293.0 end
 sm-149   102  0  #{sm1492}   293.0 end
 sm-150   102  0  #{sm1502}   293.0 end
 sm-151   102  0  #{sm1512}   293.0 end
 sm-152   102  0  #{sm1522}   293.0 end
 eu-151   102  0  #{eu1512}   293.0 end
 eu-153   102  0  #{eu1532}   293.0 end
 gd-155   102  0  #{gd1552}   293.0 end
…….
end sequence

' random numbers drawn from standard normal distribution,
' one random number per burnup credit nuclide, per perturbed case
read variable [R_u235]
 distribution=normal
 stddev=1
 value=0
end variable
…….
read variable [R_gd155]
 distribution=normal
 stddev=1
 value=0
end variable
…….

' variable for the concentrations C^m_{i,Bu} defined in Eq.(5)
' if value is less than 0, is set to 0

read variable [u2351]
  distribution=expression
```

```
  expression="if((6.01776E-04+R_u235*2.33209E-05<0),0,(6.01776E-
04+R_u235*2.33209E-05))"
end variable
read variable [u2352]
  distribution=expression
  expression="if((3.85104E-04+R_u235*3.15434E-05<0),0,(3.85104E-
04+R_u235*3.15434E-05))"
end variable
……..
end case

' desired calculated response
read response[keff]
type = grep
regexp = ":kenovi.keff:"
end response

end
```

A.2 Convergence of the keff Bias Uncertainty Estimate

The stochastic sampling approach summarized in Section 2.3 requires many random samples, or perturbed k_{eff} calculations, to ensure that the values of the mean and standard deviation for the resulting k_{eff} distribution are meaningful and converge to their best-estimate values. Figure A-1 is a histogram illustrating, for the 55 GWd/MTU burnup, an example of k_{eff} distribution corresponding to 1,000 perturbed cases. As indicated by a Shapiro-Wilk normality test at the 0.05 level, the illustrated k_{eff} values are drawn from a normal distribution.

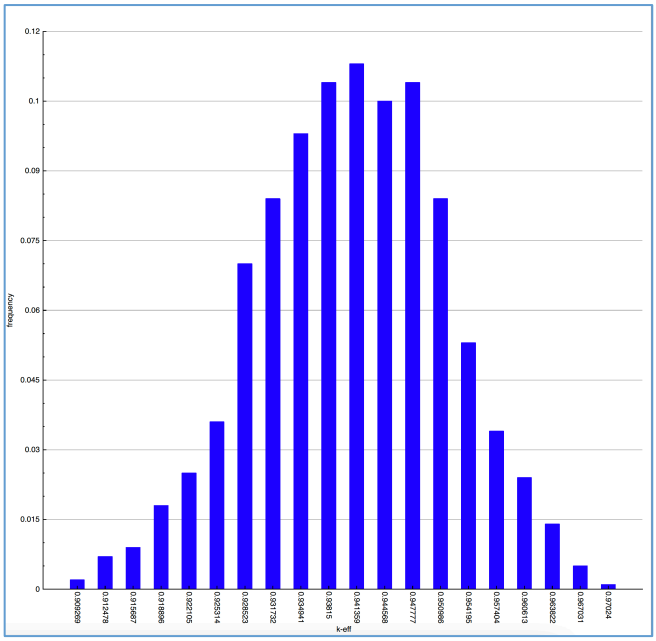

Figure A-1 Histogram of k_{eff} Perturbed Values for 55 GWd/MTU Burnup

The stochastic sampling method is inherently computationally intensive. Studying its convergence and understanding how convergence varies according to the number of perturbed cases is a prerequisite for making the application of the method practical. Convergence is considered reached when the difference in k_{eff} bias uncertainty values corresponding to N and N+1 perturbed cases is smaller than a target value, which in this case is set 0.0001. The minimum number of perturbed cases sufficient to reach convergence can indicate how the total computing time can be minimized.

Figures A-2 and A-3 present variations of k_{eff} bias and bias uncertainty as a function of the number of perturbed cases for burnup values of 10 and 45 GWd/MTU. Each figure illustrates (a) the mean k_{eff} calculated per Eq. (6) as a green line; (b) the k_{eff} bias calculated per Eq. (8) as the distance between the 0.94 reference value in blue and the mean k_{eff}, and (c) the k_{eff} bias uncertainty calculated per Eq. (9) as the distance between the green and red lines. Visual inspection of the plots in A2-2 and A2-3 indicates that k_{eff} convergence of bias and bias uncertainty is reached at around 200 perturbed cases. It also indicates that the number of perturbed cases required to reach convergence varies slightly with burnup.

Figure A-4 illustrates the variation of the k_{eff} bias uncertainty with the number of perturbed cases for burnups of 10, 30, and 50 GWd/MTU, indicating that the minimum number of perturbed cases to reach convergence can vary slightly with burnup. Per this convergence criterion, convergence is achieved for burnups of 10, 30, and 50 GWd/MTU at 180, 200, and 280 perturbed cases, respectively. The highest value of the minimum number of cases to reach convergence for a 70 GWd/MTU burnup is 280. Based on these observations, it can be concluded that 300 perturbed cases are sufficient to achieve convergence for this model.

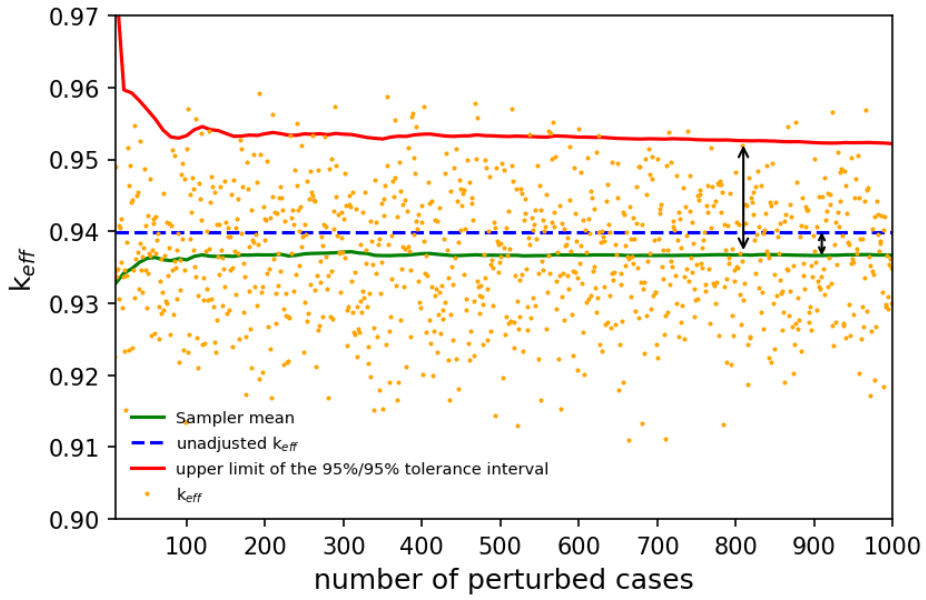

Figure A-2 k_{eff} Bias and Bias Uncertainty vs. Number of Perturbed Cases for 10 GWd/MTU Burnup

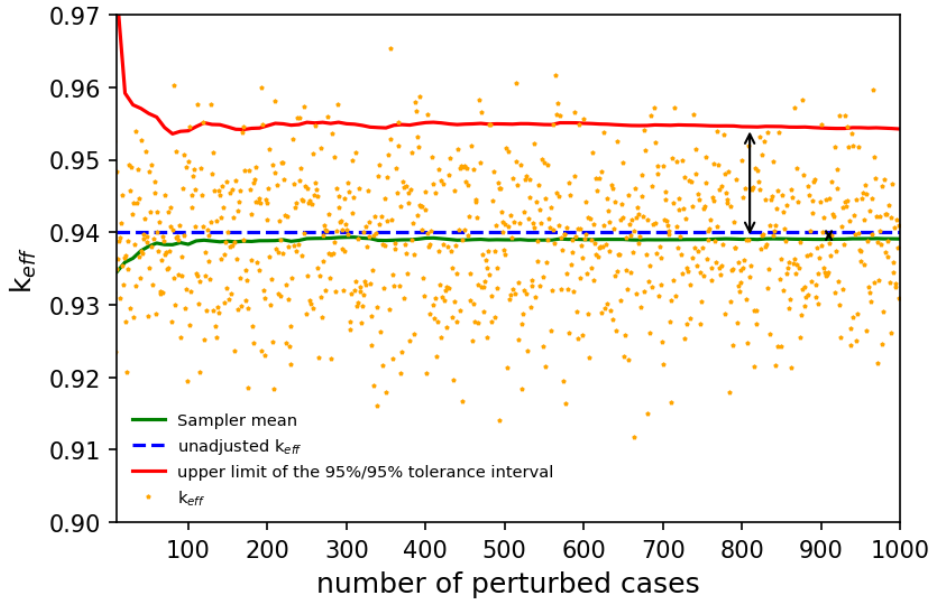

Figure A-3 k_{eff} Bias and Bias Uncertainty vs. Number of Perturbed Cases for 45 GWd/MTU Burnup

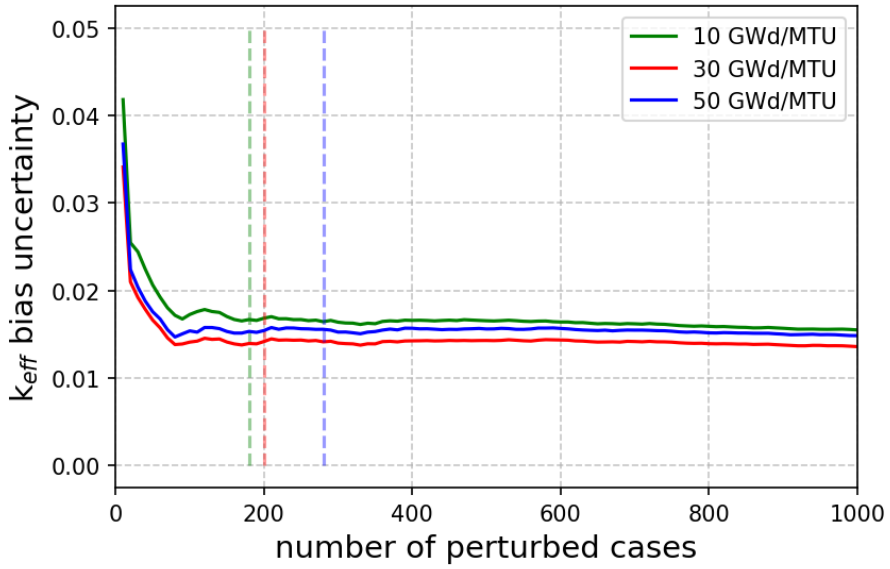

Figure A-4 k_{eff} Bias Uncertainty Estimate vs. Number of Perturbed Cases

For any assembly-average burnup under consideration, the minimum number of perturbed cases to achieve convergence is inherently dependent on the considered convergence criterion and the analyzed criticality model. The 300 number of converged cases determined in this work is considered a representative estimate of the order (few hundreds) of perturbed cases that would be necessary for other, different criticality models.

A.3 Verification of Stochastic Sampling Method Implementation

The new implementation of the stochastic sampling method was verified for consistency with the method as applied in NUREG/CR-7108 using the 30 GWd/MTU burnup as a test case. The same CSAS5 reference input model for this burnup, 238-group ENDF/B-VII.0 cross section library, and the same bias and bias uncertainty for burnup credit nuclide concentrations that were applied in NUREG/CR-7108 were used in the present work with the Sampler-based implementation of the method and SCALE 6.2.4. The difference between the old and the new CSAS5 input was the source definition (here, gen=1200, nsk=200, npg=100,000) to ensure that all CSAS5 convergence tests pass. All calculations were performed using CSAS5 and Sampler in SCALE 6.2.4.

Results for 500 Sampler "samples" (CSAS5 inputs) with stochastically sampled nuclide concentrations vs. results reported in NUREG/CR-7108 for the same burnup are compared in Table A-1. The results obtained with the new implementation and those reported previously are relatively consistent. The differences of 0.0002 in bias and 0.0028 in bias uncertainty between the current and previous values are small. These small differences could be the result of differences in the stochastic method implementation, or differences between SCALE 6.1.3 and SCALE 6.2.4 for the CSAS5 criticality sequence, including in the self-shielding method applied to the multigroup cross sections used with the Monte Carlo neutron transport solver KENO-V.a.

Table A-1 Comparison of Results Obtained with New and Previous Implementations of the Stochastic Sampling Method for Consistent Input at 30 GWd/MTU Burnup

Results for current method implementation			Results reported in NUREG/CR-7108 [4]		
k_{eff} bias	k_{eff} bias uncertainty	k_{eff} bias + bias uncertainty	k_{eff} bias	k_{eff} bias uncertainty	k_{eff} bias + bias uncertainty
0.0029	0.0133	0.0162	0.0031	0.0161	0.0192

A.4 Comparison of Results Obtained with the New Implementation Using CSAS5 and CSAS6 with the Criticality Model

The k_{eff} bias and bias uncertainty calculated using the GBC-32 cask criticality model with the CSAS6 criticality sequence in SCALE 6.2.4 and the corresponding values obtained using the same criticality model represented with the CSAS5 criticality sequence in SCALE 6.2.4 are the same. This similarity is as expected; criticality calculations for the same model represented with either KENO-V.a or KENO-VI are expected to produce the same k_{eff} for that model within the statistical uncertainty associated with the calculation.

Table A-2 includes more detailed information relevant to the bias and bias uncertainty values presented in Table 6-1. These data correspond to the use of CSAS6 for the cask criticality model. In addition to data in Table 6-1, Table A-2 includes:

- k_{eff} and the associated standard deviation for the reference criticality model; in this model, calculated burnup credit nuclide concentrations are not adjusted in any way.

- k_{eff} and associated standard deviation for the "unperturbed" case in Sampler; calculated nuclide concentrations are modified to their best estimate values to account for the bias in nuclide concentrations $C_{i,Bu}\ \overline{X}_{i,Bu}$, where the terms are as defined for Eq. (5).

- k_{eff} mean and associated standard deviation from the distribution of k_{eff} values obtained with Sampler for the 1,000 instances of the criticality model with stochastically sampled concentrations for the burnup credit nuclides.

Table A-2 presents the same type of data as shown in Table A-1 for burnups up to 50 GWd/MTU, with the difference being that the criticality sequence used is CSAS5. The bias and bias uncertainty values obtained using CSAS5 are the same as those obtained using CSAS6, for the considered model. This is of consequence for computationally expensive calculations with Sampler. The use of a CSAS5 model would result in a significantly smaller total Sampler computing time than that applicable to CSAS6. However, the use of CSAS6 would provide capabilities of modeling more heterogeneous, complex geometries for which CSAS5 may have limited capabilities, depending on the actual configuration of interest.

Table A-2 Detailed k_{eff} Results Obtained with the CSAS6 Criticality Model

Assembly burnup (GWd/MTU)	Initial enrichment (%)	k_{eff} nominal		k_{eff} data from Sampler				k_{eff} bias	k_{eff} bias uncertainty	k_{eff} bias + k_{eff} bias uncertainty
				unperturbed		mean				
		value	stdev	value	stdev	value	stdev			
5	2.13	0.94002	0.00008	0.93683	0.00007	0.93671	0.00957	0.0033	0.0165	0.0198
10	2.36	0.93990	0.00008	0.93661	0.00008	0.93672	0.00899	0.0032	0.0155	0.0187
18	3.03	0.94002	0.00007	0.93717	0.00007	0.93725	0.00820	0.0028	0.0142	0.0169
25	3.48	0.93993	0.00007	0.93777	0.00008	0.93763	0.00798	0.0023	0.0138	0.0161
30	3.98	0.93994	0.00006	0.93805	0.00007	0.93792	0.00788	0.0020	0.0136	0.0156
40	4.70	0.93989	0.00007	0.93824	0.00007	0.93908	0.00808	0.0017	0.0139	0.0157
45	5.06	0.94008	0.00007	0.93909	0.00006	0.93908	0.00878	0.0010	0.0152	0.0162
50	5.42	0.93986	0.00008	0.93876	0.00007	0.93877	0.00861	0.0011	0.0149	0.0160
55	5.79	0.94019	0.00007	0.94090	0.00007	0.94067	0.01083	-0.0005[a]	0.0187	0.0187
60	6.16	0.94007	0.00007	0.94090	0.00007	0.94074	0.01067	-0.0007	0.0184	0.0184
65	6.52	0.93989	0.00007	0.94055	0.00007	0.94048	0.01054	-0.0006	0.0182	0.0182
70	6.89	0.94006	0.00007	0.94064	0.00007	0.94041	0.01041	-0.0003	0.0180	0.0180

[a] A bias that reduces the calculated value of k_{eff} is not considered; set to 0 in Table 6-1.

Table A-3 Detailed k_{eff} Results Obtained with the CSAS5 Criticality Model

Assembly burnup (GWd/MTU)	Initial enrichment (%)	k_{eff} nominal		k_{eff} data from Sampler				k_{eff} bias	k_{eff} bias uncertainty	k_{eff} bias + k_{eff} bias uncertainty
				unperturbed		mean				
		value	stdev	value	stdev	value	stdev			
5	2.13	0.93971	0.00007	0.93654	0.00008	0.93642	0.00957	0.0033	0.0165	0.0198
10	2.36	0.93988	0.00006	0.93651	0.00007	0.93646	0.00897	0.0034	0.0155	0.0189
18	3.03	0.93969	0.00008	0.93727	0.00007	0.93704	0.00820	0.0026	0.0142	0.0168
25	3.48	0.93976	0.00008	0.93750	0.00009	0.93743	0.00797	0.0023	0.0138	0.0161
30	3.98	0.93986	0.00007	0.93783	0.00008	0.93771	0.00788	0.0022	0.0136	0.0158
40	4.70	0.93967	0.00008	0.93815	0.00009	0.93797	0.00808	0.0019	0.0139	0.0145
45	5.06	0.93986	0.00007	0.93892	0.00008	0.93887	0.00878	0.0010	0.0152	0.0162
50	5.42	0.93970	0.00009	0.93866	0.00008	0.93857	0.00861	0.0011	0.0149	0.0160
55	5.79	0.93992	0.00008	0.94069	0.00007	0.94046	0.01083	-0.0005[a]	0.0187	0.0187
60	6.16	0.94006	0.00008	0.94060	0.00007	0.94053	0.01067	-0.0005	0.0184	0.0184

[a] A bias that reduces the calculated value of k_{eff} is not considered; set to 0.

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11. ABSTRACT (200 words or less)

Criticality safety evaluations accounting for burnup credit require validation of the computational tools and associated nuclear data used to perform the fuel depletion and criticality calculations. The NUREG/CR-7108 report published in 2012 documents an approach for establishing the bias and bias uncertainty in the calculated neutron multiplication constant (k_{eff}) that is associated to calculated spent nuclear fuel nuclide concentrations, using as validation basis radiochemical assay (RCA) measurement data for nuclide concentrations. Studies documented in the current report build upon the NUREG/CR 7108 approach. The bias and bias uncertainty in calculated k_{eff} for a representative analysis model that includes pressurized water reactor (PWR) spent fuel is determined by applying a stochastic sampling method. The validation basis presented herein is extended compared to that in NUREG/CR-7108 to fill data gaps at high burnups, with practical impact on burnup credit for modern discharged fuel. This extended validation basis includes recent RCA measurements performed at Oak Ridge National Laboratory under an experimental program jointly funded by the Nuclear Regulatory Commission and the Department of Energy. The improvement in the validation basis led to an overall decrease in the keff bias uncertainty for the considered analysis model compared to the corresponding values in ISG-8 Rev.3.

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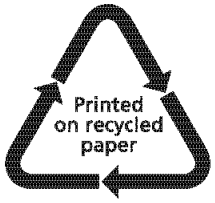

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