

## Burnup Credit Methodology

Spent Nuclear Fuel Transportation Applications

## **Burnup Credit Methodology**

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### **REPORT SUMMARY**

This report describes a practical methodology for actinide-only and fission product burnup credit in concert with a methodology to validate the isotopic and reactivity calculations. The methodology supports initial enrichments up to 5.0 wt% <sup>235</sup>U and burnup beyond 50 gigawattdays per metric ton of uranium (GWd/MTU). The validation methodologies are all based upon standard methodologies, including extensions beyond traditional radiochemistry assays (RCAs) for isotopic concentrations and critical experiment benchmarking. The classical Propagation of Uncertainties methodology is used to transform documented uncertainties associated with fission-product isotope production and cross sections into equivalent reactivity bias factors. Confirmation of reactivity bias factors for fission products is provided by available data, including commercial reactor criticals, that validate the isotopic production and cross sections together and/or separate fission-product RCAs and critical experiments.

#### Background

*Burnup credit* refers to taking credit for the burnup of nuclear fuel in the performance of criticality safety analyses. *Actinide-only* burnup credit refers to a methodology that considers only the two major actinides present in spent fuel: uranium and plutonium. *Fission product* burnup credit considers a number of fission products and minor actinides. *Full* burnup credit refers to a combination of actinide-only and fission product burnup credits.

Burnup credit has been sought for the transportation, storage, and disposal of spent commercial nuclear fuel for over two decades. Progress has included the issuance of the first version of U.S. Nuclear Regulatory Commission (NRC) Interim Staff Guidance 8 in 1999. The latest version, Revision 2, has endorsed actinide-only burnup credit and was issued in 2002. Revision 3 is expected in 2011. Experimental data necessary for validation of the isotopic compositions and the nuclear cross sections of fission products have not been deemed to be adequate thus far, and approval of full burnup credit, including both actinides and fission products, has been subsequently delayed.

#### **Objectives**

• To develop a methodology that addresses actinide-only and fission product burnup credit in concert with a methodology to validate the isotopic and reactivity calculations

#### Approach

Actinide-only burnup credit calculations are divided into isotopic calculations and reactivity calculations, and a validation for these two components is performed using published data that can be used to determine the accuracy of the calculations. Fission product burnup credit builds upon this by adding isotopic calculations and reactivity calculations for fission products, and

validates the fission product calculations by evaluating the uncertainties of the physical processes that produce the fission product isotopic contents and the uncertainties of the cross sections of isotopes that affect the spent fuel reactivity.

Actinide-only burnup credit is validated by the application of RCAs to validate the isotopic calculations and to develop an actinide-only isotopic bias correction factor ( $\Delta k_{I-Act}$ ). The validation of reactivity calculations is achieved through a traditional analysis of critical experiments to determine the actinide-only cross-section bias correction factor ( $\Delta k_{C-Act}$ ). The overall bias correction factor is the statistical sum of the independent isotopic and cross-section bias factors.

Fission product cross-section validation is obtained by means of a "direct perturbation" or "indirect perturbation" study of the reactivity effects of cross-section uncertainty, resulting in the fission product cross-section bias correction factor ( $\Delta k_{C-FP}$ ). The overall bias correction factor is the statistical sum of the isotopic and cross-section bias factors, with the exception of the neutron capture term of the isotopic validation, which is dependent upon both isotopic and cross-section data and is summed algebraically.

#### Results

A validated methodology is described. The overall bias correction factors for actinide-only and full (actinide plus fission product) burnup credit for PWR applications are  $\Delta k_{Act} = 0.014$  and  $\Delta k_{Act+FP} = 0.037$ , based on a 21 PWR waste package design that employed boron/stainless steel absorber. Confirmatory data from a variety of sources are also provided. These sources indicate that the methodology and its validation are conservative by 0.02  $\Delta k$  or more.

#### **EPRI** Perspective

Full burnup credit including fission products may be gaining regulatory acceptance as additional data are obtained and calculational methodologies are refined. Burnup credit, especially including fission products, makes allowance for the much lower reactivity of spent light water reactor fuel by means of calculations. Greater capacities are possible in large dual-purpose casks, increasing from 21 PWR assemblies for early designs to 37 PWR assemblies in today's casks that take credit for burnup. Higher enrichments in fresh fuel apply pressure on today's cask certifications, and full burnup credit can address this without requiring more difficult and expensive hardware solutions.

#### Keywords

Actinide-only Burnup credit Fission product Full burnup credit Isotopic validation Reactivity validation

### ABSTRACT

Burnup credit has been sought for transportation, storage, and disposal of spent commercial nuclear fuel for over two decades. Progress has included the issuance of U.S. NRC Interim Staff Guidance – 8, Revision 2, which describes actinide-only burnup credit. Experimental data and critical experiments necessary for validation of the isotopic compositions and the nuclear cross sections of spent fuel fission products have not been deemed to be adequate thus far, and approval of burnup credit including fission products has been delayed. Recently, a proprietary form of burnup credit including fission products was approved for the Holtec MPC-32 storage/transport canister system. The Holtec approach is similar to methodologies used for the licensing of burnup credit for spent fuel pools.

A consistent methodology and validation that makes maximum use of existing data has been developed. This methodology divides the validation into two parts: actinide-only burnup credit that is validated directly by traditional experimental data, and fission product burnup credit that is an add-on to actinide-only burnup credit and is validated by analyses of the uncertainties of physical processes (sensitivity analyses). This methodology diverges from the methods used for reactor and spent fuel pool analyses, which traditionally rely upon validation of "integral" experiments, as described in the Burnup Credit Standard [ANSI/ANS-8.27].

Actinide-only burnup credit is validated by the application of radiochemistry assays (RCAs) to validate the isotopic calculations and develop an actinide-only isotopic bias correction factor ( $\Delta k_{I-Act}$ ). The validation of reactivity calculations is achieved through the analysis of critical experiments to determine the actinide-only cross-section bias correction factor ( $\Delta k_{C-Act}$ ). The overall bias correction factor is the statistical sum of the independent isotopic and cross-section bias factors.

Radiochemistry assay data and critical experiments that are currently available are sufficient to validate actinides, but fission product data are less comprehensive. Fission product isotopic validation is obtained, in this report, through analysis of the processes that produce fission products: fission yield, radioactive decay, and neutron capture. This approach is an application of the classical "propagation of errors" or "propagation of uncertainties" methodology. The individual  $\Delta k$  factors are summed to produce an overall fission product isotopic bias correction factor ( $\Delta k_{I-FP}$ ).

Fission product cross-section validation is obtained by means of a "direct perturbation" or "indirect perturbation (available in TSUNAMI [SCALE 6.0])" study of the reactivity effects of cross-section uncertainty, resulting in the fission product cross section bias correction factor ( $\Delta k_{C-FP}$ ). The overall bias correction factor is the statistical sum of the isotopic and cross-section

bias factors, with the exception of the neutron capture term of the isotopic validation, which is dependent upon both isotopic and cross-section data and must be summed algebraically.

The general form for the reactivity of a full burnup credit system is:

 $k_{eff} = k_{eff,calculated} + SQRT(\Delta k_{I}^{2} + \Delta k_{C}^{2} + \Delta k_{C$ 

For PWRs, evaluation of the five bias correction factors was performed with the results as follows:

$$\begin{split} \Delta k_{I\text{-Act}} &= 0.011 \text{ [Triton validation, typical, SCALE 6.0]} \\ \Delta k_{C\text{-Act}} &= 0.008 \text{ [NUREG/CR-6979]} \\ \Delta k_{I\text{-FP}} &= 0.0046 \text{ [This report]} \\ \Delta k_{I\text{-FP(capture)}} &= 0.0058 \text{ [Wells 2006]} \\ \Delta k_{C\text{-FP}} &= 0.0170 \text{ [Wells 2005]} \end{split}$$

The overall PWR bias correction factors for actinide-only and full, actinide plus fission product, burnup credit are thus  $\Delta k_{Act} = 0.014$  and  $\Delta k_{Act+FP} = 0.037$ , respectively, and are added to  $k_{eff,calculated}$ .

By comparison, the bias and uncertainty from the Commercial Reactor Critical state points is 0.0143, which is about half of the  $\Delta k_{Act+FP}$  value, so the methodology described in this Topical Report is conservative by 2  $\frac{1}{2}$  percent  $\Delta k$ . Thus the methodology presented in this report is conservative.

The methodology also provides a means for applying recent fission product data that are more suited to a confirmatory role than a means of calculating FP bias correction factors. Fission product critical experiments [JAEA 2009] can be used to confirm the calculated  $\Delta k_{C-FP}$ . For these critical experiments, <sup>133</sup>Cs, <sup>103</sup>Rh, <sup>149</sup>Sm and <sup>151</sup>Eu, Calculated/Experimental ratios confirm the uncertainties used in the Direct Perturbation calculations. Similarly, fission product RCA data from reactors such as Gravelines and Bugey can be used to confirm the calculated  $\Delta k_{I-FP}$  for, <sup>145</sup>Nd, <sup>133</sup>Cs, and <sup>241</sup>Am.

Enhancements of the fission product isotopic and cross-section validations are foreseeable based upon future versions of the SCALE code system and MCNP. In particular, the TRITON code sequence of SCALE 6.0 allows more precise, two-dimensional representations of a fuel assembly for the calculation of the neutron spectrum and the isotopic contents of a fuel sample.

Parameters that define the irradiation history (e.g., specific power, temperature of moderator and fuel, axial burnup profile) of spent fuel are selected based upon studies that investigate the behavior of each parameter and their effect upon the reactivity of the discharged fuel. Together, these approaches produce conservative burnup credit  $k_{eff}$  values with a complete validation of all actinides and fission products. The methodology is applicable to storage, transport, and eventual disposal of spent fuel.

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## **1** INTRODUCTION

"Burnup credit" refers to taking credit for the burnup of nuclear fuel in the performance of criticality safety analyses. Historically, criticality safety analyses for transport of spent nuclear fuel have assumed the fuel to be unirradiated (i.e., "fresh" fuel). Considerable improvement in cask capacity can be realized if credit for the depletion of the fresh fuel can be credited ("burnup credit"). The methodology used to perform criticality analyses using burnup credit, and the validation of the calculations, has not been fully resolved primarily due to validation issues. Also, the ability to rely on reactor records in establishing fuel burnup rather than on an in-pool measurement, as presently prescribed in NRC guidance, is an important implementation issue.

#### 1.1 Regulatory Requirements

In 1999, the U.S. Nuclear Regulatory Commission (NRC) Spent Fuel Project Office issued Interim Staff Guidance – 8 (ISG-8) with recommendations for the use of burnup credit in storage and transportation of pressurized water reactor (PWR) spent fuel. On September 27, 2002, the NRC issued ISG-8, Revision 2 "*Burnup Credit in the Criticality Safety Analyses of PWR Spent Fuel in Transport and Storage Casks*". The ISG is a document meant to provide guidance for NRC staff regarding acceptable methods of analysis of various issues, but ISGs do not preclude other methodologies that may also be shown to be acceptable.

ISG-8, Revision 2 provides for burnup credit based upon only on the two major actinides (uranium and plutonium) present in spent UO<sub>2</sub> fuel. ISG-8 Rev. 2 requires that validation be performed for all actinide isotopes that are credited in the criticality calculations. ISG-8 Rev. 2 is limited to UO<sub>2</sub> fuel with initial enrichments up to 5.0 wt% U-235 and burnup up to 50 GWd/MTU. Calculations of isotopics for spent fuel are meant to be conservative, employing conservative axial burnup profiles and conservative parameters describing the assembly irradiation history, including the presence of burnable absorbers in the assembly. The calculations of reactivity must also be conservative, based upon validation of the cross sections that are used.

ISG-8 Rev. 2 also includes a measurement of burnup to confirm the assigned burnup from reactor reactors. ISG-8 Rev. 2 allows the calculation of "uncredited margin", i.e., a lower reactivity based upon other knowledge of burnup. An example is the calculation employing additional fission products that are not validated, resulting in a lower, but not validated  $k_{eff}$ .

#### 1.2 Current Status of Burnup Credit

Burnup credit for spent fuel pools has been a reality for many years, based in part upon NRC staff guidance [Kopp 1998]. The basic approach of spent fuel pool burnup credit calculations is to use isotopic concentrations computed in core follow (reactor physics) calculations and a computer code such as KENO or MCNP for calculating the reactivity of the spent fuel rack. A proprietary methodology developed by Holtec [Holtec 2006] extends this approach to spent fuel storage and transport canisters.

#### 1.3 Burnup Credit – US Industry Goals

A set of goals for burnup credit were described in [EPRI 2002]. A practical burnup credit methodology must be focused on achievable goals that add value to the use of burnup credit.

The methodology described in this report meets all of the industry goals for burnup, as described in Appendix A. Improvements to the technical means for validation of burnup credit isotopics and fission product cross sections may be forthcoming based upon additional data and enhancements of the sensitivity tools of SCALE. Modifications to the methodology itself are at the discretion of the user.

#### 1.4 Burnup Credit Terminology

Actinide-Only burnup credit refers to calculations employing only uranium (<sup>234</sup>U, <sup>235</sup>U, <sup>236</sup>U, and <sup>238</sup>U) and plutonium (<sup>238</sup>Pu, <sup>239</sup>Pu, <sup>240</sup>Pu, <sup>241</sup>Pu, and <sup>242</sup>Pu) isotopes. "Full" burnup credit refers to a combination of the uranium and plutonium isotopes evaluated in Actinide-Only burnup credit, plus a number of fission products and minor actinides. In this report, "Fission Product" or "FP" burnup credit is used to refer to 16 fission product isotopes and 4 minor actinide (neptunium and americium) isotopes. Different numbers of fission product isotopes have been used by different researchers in the burnup credit field, but the consensus is that the most important six are <sup>149</sup>Sm, <sup>103</sup>Rh, <sup>143</sup>Nd, <sup>151</sup>Sm, <sup>155</sup>Gd, and <sup>133</sup>Cs. <sup>241</sup>Am is the most significant minor actinide, contributing a reactivity worth (as a neutron absorber, this is beneficial to burnup credit) that is ranked third or fourth among the fission product isotopes. <sup>237</sup>Np is also significant, although not as strong in terms of reactivity worth as the six fission products and <sup>241</sup>Am.

#### 1.5 Burnup Credit Methodology

In this report, the flow of calculations involved in burnup credit calculations and their validation is described in Section 2. The validation of isotopic calculations for Actinide-Only and Actinide-Only plus Fission Product (FP) burnup credit is described in Section 3, noting that the validation of Actinide-Only burnup credit has matured and is therefore not discussed in as much detail as Fission Product burnup credit. The validation of cross sections for reactivity calculations is described in Section 4, again with more detail provided for FP calculations.

Confirmatory data supporting FP calculations are discussed in Section 5, including confirmation of both cross section and isotopic calculations. Confirmation is desirable because the validation of fission product calculations relies upon analyses of the uncertainties of isotopic and reactivity calculations via sensitivity studies. Section 6 discusses the application of burnup credit to the loading of burnup credit casks for storage and/or transport. Section 7 summarizes the burnup credit methodology and validation. Section 8 provides references.

Appendix A compares the burnup credit methodology of this report to industry goals for burnup credit.

## **2** CALCULATION PROCESS

The process of performing burnup credit calculations begins with the specification of fresh fuel design and the core support structure (e.g., canister basket). Once the fuel and core structure are known, information specific to burnup credit calculations is needed:

- Isotopic depletion parameters (e.g., specific power, temperature of moderator and fuel, axial burnup profile)
- Isotopic contents of the spent fuel (e.g., output from SAS2H [SCALE 5.1] or TRITON [SCALE 6.0])
- Reactivity calculations using nuclear cross section data (e.g., MCNP [Breismeister 1997] or KENO [SCALE 6.0])

#### **Actinide-Only Burnup Credit**

- Validation of actinide isotopics to provide the isotopic bias and uncertainty,  $\Delta k_{I-Act}$ , via radiochemistry assay (RCA) experiments
- Validation of cross sections to provide the cross section bias  $\Delta k_{C-Act}$ , via critical experiments

#### **Fission-Product Burnup Credit**

- Validation of fission product and minor actinide isotopics to provide the isotopic bias,  $\Delta k_{I-FP}$ , via sensitivity studies, with confirmatory data
- Validation of cross sections to provide the cross section bias  $\Delta k_{C-FP}$ , via sensitivity studies, with confirmatory data

#### **Confirmatory Calculations for Fission Product Burnup Credit**

- Calculation of  $\Delta k_{I-FP}$  plus  $\Delta k_{C-FP}$  via alternative methods, e.g., integral reactivity data such as reactor restart data
- Calculation of  $\Delta k_{I-FP}$  via additional RCA data that do not possess the desired range of experiments and/or statistical properties<sup>1</sup>
- Calculation of  $\Delta k_{C-FP}$  via additional critical experiment data that do not possess the desired range of experiments and/or statistical properties<sup>1</sup>

<sup>&</sup>lt;sup>1</sup> Limited datasets with few datapoints or insufficient range of enrichment and burnup values may not support normal distribution statistical methods.

#### Propagation of Errors (Uncertainties) and Confirmatory Data Application

The traditional approach to validation of neutron cross sections is via purpose-built benchmark critical experiments that are designed to closely represent the configurations of the desired criticality application. Similarly, the traditional approach to validation of isotopic contents of spent fuel is via RCA data obtained from actual fuel assemblies or pellets. Such experimental data incorporate the parameters of the burnup credit application explicitly, using enrichments and isotopic concentrations that are typical of spent fuel. When sufficient data of these types are available, the bias and uncertainty of the cross sections and isotopic depletion calculations are obtained through statistical evaluations from the datasets. Actinide-Only burnup credit can be validated with such data because sufficient fresh fuel and MOX critical experiments and sufficient RCA data for major actinides are available.

The validation of fission products and minor actinides is more difficult because few critical experiments are available for the strongest fission products and none are available at all for many less important, but still valuable, fission products. No critical experiments are available for the minor actinides of americium and neptunium. Critical experiments including actual spent fuel rods have been considered, but the cost and difficulty of working with such radioactive components in critical experiment facilities have discouraged such experiments.

Similarly, only a few RCA datapoints are available in the U.S. for fission products and minor actinides. These few RCA datapoints are not amenable to the statistics of normal distributions and are best suited for the purposes of confirmatory data. Confirmatory data may be considered to be any data that do not have the necessary statistical significance to act as a primary means of validation, but can act as check on validation obtained through other means.

The alternative means used in the burnup credit methodology of this report is to validate the cross sections of the fission products and minor actinides via computer simulations that determine the reactivity effects of uncertainties in the cross sections of each individual isotope; and the isotopic depletion calculation uncertainties are obtained by separately evaluating the uncertainties of the physical phenomena of fission yield, isotopic transmutation through neutron capture, and radioactive buildup and decay. The reactivity effects of the uncertainties of each part of the validation process are then combined with the uncertainties obtained for the Actinide-Only burnup credit validation through the "Propagation of Errors".

The different types of uncertainties in any experiment may be determined by an experimenter and then combined using the "Propagation of Errors" methodology. The different uncertainties are evaluated to determine if they are independent of each other, or if a dependence between two or more types of uncertainties exists. For burnup credit, the reactivity effects ( $\Delta k$ ) of fission product and minor actinide cross section uncertainties, fission yield uncertainties, and radioactive buildup and decay are independent. The reactivity effect of uncertainties in neutron capture and transmutation upon isotopic quantities is dependent upon the uncertainty of the neutron capture cross sections, so a dependency exists for this isotopic phenomenon. The "Propagation of Errors" methodology provides that independent types of uncertainty may be combined as the square root of the sum of the squares, while dependent types are combined arithmetically. Thus the total bias and uncertainty for full burnup credit is expressed as:

$$k_{eff} = k_{eff,calculated} + SQRT(\Delta k_{I}^{2} - Act + \Delta k_{C}^{2} - Act ) + SQRT(\Delta k_{I}^{2} - FP + \Delta k_{C}^{2} - FP ) + \Delta k_{I} - FP(capture)$$

where

$\Delta k_{I-Act}$	=	Actinide-Only Isotopic bias and uncertainty
$\Delta k_{C-Act}$	=	Actinide-Only Cross Section bias and uncertainty
$\Delta k_{I-FP}$	=	<u>F</u> ission <u>P</u> roduct and minor actinide bias and uncertainty due to <i>fission yield</i> and <i>radioactive decay</i> uncertainties on <u>I</u> sotopic content, such that:
		$\Delta k_{I-FP} = SQRT(\Delta k_{I}^{2} - FP(fission yield) + \Delta k_{I}^{2} - FP(radioactive decay))$
$\Delta k_{I-FP(capture)}$	=	<u>F</u> ission <u>P</u> roduct and minor actinide bias and uncertainty due to uncertainty in <u>I</u> sotopic content due to uncertainties in neutron <i>capture</i> cross sections
$\Delta k_{C-FP}$	=	<u>Fission</u> <u>Product and minor actinide bias and uncertainty due to uncertainty in neutron capture</u> <u>Cross</u> Sections

Note that in the treatment of fission product and minor actinide terms, the bias is due to the uncertainties in the term. A fuller description of this principle may be found in the TSUNAMI documentation of SCALE 6.0.

#### 2.1 Isotopic Depletion Parameters

Isotopic depletion parameters include at a minimum:

- 1. Burnup, typically expressed as a function of time as part of the irradiation history
- 2. Reactivity control mechanisms such as dissolved boron, control rods/blades
- 3. Presence of fixed or removable burnable absorbers
- 4. Coolant temperature and density
- 5. Fuel temperature power density
- 6. Cooling time (varies from discharge through thousands of years)

Parameter	Suggested Value
Fuel Temperature <sup>1</sup>	1000K
Cladding Temperature <sup>1</sup>	620K
Water Temperature <sup>1</sup>	600K
Water Density <sup>1</sup>	0.67 g/cm <sup>3</sup>
Moderator Boron Concentration <sup>1</sup>	650 ppmB
Specific Power <sup>2</sup>	30 MW/MTU
UO <sub>2</sub> Density	Calculated by User or 10.521

#### Table 2-1 Typical Isotopic Depletion Parameters

Notes: <sup>1</sup>Values from NUREG/CR-6761; <sup>2</sup>Typical, adjusted to match cycle length

The UO<sub>2</sub> density may be calculated from the active fuel length and the mass of UO<sub>2</sub> as smeared over the fuel rod inner diameter. These values are typically available for any particular fuel type, and may be especially useful for early fuel designs that did not obtain the 95 percent of theoretical density that is common today. The water temperature, and hence the density, vary along the fuel length so an average density of 0.67 g/cm<sup>3</sup> is suggested. Specific power may also be determined by the user to more accurately represent the characteristics of the specific fuel type. Specific power is often specified for each burnup cycle to reflect the different conditions of the core locations that the fuel assembly occupies in each cycle.

A generic cycle length or one that is specific to the fuel type or power plant may be used, with the specific power and cycle length combining to produce the desired burnup per cycle. Isotopic contents for fresh fuel (<sup>234</sup>U, <sup>235</sup>U, <sup>236</sup>U, <sup>238</sup>U as a function of enrichment) are found in [Bowman 1995, page 20].

#### 2.2 Isotopic Calculations

An isotopic depletion code, such as TRITON or CASMO for PWRs, or HELIOS for BWRs, is used to calculate the isotopic contents of spent fuel. The recent development of the twodimensional TRITON code sequence for SCALE offers improvements for complex PWR assemblies and BWR assemblies compared to what is attainable with the older one-dimensional SAS2H.

Isotopic contents of spent fuel are somewhat dependent upon the details of the irradiation history and are more strongly affected by the presence of strong neutron absorbers (control rods/blades) and dissolved boron. Removable burnable absorbers also have a significant effect upon the calculated isotopic contents. Results of isotopic calculations are thus improved by the addition of detailed information of the irradiation history, or conservatism may be introduced by the selection of depletion parameter values that harden the neutron spectrum and encourage the production of plutonium via the transmutation of <sup>238</sup>U. A study performed at ORNL [Wagner 2000] identified a number of conservative representations for the treatment of burnable absorbers

and also addressed the presence of control rods. Application of the ORNL irradiation models can insure a conservative result without becoming unrealistic. The ORNL irradiation models have been applied to burnup credit calculations [EPRI 2008a] and are adopted for this topical report since they provide a complete solution for burnable absorber calculations.

The axial burnup profile is implemented by performing isotopic calculations at a number of nodes, e.g., 25 nodes for a core-follow calculation for a PWR or 7 nodes for a conservative axial profile [Scaglione 2003]. The 7-node conservative profiles are abstractions of a database of actual profiles, created by first determining the mean burnup and standard deviation at each of three nodes at the top end of the fuel assembly and three nodes at the bottom of the fuel assembly, and then setting the end values to the mean minus twice the standard deviation. The longer, central node is adjusted to preserve the total burnup of the fuel assembly. Both PWR and BWR axial profiles are available. An alternative burnup profile for PWR spent fuel is presented in NUREG/CR-6801 for PWR spent fuel only, with enrichments up to 4.0 wt% <sup>235</sup>U initial. It should be noted that the axial profile database used in the 7-node axial profiles is an expansion of the data described in NUREG/CR-6801.

#### 2.3 Reactivity Calculations

The reactivity of a canister or cask using burnup credit may be calculated in the same fashion as a fresh fuel calculation, with the addition of detailed isotopic contents at each axial node. The calculated  $k_{eff}$  is then adjusted for the isotopic and cross section bias and uncertainty terms and compared to the reactivity limit equal to 1 minus the administrative margin that is typically 0.05 for storage and transport and zero for disposal.

Calculations are performed at a number of initial enrichments, typically ranging from 2.0 wt% <sup>235</sup>U initial through 5.0 wt%. A spacing of a half a weight percent is suggested to insure that a conservative interpolation will result for enrichments between the data points.

For each enrichment,  $k_{eff}$  is calculated at different burnup (including bias and uncertainty) and the results are plotted against the reactivity limit (e.g., 0.95). The burnup at which the  $k_{eff}$  plot crosses the reactivity limit is the required minimum burnup for that enrichment. If desired, a mathematical curve fit may be used to determine the intersection of the reactivity curve and the reactivity limit.

This process is repeated for all initial enrichment data points and the results are tabulated and plotted to provide a loading curve. Spent fuel assemblies with greater than the required minimum burnup may be loaded into the canister or cask, while assemblies with less than the minimum burnup may not be loaded.

#### 2.4 Isotopic Validation

Validation of isotopic contents may be achieved with any of three different approaches: Radiochemistry Assays (RCAs) for an entire spent fuel assembly, RCAs for specific pellets, and using a "Propagation of Errors" approach. RCAs for specific fuel pellets are currently not

#### Calculation Process

available for all of the 16 fission products selected by the methodology of this report, and data for fission products are quite sparse.

Countries with active reprocessing programs (e.g., Japan, France, and Great Britain) have isotopic measurements obtained through dissolving an entire spent fuel assembly or even several assemblies, which produces isotopic contents associated with the average fuel burnup. This approach may provide a more accurate validation of isotopic contents at a number of average burnup, which might then be related to the burnup at each axial node of a burnup credit calculation. However, assembly-based RCAs are not currently available within the United States.

#### 2.4.1 Actinide-Only Isotopic Validation

Validation of isotopic calculations is achieved by calculating the reactivity  $(k_{\infty})$  of a fuel sample using the RCA isotopic contents and comparing it to the reactivity obtained by using the isotopic depletion code. This process is repeated for each fuel sample and the bias and uncertainty for the dataset are calculated. Fission product and minor actinide isotopes are not included in the reactivity calculations in the Actinide-Only methodology.

The advent of the TRITON isotopic depletion code sequence allows greater accuracy than the older one-dimensional SAS2H code sequence, but this accuracy demands greater precision of the benchmark validation data. For example, the Yankee Rowe RCA data include samples that were burned very near a reactor control blade. TRITON calculations yielded a  $\Delta k/k$  [defined as  $(k_{\infty\text{-calc}} - k_{\infty\text{-exp}})/k_{\infty\text{-exp}})$ ] greater than six percent for these two samples without the control blade representation [Napolitano 2009 and YAEC 1961], but was reduced to less than a percent with the inclusion of the blade. By comparison, the same two data points yielded a  $\Delta k/k$  of just over a percent for a SAS2H analysis, indicating that the one-dimensional code is relatively insensitive to the treatment of the control blade region.

#### 2.4.2 Fission Product and Minor Actinide Isotopic Validation

The approach is to investigate the reactivity effect of each physical process involved in the production of isotopes in spent fuel. These processes are:

- The fission yield curve
- Radioactive buildup and decay
- Transmutation via neutron capture

#### **Fission Yield Curve**

The fission yield curve has been studied since the 1940s and a comprehensive evaluation of fission product yields is described in the "Compilation and Evaluation of Fission Yield Nuclear Data" developed by an IAEA Coordinated Research Project (CRP) [IAEA 2000].

#### Radioactive Buildup and Decay

Many isotopes created by the fission process are radioactive, and decay to produce other isotopes. The isotopic mixture is thus changing as decay chains are followed, and the isotopic depletion computer code must track these changes based upon half-life and branching ratio data.

#### Transmutation via Neutron Capture

The actinides and fission products in irradiated fuel have neutron capture (and scattering) cross sections, and can absorb a neutron and transmute to a heavier isotope. Neutron capture is often followed by radioactive decay, further complicating the radioactive buildup and decay process. The uncertainty in the neutron transmutation process is determined by the uncertainty in the cross section data, so this process is not independent of the cross section validation and its uncertainty term must be added algebraically to the total uncertainty.

#### 2.5 Cross-Section Validation through Critical Experiments

A Handbook of Evaluated Benchmark Critical Experiments [NEA 2009] provides hundreds of critical experiments applicable to the validation of fresh LWR fuel. This Handbook also contains Mixed-Oxide (MOX) experiments that are applicable for spent LWR fuel. MOX experiments generally have Pu contents that are greater than in spent LWR fuel, so the combination of fresh (UO<sub>2</sub>) experiments and MOX experiments encompasses the U and Pu mixture of spent fuel.

Uranium and plutonium isotopes in spent nuclear fuel are particularly well represented by a special series of critical experiments (HTC) performed in France and analyzed by ORNL [NUREG/CR-6979]. In the HTC experiments, the plutonium-to-uranium ratio and the isotopic compositions of both the uranium and plutonium used in the simulated fuel rods were designed to be similar to what would be found in a typical pressurized-water reactor fuel assembly that initially had an enrichment of 4.5 wt % <sup>235</sup>U and a burnup of 37.5 GWd/MTU. The HTC experiments included configurations to simulate fuel handling activities, pool storage, and transport in casks constructed of thick lead or steel. The ORNL analysis accounts for the extrapolation of this experiment set over the range of enrichments and burnup of interest for burnup credit. The bias plus the associated uncertainty,  $\Delta k_c$ , are provided in [NUREG/CR-6979] (taken from the Upper Subcritical Limit provided as Case 6, Table 7.1, NUREG/CR-6979), so that  $\Delta k_{C-Act} = 0.008$ . Note that the distribution of the HTC data is restricted and each user is expected to recreate the computer code inputs necessary to evaluate  $\Delta k_{C-Act}$ .

A combination of critical experiments from the Handbook plus the HTC experiments can provide a very complete range of applicability for fresh and spent LWR fuel. The availability of the analyses of the evaluated critical experiments and HTC experiments thus provides sufficient information for the actinide isotopes of uranium and plutonium.

Future analyses of the French fission product critical experiments may eventually be forthcoming, which would address the critical experiment validation of the fission product

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isotopes <sup>103</sup>Rh, <sup>143</sup>Nd, <sup>149</sup>Sm, <sup>151</sup>Sm, <sup>155</sup>Gd, and <sup>133</sup>Cs. These experiments employed the HTC fuel rods plus solutions of individual fission product isotopes.

Additional fission product critical experiments were performed in Japan at the Static Experiment Critical Facility (STACY) of the Japan Atomic Energy Agency (JAEA). These experiments used 5.0 wt% enriched UO<sub>2</sub> fuel rods in a 6% uranyl nitrate solution poisoned with elemental samarium, cesium, rhodium, and europium. Since natural elemental cesium consists only of <sup>133</sup>Cs and elemental rhodium consists only of <sup>103</sup>Rh, experiments with these isotopes are easily interpreted. The experiments were intended to represent a burnup of 30 GWd/MTU. The uranyl nitrate solution was included to simulate the environment in a reprocessing plant dissolver, which discourages the use of these critical experiments as a primary means of validating fission product cross sections for spent fuel storage and transport. However, these data provide a resource for confirmatory validation of fission product cross sections.

## 2.6 Cross-Section Validation for Fission Products and Minor Actinides Not Represented in Critical Experiments

The validation of fission products that are not represented in critical experiments is more challenging. The sixteen fission product isotopes considered in this report include the six mentioned above, plus <sup>95</sup>Mo, <sup>99</sup>Tc, <sup>101</sup>Ru, <sup>109</sup>Ag, <sup>145</sup>Nd, <sup>147</sup>Sm, <sup>150</sup>Sm, <sup>152</sup>Sm, <sup>151</sup>Eu, and <sup>153</sup>Eu. Lacking the results for the French fission product critical experiments, a value for  $\Delta k_C$  may be obtained by applying either the Direct Perturbation approach or the Indirect Perturbation approach, as described in [EPRI 2008a].

The Direct Perturbation approach used in [Wells 2005] is a "brute force" method to determine the reactivity worth of each individual isotope; the latter is then modified by reducing the cross section by the thermal cross-section uncertainty extracted from ENDF [LANL 2004]. The reactivity worth and the worth of the cross section uncertainty are thus determined for each isotope. This approach also works for minor actinide isotope absorbers included in the burnup credit isotope list: <sup>237</sup>Np, <sup>241</sup>Am, <sup>242m</sup>Am, and <sup>243</sup>Am. The results may be expressed as a  $\Delta k_{C-FP}$ for a given canister or a group of canisters with a particular boron fixed neutron absorber built into the basket. The  $\Delta k_C$  may also be represented as a percentage of the reactivity worth of the fission products and minor actinides, so that if the reactivity with only uranium and plutonium isotopes is calculated for each loading curve enrichment data point, a  $\Delta k_{C-FP}$  for that enrichment may be computed. This is important because the effect of fission products and minor actinides is very small at 2.0 wt% but much larger at 5.0 wt%, and a single value of  $\Delta k_{C-FP}$  would not be realistic.

The Indirect Perturbation approach is implemented at ORNL in TSUNAMI [Rearden 2005], and this provides a more powerful tool able to address the uncertainty of each isotope as a function of energy. This approach could provide a more accurate value for  $\Delta k_{C-FP}$  in the near future, using existing ENDF data or the new ENDF/B-VII data.

It should be noted that both the direct (MCNP) and indirect (TSUNAMI) methods are implementations of the "Propagation of Uncertainty" approach. In both methods, uncertainties in neutron cross sections are propagated to uncertainties in reactivity via a computer code system.

## **3** CRITICALITY VALIDATION

The validation of the cross sections for a burnup credit calculation is presented here as a two-step process. First, validation of the cross sections for an Actinide-Only burnup credit analysis is discussed using traditional methods using published [NEA 2009] fresh fuel and MOX evaluated benchmark critical experiment data plus the HTC [NUREG/CR-6979] simulated burned fuel critical experiment data.

Second, when fission products and minor actinides are included ("full burnup credit"), the user must select the desired isotopes to include and perform the perturbation analyses. MCNP users are provided the cross-section uncertainties that are required for a Direct Perturbation calculation; KENO and TSUNAMI users do not require this data for an Indirect Perturbation calculation.

#### 3.1 Actinide Cross-Section Validation

Validation of the actinide cross-sections can be performed by using the International Handbook of Evaluated Criticality Safety Experiments [NEA 2009] or by using the French HTC data described in [NUREG/CR-6979].

The evaluated benchmark critical experiment data [NEA 2009] consist of published critical experiments that have been reviewed for consistency and accuracy, and have been found to be acceptable for the validation of cross-section datasets for criticality safety calculations. The Handbook provides sufficient critical experiment data to validate UO<sub>2</sub> Fresh Fuel calculations and also provides sufficient MOX data to validate Actinide-Only burnup credit. The MOX critical experiments contain U and Pu in ratios that are not exactly what would be expected in spent LWR fuel, with the Pu content higher than in spent fuel. Evaluations of the UO<sub>2</sub> and MOX result in very similar bias values, as would be expected by systems with large quantities of <sup>238</sup>U. Since the bias values are very close, and no trends are manifested with Pu content, it is appropriate to collect the data into two sets: one comprised of the UO<sub>2</sub> experiments for fresh fuel validation and one comprised of the UO<sub>2</sub> and MOX experiments for Actinide-Only burnup credit. A typical Actinide-Only benchmark critical experiment validation will produce a bias of about – 0.007 to – 0.008, indicating an underprediction of k<sub>eff</sub> of less than one percent.

The French Haut Taux de Combustion (HTC) data are particularly useful in that the experiments simulated a typical pressurized-water reactor fuel assembly that initially had an enrichment of 4.5 wt % <sup>235</sup>U and was burned to 37.5 GWd/MTU. The HTC data were analyzed via the TSUNAMI computer code system [NUREG/CR-6979] to confirm its applicability for burnup

#### Criticality Validation

credit, and a  $\Delta k_{C-Act}$  was calculated, as shown in Table 3-1. The value listed is extracted from the "Upper Subcritical Limit" (USL) calculation.

Actinide (U+Pu) Cross-section Validation		
Fresh Fuel	0.0067	NEA 2009 evaluation
UO <sub>2</sub> + MOX	0.0078	NEA 2009 evaluation
HTC + MOX*	0.0072*	NUREG/CR-6979

## Table 3-1 Actinide-Only Cross-Section Bias and Uncertainties

\* From NUREG/CR-6979, Table 7.1, Case #6

It is expected that each cask vendor or applicant would re-perform the calculation of the HTC, MOX, and Fresh Fuel benchmark critical experiment datasets for their particular computer code version.

#### 3.2 Fission Product and Minor Actinide Cross-Section Validation

A study of the effects of cross-section uncertainty was performed for the Yucca Mountain Project [Wells 2005], which addressed the uncertainties for the "Principal Isotope" set. The latter includes 15 fission products (FPs) plus <sup>237</sup>Np and three americium isotopes. <sup>133</sup>Cs was also included in the study, although it is not part of the "Principal Isotope" set because of early concerns related to the volatility of cesium observed in the Three Mile Island accident. It was recognized that for commercial PWR and BWR fuel types, thermal cross-sections dominate the reactions with neutrons, and thus, uncertainties of the thermal cross-sections dominate the overall uncertainty. A study was performed at Los Alamos to evaluate the thermal neutron cross-section and also the range of values reported in different ENDF evaluations. These data were used to calculate a conservative estimate of the overall uncertainty in the Yucca Mountain Project report.

The reactivity effect of a thermal neutron cross-section uncertainty may be determined using the Direct Perturbation methodology and the MCNP computer code. Another methodology, the Indirect Perturbation methodology, is implemented in the TSUNAMI computer code system [Rearden 2005]. The Indirect Perturbation provides an automated means to incorporate ENDF uncertainties for all energies and uses the KENO computer code.

#### 3.2.1 Direct Perturbation Methodology

The process used for the validation of the fission product and minor actinide cross-sections is:

- Calculate k<sub>eff</sub> for U and Pu in system (Actinide-Only Burnup Credit). Bias is obtained from the Handbook of critical experiments [NEA 2009] and from the HTC experiments [NUREG/CR-6979].
- Calculate  $k_{eff}$  for each FP, adding one at a time to the base "Actinide-Only" case. The difference between the base case and the calculated  $k_{eff}$  with each FP is the  $k_{Worth}$  of that FP.
- Adjust the content of FP based upon the thermal cross-section uncertainty to maximize the uncertainty effect. The adjustments for the thermal neutron cross section, which dominates the interaction rates of the fission products and minor actinides, are provided in Tables 3-2 and 3-3.
- Recalculate  $k_{eff}$  for each FP, adding each individually to the base "Actinide-Only" case. The difference between the adjusted and unadjusted  $k_{eff}$  values is the  $\Delta k_U$  for that FP.
- Sum the  $\Delta k_U$  values to determine the total reactivity worth of the fission product and minor actinide uncertainties.

If the bias is completely due to ENDF uncertainties (ENDF bias), then the worst-case calculated bias for fission products is the sum of the individual  $\Delta k_U$  for all FPs. One would expect that the uncertainties at different energies would vary from maximum over-prediction to maximum under-prediction and would not always be the worst case, and the calculated bias would thus bound the bias that would be observed in traditional critical experiments.

The Direct Perturbation methodology can be applied using MCNP. Analysts are cautioned that the convergence of reactivity does not assure the convergence of the small  $\Delta k$  of a Direct Perturbation Calculation.

# 3.3 Cross-Section Uncertainties for Fission Product and Minor Actinide Validation

Tables 3-2 and 3-3 provide the cross-section uncertainties for fission products and minor actinides. The Evaluated Nuclear Data File (ENDF/B) provides the thermal cross-section uncertainty value for many isotopes as part of the dataset. In addition, the values of the thermal cross sections from ENDF/B-IV, ENDF/B-V, ENDF/B-VI and ENDF/B-VII (pre-release) were obtained [Bowen 2004] and treated with a methodology used for evaluation of expert elicitations (because the values are provided by expert cross section reviewers). Guidance for combining the results of expert elicitations and experimental results is not available, so the two values were simply summed to provide a "Total Uncertainty". An alternative approach that was considered was to use only the value that was larger numerically, or sum the two values as the square root of the sum of the squares because the values were independent. In any case, the approach of simply summing the two values is likely to provide a conservative result.

Note that the Confidence Factor applied to the MCNP calculations<sup>2</sup> has been multiplied by a factor of two. This additional margin could be adjusted to "fine tune" the uncertainties to be consistent with confirmatory data. The factor of two is sufficiently large that no additional margin is needed, and the individual isotopic factors could be reduced for some isotopes and still provides conservative results compared to the confirmatory data.

Principal Isotope	Cross Section Elicitation Uncertainty (%)	Experimental Measurement Uncertainty (%)	Total Uncertainty (%)	Confidence Factor Used In MCNP
Mo-95	6.26	3.57	9.83	0.80
Tc-99	0.00	5.00	5.00	0.90
Sm-149	2.80	2.00	4.80	0.90
Ru-101	0.00	26.47	26.47	0.26
Rh-103	3.00	2.00	5.00	0.90
Ag-109	1.00	2.00	3.00	0.94
Nd-143	1.00	3.08	4.08	0.92
Nd-145	17.00	4.76	21.76	0.58
Sm-147	26.00	6.00	32.00	0.34
Sm-150	1.97	3.85	5.81	0.88
Sm-151	1.32	2.00	3.00	0.96
Sm-152	0.00	2.91	2.91	0.96
Eu-151	0.00	1.09	1.09	0.98
Eu-153	0.00	2.24	2.24	0.96
Gd-155	1.00	1.00	2.00	0.96
Cs-133	0.00	3.00	3.00	0.94

### Table 3-2 Thermal Cross-Section Uncertainties for Fission Products

Note: Wells 2005, Table 1

Factor used in MCNP is one minus twice the "Total Uncertainty"

The six isotopes with bold font are the most important and are the subject of the French fission product critical experiments.

 $<sup>^{2}</sup>$  The Confidence Factor is the value used to adjust the macroscopic cross section in the MCNP calculations for the Direct Perturbation calculations of an isotope. The confidence factor is nominally the sum of the uncertainties of the thermal cross section obtained from a survey of ENDF values (elicitation) and experimental values reported by evaluators (measurement). The values are rounded in some cases (e.g., Mo-95).

Principal Isotope	Cross Section Elicitation Uncertainty (%)	Experimental Measurement Uncertainty (%)	Total Uncertainty (%)	Confidence Factor Used In MCNP
Np-237	12.65	2.00	14.65	0.68
Am-241	10.11	2.04	12.15	0.74
Δm-242m	0.71 (fission)	4.03	4.74	1.22*
7111-242111	0 (capture)	30.00	30.00	
Am-243	6.45	2.40	8.85	0.82

 Table 3-3

 Cross-Section Uncertainties for Minor Actinides

Note: Wells 2005, Table 2

Factor used in MCNP is one minus twice the "Total Uncertainty"

\* The Am-242m Confidence Factor is weighted by the relative strengths of the fission cross section and the capture cross section.

 $^{237}$ Np and  $^{242m}$ Am and  $^{243}$ Am, shown in Table 3-3, are not as important as the six most important fission products, and do not contribute substantially to the overall results. However, the presence of a substantial  $^{242m}$ Am fission cross section suggests that americium isotopes be included since the  $^{242m}$ Am could increase the value of k<sub>eff</sub>.  $^{237}$ Np is present in significant quantities, but has a relatively small cross section, and also could be omitted at the discretion of the user of the methodology.

The use of thermal cross-section uncertainties in the Direct Perturbation calculation of the reactivity effect of the cross-section uncertainties is justifiable only if thermal interactions dominate the interaction rates for each isotope. The relative ratio of interactions in the thermal range versus the epithermal + fast range is provided for each isotope in Table 3-4. MCNP provides optional tally functions that allow the convenient determination of the number of interactions in various user-supplied energy bins, and the ratio is easily determined.

Nuclide	Absorption Thermal/(Epithermal+Fast) Rate Ratio	Fission Thermal/(Epithermal+Fast) Rate Ratio	Overall Ratio*
Mo-95	12.5	0	12.5
Tc-99	12.2	0	12.2
Sm-149	849.4	0	849.4
Ru-101	1.8	0	1.8
Rh-103	39.9	0	39.9
Ag-109	24.1	0	24.1
Nd-143	19.0	0	19.0
Nd-145	9.2	0	9.2
Sm-147	5.8	0	5.8
Sm-150	16.9	0	16.9
Sm-151	142.2	0	142.2
Sm-152	54.2	0	54.2
Eu-151	89.3	0	89.3
Eu-153	8.5	0	8.5
Gd-155	676.1	0	676.1
Cs-133	8.7	0	8.7
Np-237	9.9	0	9.9
Am-241	34.0	1.0	34.0
Am-242m	160.7	114.5	124.8
Am-243	18.6	0	18.6

 Table 3-4

 Thermal to (Epithermal + Fast) Interaction Rate Ratios

\* Weighted by the number of neutrons

Inspection of Table 3-4 shows that, for Sm-149, thermal interactions take place at a ratio of 849.4 versus epithermal + fast interactions. The ratio of thermal interactions is also large for the most important six isotopes, but the ratio for <sup>101</sup>Ru is less than two. This raises the possibility of removing <sup>101</sup>Ru from the analysis methodology.

#### 3.4 MCNP Direct Perturbation Calculation

Once uncertainties in the thermal cross sections for each isotope are obtained and shown to be representative of the behavior of the cross section in spent fuel, a computer code such as MCNP
can be used to perform a Direct Perturbation calculation. SCALE users can employ the TSUNAMI computer code sequence, which includes KENO for reactivity calculations and includes both Direct and Indirect Perturbation calculations.

Direct Perturbation calculations using MCNP [Wells 2005] were performed using a Yucca Mountain Project waste package design that employed boron/stainless steel neutron absorber. (An applicant would perform such calculations for the container design defined in the application.) In these calculations, the microscopic cross section is not directly adjusted to reflect the uncertainty of a given fission product; rather, the macroscopic cross section actually used in the Monte Carlo process are adjusted by changing the isotopic contents by the amount of the cross-section uncertainty. Reactivity calculations for each fission product and minor actinide isotope were performed for both the original, unperturbed macroscopic cross section, and the perturbed cross section. Since these isotopes are neutron absorbers, the quantity of each isotope was reduced by the uncertainty value.

Calculations were performed for each individual fission product and minor actinide isotope, and also for the mixture. The results of these calculations were then normalized by the total worth of all of the fission products and minor isotopes, so that the change in reactivity ( $\Delta k_U$ ) for each isotope could be expressed as a fraction of the total fission product worth. The reactivity changes for several isotopes were very small and convergence was not assured, so the  $\Delta k_U$  for these isotopes were conservatively increased to a fractional reactivity worth of 0.2 percent. This value was determined to be the "minimum resolution" of the  $\Delta k_U$  calculations using the Direct Perturbation method for the waste packages and computer resources that were available.

The calculations for a PWR spent fuel container (a 21-PWR waste package design) are shown in Table 3-5. The reactivity of the container,  $k_{Worth}$ , with the major actinides (U and Pu isotopes) alone (Base Case) and the reactivity of the container with all isotopes including the fission products and minor actinides (Complete Case) are given at the upper part of the 4<sup>th</sup> column from the left in the table. The isotope name and the overall cross section uncertainty "Confidence Factor" from Tables 3-2 and 3-3 are listed in the 2<sup>nd</sup> and 3<sup>rd</sup> columns from the left in the table, followed by the reactivity (k<sub>Worth</sub>) and standard deviation of the spent fuel container with the major actinides plus only one isotope. The difference between the Base Case and each isotope k<sub>Worth</sub>,  $\Delta k_{Worth}$ , is the effect of adding that isotope to the mixture in the spent fuel pellets. For neutron absorbers including the fission products and minor actinides except for <sup>242m</sup>Am, this has the effect of increasing the neutron capture and decreases k<sub>eff</sub> by a small amount. The <sup>242m</sup>Am isotope has a strong fission cross section, so it contributes to the neutron chain reaction and its  $\Delta k_{Worth}$  has a different sign compared to the other isotopes.

The effect of the thermal cross section uncertainty is obtained by recalculating  $k_{Worth}$  but with the macroscopic cross section adjusted by the isotopic Confidence Factor, giving  $k_{Uncertainty}$ . The difference between each isotopes  $\Delta k_{Worth}$  and  $k_{Uncertainty}$  is the reactivity effect of cross section uncertainty,  $\Delta k_U$ , for that isotope. The isotopic  $\Delta k_U$  values are then renormalized by the total of all isotopic  $\Delta k_U$  values to give the percentage reactivity effect so that the individual contributions to the overall uncertainty may be distinguished.

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An isotope with a small  $\Delta k_{Worth}$  but a large  $\Delta k_U$  contributes little to the benefit of burnup credit while increasing the total bias and uncertainty  $\Delta k_{I-FP}$  disproportionately. Note that <sup>149</sup>Sm has a  $\Delta k_U$  of 1.2 percent but is one of the strongest contributors, while <sup>147</sup>Sm contributes only 13 percent (-0.00198/-0.01545) of the benefit of <sup>149</sup>Sm, but has an uncertainty penalty of 2/3rds (0.8/1.2) of the  $\Delta k_U$  of <sup>149</sup>Sm. Coupled with the poorer thermal/(epithermal + fast) interaction rate ration from Table 3-4, the <sup>147</sup>Sm isotope might be omitted from full burnup credit calculations. This could be at the discretion of the user of the burnup credit methodology.

## Table 3-5Reactivity Effects of Cross-Section Uncertainty

			(21-PW	R Waste Pa	ackage)					
	C	onfidenc	e							Percent
	Case	Factor	<b>k</b> worth	σ	$\Delta \mathbf{k}_{Worth}$		<b>k</b> <sub>Uncertainty</sub>	σ	∆k <sub>U</sub>	∆k <sub>u</sub>
Com	plete, U, Pu, FP	1.00	0.82589	0.00012	-0.1026	F.P.				∆k <sub>Worth</sub>
I	Base U,Pu Only	1.00	0.92845	0.00014		Reacti	vity			
	Nuclide					Rank	-			
1	Mo-95	0.80	0.92658	0.00021	-0.00187		0.92685	0.00022	0.00027	0.2
2	Tc-99	0.90	0.92421	0.00022	-0.00424		0.92502	0.00022	0.00081	0.7
3	Sm-149	0.90	0.91300	0.00014	-0.01545	2	0.91430	0.00014	0.00130	1.2
4	Ru-101	0.26	0.92745	0.00022	-0.00100		0.92775	0.00022	0.00030	0.3
5	Rh-103	0.90	0.91687	0.00014	-0.01158	4	0.91761	0.00014	0.00074	0.7
6	Ag-109	0.94	0.92645	0.00017	-0.00200		0.92659	0.00017	0.00014	0.1
7	Nd-143	0.92	0.91215	0.00014	-0.01630	1	0.91320	0.00015	0.00105	0.9
8	Nd-145	0.58	0.92530	0.00017	-0.00315		0.92666	0.00017	0.00136	1.2
9	Sm-147	0.34	0.92647	0.00017	-0.00198		0.92739	0.00017	0.00092	0.8
10	Sm-150	0.88	0.92667	0.00013	-0.00178		0.92681	0.00013	0.00014	0.1
11	Sm-151	0.96	0.91774	0.00014	-0.01071	5	0.91837	0.00014	0.00063	0.6
12	Sm-152	0.96	0.92401	0.00017	-0.00444		0.92406	0.00017	0.00005	0.0
13	Eu-151	0.98	0.92809	0.00017	-0.00036		0.92832	0.00017	0.00023	0.2
14	Eu-153	0.96	0.92503	0.00017	-0.00342		0.92539	0.00017	0.00036	0.3
15	Gd-155	0.96	0.92139	0.00014	-0.00706	6	0.92220	0.00014	0.00081	0.7
16	Cs-133	0.94	0.92230	0.00014	-0.00615	7	0.92266	0.00014	0.00036	0.3
17	Np-237	0.68	0.92267	0.00014	-0.00578		0.92570	0.00014	0.00303	2.7
18	Am-241	0.74	0.91604	0.00014	-0.01241	3	0.91926	0.00014	0.00322	2.9
19	Am-242m	1.22	0.92868	0.00013	0.00023		0.92922	0.00014	0.00054	0.5
20	Am-243	0.82	0.92671	0.00014	-0.00174		0.92731	0.00016	0.00060	0.5
	Total		Tota	I ∆k <sub>Worth</sub>	-0.1112				0.01686	15.2
								S	Sum ∆k <sub>∪</sub>	Reactivity
			NET	Ր <b>∆k<sub>Worth</sub></b>	-0.0943					Effect (Percent)

The results of the Direct Perturbation calculations using the thermal cross-section uncertainties are tabulated in Table 3-6. The values are expressed as percentages of the reactivity worth of all fission products and minor actinides. Because each isotope  $\Delta k_U$  was calculated separately, competition for neutron absorption from other fission products was not included and the values for  $\Delta k_U$  are thus conservatively overestimated by eight percent.

Table 3-6
Reactivity Effects of Cross-Section Uncertainty

	PWR	BWR
Mo-95	0.2	0.2
Tc-99	0.7	0.4
Sm-149	1.2	2.7
Ru-101	0.2	0.9
Rh-103	0.7	0.6
Ag-109	0.2	0.5
Nd-143	0.9	0.6
Nd-145	1.2	0.7
Sm-147	0.8	1.0
Sm-150	0.2	0.2
Sm-151	0.6	0.5
Sm-152	0.2	0.2
Eu-151	0.2	0.2
Eu-153	0.3	0.2
Gd-155	0.7	0.2
Cs-133	0.3	0.2
Np-237	2.7	1.8
Am-241	2.9	2.5
Am-242m	0.5	0.2
Am-243	0.5	0.3
Sum	15.2%	14.1%

The method of determining the reactivity effect of cross section described in this section is a conservative tool for the benchmarking of fission product and minor actinide cross sections.

### 3.5 Results and Discussion

The Direct Perturbation bias for spent PWR fuel for sixteen fission products isotopes is provided in Table 3-7.

Fuel Type	Number of FP and Minor Actinides Isotopes*	Total Δk <sub>worth</sub>	Δk <sub>u</sub> Percent of Δk <sub>worth</sub>	Net ∆k <sub>worth</sub>	Δk <sub>c-⊧</sub> , (Cross Section Uncertainty)
PWR	20	-0.1112	15.2	-0.0943	0.017
BWR	20	-0.1056	14.1	-0.0907	0.015

Table 3-7	
Bias for Spent Fuel Burnup Methodology Using Direct Perturbation Methodo	logy

\*16 FP and 4 minor actinides

The negative sign indicates that the fission products decrease keff. The comparison of the Total  $\Delta k$ Worth to the Net  $\Delta k$ Worth shows that the value of the effect of cross-section uncertainty reduces the benefit of fission products and minor actinides for burnup credit from about 0.11 to 0.094, a ~15 percent reduction in reactivity worth. The PWR cross section uncertainty,  $\Delta k$ C-FP, is 0.017 and the BWR cross section uncertainty is 0.015.

Each cask vendor would have to perform the direct (or indirect) perturbation calculation for their specific cask or canister designs. Users of this methodology are encouraged to choose the fission products and minor actinides that are included in "full" burnup credit from the 16 isotopes evaluated in this report. In particular, users might wish to remove the minor actinides, which contribute substantially to the total uncertainty, but are small contributors to the reactivity worth and provide little benefit to fission product burnup credit. Also, the possibility of removing <sup>101</sup>Ru, <sup>145</sup>Nd, and <sup>147</sup>Sm from the analysis methodology should be considered because their reactivity worth contributions are small, but their contributions to uncertainty are significant.

# **4** ISOTOPIC VALIDATION

Isotopic validation is traditionally achieved through comparisons of reactivity of spent fuel using calculated isotopic contents against reactivity using Radiochemistry Assays (RCAs). RCAs have been used successfully to validate the actinide contents of spent fuel, but difficulties have arisen with the validation of fission products and minor actinides via spent fuel pellets. Essentially, the complexity of modeling the behavior of a single pellet plus the difficulty of accurate chemical assays of a single pellet, for isotopes present in small quantities, have been quite challenging. The RCAs available to date [Scaglione 2002] have not provided a statistically significant dataset for the fission product isotopes, so a distribution-free tolerance limit approach was taken to calculate a fission product bias and uncertainty,  $\Delta k_{I}$  (major actinides + minor actinides + fission products), of 0.0249. A recent re-evaluation [NUREG/CR-6968] of the TMI-1 RCAs, which provide much of the available fission product RCA data, has yielded a more accurate representation of the fuel assemblies and used the TRITON code sequence. In addition, an adjustment was made to match the specific power derived from reactor records to the burnup calculated from the <sup>148</sup>Nd isotopic content. In spite of these improvements in the treatment of TMI-1 RCA data, agreement between the calculated and measured data for the actinides and fission products remains poor. Regardless, additional data may eventually make it possible to develop a dataset with a normal distribution amenable to traditional statistical methods.

## 4.1 Actinide-Only Isotopic Validation

A typical Actinide-Only isotopic validation uses Radiochemistry Assay (RCA) data from seven or eight different PWR reactors providing 55 to 63 data points, as described in Table 4-1. Eight data points from Yankee Rowe may be omitted if sufficient information regarding the control blades [YAEC 1997] is not included.

Reactor	Assembly Design	Numbers of Samples/ Assemblies/Rods	Sample Burnup (GWd/MTU)	Initial Enrichment (wt% U-235)
Tripo Vorcollos	Westinghouse,	14/2/6	12.042	3.897
Third vercenes	Irregular, Blades	14/3/0	11.529-24.548	3.13
Turkey Point	W 15x15, 20 GT	5/2/5	30.72-31.56	2.556
			6.92-8.3	3.208
Mihama	W 15x15, 20 GT	9/3/NA	14.66-21.29	3.203
			29.5-34.32	3.210
Takahama	Westinghouse 17x17 (Mitsubishi)	11/2/2	14.30-47.25	4.11
H.B. Robinson	W 15x15, 20 GT, 12 BP	4/1/1	16.02-31.66	2.561
Obrigheim	Siemens 14x14	6/5/special	25.93-29.52	3.13
			27.35-44.34	3.038
Calvert Cliffs	BB propert	6/3/3	18.68-33.17	2.72
	br present		31.40-46.46	2.453
Yankee Rowe	W, irregular, control blade followers	8/1/3	15.95-35.97	3.4

#### Table 4-1 Radiochemical Assay Information

Evaluations of these RCAs may be performed with a variety of computer code systems, commonly TRITON and the older SAS2H. SAS2H uses a one-dimensional code to solve the neutron transport equation prior to creating the requisite data libraries for the ORIGEN-S isotopic depletion code. TRITON uses a recent two-dimensional code to solve the neutron transport equation, and is able to more accurately represent the neutron energy spectrum in a fuel rod near a strong absorber such as a control rod, control blade, or control blade follower. Several of the RCA samples for Yankee Rowe are of particular interest since they were irradiated only two rows in from the control blade follower, which displaces water from the slot between assemblies when the control blades are withdrawn for reactor operation. This water displacement substantially hardens the neutron energy spectrum and causes the production and burning of more Pu<sup>239</sup>, with the result that the discharge reactivity of these samples is greater than if water were present in the slot. SAS2H is insensitive to this issue because if the control blade slot is treated as water-filled, the change in the total water in the assembly is small and there is only a small reactivity consequence. A TRITON calculation with a water-filled slot results in a six percent or more decrease in calculated reactivity because TRITON can accurately represent the slot. Thus the use of a more modern code such as TRITON places greater demands upon the accuracy of the data that describe the irradiation environment of an RCA sample. With

the control blade follower modeled in the slot with TRITON, the TRITON calculated reactivity is much better than SAS2H.

Each RCA sample modeled in TRITON, SAS2H or another computer code includes the geometry and irradiation history, and the isotopic contents of the spent fuel sample are calculated. A reactivity calculation is performed with the isotopic contents and compared to the reactivity obtained using the measured isotopic contents, and a  $\Delta k_{I-Act}$  is calculated for each RCA sample. The individual sample  $\Delta k_{I-Act}$  values are averaged and a statistical uncertainty  $\sigma$  is calculated, providing the bias and uncertainty of the Actinide-Only isotopic calculations. Trend analyses are performed to insure that any dependencies of the bias are included, although typically no trends are observed. The final  $\Delta k_{I-Act}$  value is summed statistically with the value of  $\Delta k_{C-Act}$ . A typical calculation of the bias and uncertainty using TRITON results in a value of  $\Delta k_{I-Act}$  around 0.011. Each user is expected to perform an Actinide-Only isotopic validation analysis.

### 4.2 Fission Product and Minor Actinide Isotopic Validation

An alternative approach is one that is often used is the Propagation of Errors method. This method breaks down the complex phenomenon under study into its component physical processes so that the uncertainties, or error, of each process can be individually established. Then the individual process errors are statistically summed to produce the uncertainty of the desired complex phenomenon. In the case of isotopic calculations for fission products, the physical processes that establish the concentrations of fission products are:

- Fission yield
- Radioisotope buildup and decay
- Transmutation by neutron capture

The fission yield and radioactive decay processes are independent and their evaluations are independent, so their contributions to  $\Delta k_{I-FP}$  may be summed statistically. The process of transmutation of isotopes by neutron capture depends upon the neutron cross section, and the evaluation of the contribution to  $\Delta k_{I-FP}$  from capture employs a sensitivity study that computes the change in isotopic content resulting from an uncertainty in capture cross section. This dependence upon cross sections mandates that the capture effect contribution be added algebraically to  $\Delta k_{I-FP}$ .

### 4.2.1 Fission Yield Uncertainty

The fission yield curves for <sup>235</sup>U, <sup>239</sup>Pu, and a variety of other fissionable actinides, have been extensively studied beginning in the 1940s. The most complete published evaluation of the fission yield curves was described in the "Compilation and Evaluation of Fission Yield Nuclear Data" developed by an IAEA Coordinated Research Project (CRP) [IAEA 2000]. The abundances and uncertainties for the fission products and minor actinides are evaluated in this report. The uncertainties for the fission yield for the atomic masses of interest range from about

#### Isotopic Validation

a percent in the peak regions for <sup>235</sup>U and <sup>239</sup>Pu, which dominate in terms of their contributions to the number of fissions.

Uncertainties of up to four percent are suggested for the burnup credit isotopes [IAEA 2000, Table 7.1.2]. The uncertainties for the fission product and minor actinide isotopes are provided in Table 4-2. The uncertainties are applied by adjusting the number density in MCNP inputs for each of the 16 isotopes by a multiplier of 1 minus the uncertainty. A Direct Perturbation calculation is then performed for each isotope to obtain the reactivity effect of the fission yield uncertainty.

Table 4-2 Fission Yield Uncertainties

	Fission Yield Uncertainty (Percent) [IAEA 2000, Table 7.1.2]	Fission Yield Adjustment Factor (1-U)
Mo-95	3.00	0.97
Tc-99	4.30	0.96
Sm-149	4.30	0.96
Ru-101	4.30	0.96
Ru-103	4.30	0.96
Ag-109	4.30	0.96
Nd-143	2.09	0.98
Nd-145	2.02	0.98
Sm-147	3.32	0.97
Sm-150	2.00	0.98
Sm-151	4.30	0.96
Sm-152	4.30	0.96
Eu-151	4.30	0.96
Eu-155	4.30	0.96
Gd-155	4.30	0.96
Cs-133	3.94	0.96

A reactivity worth of 4.1 percent due to uncertainty was obtained for the fission yield process, as shown in Table 4-3. The  $\Delta k_{I-FP(fission yield)}$  contribution for the fission yield curve is determined by applying the fission yield adjustment factors to the direct perturbation data provided in Table 3-5. The procedure used to determine the reactivity worth via Direct Perturbation calculations is

described in Section 3.2.1. The direct perturbation data provide the reactivity effect (or sensitivity) for each isotope, determined by MCNP calculations using the thermal cross section uncertainties shown in Tables 3-2 and 3-3. The reactivity effect of fission yield uncertainties is independent of the cross section uncertainty; the use of the cross section uncertainty table is only a convenience as a source of the sensitivity information relating a change in the macroscopic cross section to a change in  $k_{eff}$ . The macroscopic cross section, used by MCNP, is the product of the isotopic content (number density) and microscopic cross section (generally referred to just as the cross section), so perturbation calculations performed for the purpose of evaluating uncertainties in any other parameter (isotopic contents).

Table 4-3 shows that the reactivity effect of fission yield uncertainties is 4.1 percent of the total reactivity worth of fission products and minor actinides, as compared to the 15.2 percent of the reactivity effect resulting from the cross section uncertainties (Table 3-5). The value of 4.1 percent is obtained in Table 4-3 by calculating the "Ratio of Uncertainties," i.e., the ratio of fission yield uncertainties to cross section uncertainties. The cross section uncertainty is equal to one minus the "Cross Section Confidence Factor" in the third column from the left, and is converted to percent and tabulated in the column labeled "Cross Section Uncertainty" (2<sup>nd</sup> column from the right). The Ratio of Uncertainty. The "Fission Yield  $\Delta k_U / \Delta k_{Worth}$ " is the product of the "Ratio of Uncertainties" and the "Cross Section  $\Delta k_U / \Delta k_{Worth}$ ". The individual values for each isotope are summed and the total reactivity effect of fission yield uncertainty is 4.1 percent.

The  $\Delta k_{I-FP(fission yield)}$  is thus (4.1 percent divided by 15.2 percent, times the  $\Delta k_{C-FP}$  value of 0.017 in Table 3-7) equal to 0.0046. Similarly, the  $\Delta k_{I-FP(fission yield)}$  for BWR is (4.1%/14.1% times 0.015 in Table 3-7) equal to 0.0044.

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#### Table 4-3

**Reactivity Effect of Fission Yield Uncertainties** 

Reactivity Effect of Cross Section Uncertainties X-sec (21-PWR Waste Package) Confidence								Cross Section Percent		Fission Yield Uncertainty	Cross Section Uncertainty	Ratio of Uncertainties	Fission Yield Percent		
	Case	Factor	<b>k</b> <sub>Worth</sub>	σ	$\Delta \mathbf{k}_{Worth}$		<b>k</b> <sub>Uncertainty</sub>	σ	∆k <sub>u</sub>	Δk <sub>U</sub>		(Percent)	(Percent)		Δk <sub>U</sub>
Com	olete, U, Pu, FP	1.00	0.82589	0.00012	-0.1026	F.P.				$\Delta k_{Worth}$		[Table 7.1.2]	(1-Confidence	-	$\Delta \mathbf{k}_{Worth}$
E	Base U,Pu Only	1.00	0.92845	0.00014		Reacti	vity						Factor)		
	Nuclide					Rank								0.45	
1	Mo-95	0.80	0.92658	0.00021	-0.00187		0.92685	0.00022	0.00027	0.2	M0-95	3.00	20.0	0.15	0.04
2	I c-99	0.90	0.92421	0.00022	-0.00424		0.92502	0.00022	0.00081	0.7	IC-99	4.30	10.0	0.43	0.31
3	Sm-149	0.90	0.91300	0.00014	-0.01545	2	0.91430	0.00014	0.00130	1.2	Sm-149	4.30	10.0	0.43	0.50
4	Ru-101	0.26	0.92745	0.00022	-0.00100		0.92775	0.00022	0.00030	0.3	RU-101	4.30	74.0	0.06	0.02
5	Rh-103	0.90	0.91687	0.00014	-0.01158	4	0.91761	0.00014	0.00074	0.7	Rn-103	4.30	10.0	0.43	0.29
6	Ag-109	0.94	0.92645	0.00017	-0.00200		0.92659	0.00017	0.00014	0.1	Ag-109	4.30	6.0	0.72	0.09
7	Nd-143	0.92	0.91215	0.00014	-0.01630	1	0.91320	0.00015	0.00105	0.9	Nd-143	2.09	8.0	0.26	0.25
8	Nd-145	0.58	0.92530	0.00017	-0.00315		0.92666	0.00017	0.00136	1.2	Nd-145	2.02	42.0	0.05	0.06
9	Sm-147	0.34	0.92647	0.00017	-0.00198		0.92739	0.00017	0.00092	0.8	Sm-147	3.32	66.0	0.05	0.04
10	Sm-150	0.88	0.92667	0.00013	-0.00178		0.92681	0.00013	0.00014	0.1	Sm-150	2.00	12.0	0.17	0.02
11	Sm-151	0.96	0.91774	0.00014	-0.01071	5	0.91837	0.00014	0.00063	0.6	Sm-151	4.30	4.0	1.08	0.61
12	Sm-152	0.96	0.92401	0.00017	-0.00444		0.92406	0.00017	0.00005	0.0	Sm-152	4.30	4.0	1.08	0.05
13	Eu-151	0.98	0.92809	0.00017	-0.00036		0.92832	0.00017	0.00023	0.2	Eu-151	4.30	2.0	2.15	0.44
14	Eu-153	0.96	0.92503	0.00017	-0.00342		0.92539	0.00017	0.00036	0.3	Eu-153	4.30	4.0	1.08	0.35
15	Gd-155	0.96	0.92139	0.00014	-0.00706	6	0.92220	0.00014	0.00081	0.7	Gd-155	4.30	4.0	1.08	0.78
16	Cs-133	0.94	0.92230	0.00014	-0.00615	7	0.92266	0.00014	0.00036	0.3	Cs-133	3.94	6.0	0.66	0.21
17	Np-237	0.68	0.92267	0.00014	-0.00578		0.92570	0.00014	0.00303	2.7					
18	Am-241	0.74	0.91604	0.00014	-0.01241	3	0.91926	0.00014	0.00322	2.9					
19	Am-242m	1.22	0.92868	0.00013	0.00023		0.92922	0.00014	0.00054	0.5					
20	Am-243	0.82	0.92671	0.00014	-0.00174		0.92731	0.00016	0.00060	0.5					
	Total		Tota	I ∆k <sub>worth</sub>	-0.1112				0.01686	15.2					4.1
								5	Sum ∆k <sub>∪</sub>	Reactivity					Reactivity
			NET	ΓΔk <sub>Worth</sub>	-0.0943					Effect (Percent)					Effect (Percent)

## 4.2.2 Radioactive Buildup and Decay Effect

The isotopes that are included for fission product and minor actinide burnup credit are relatively stable. Fission products are produced directly by fission, but are also produced by the decay of an unstable precursor isotope. The uncertainties in the half-lives of the isotopes included in the ORIGEN-S depletion code libraries were evaluated [Hermann 1998].

A calculation of the reactivity effect of these radioactive decay isotopic uncertainties was performed by Connell and Kochendarfer [Connell 2002], who found that the effect was quite small,  $\Delta k_{I-FP(radioactive decay)} = \sim 0.000375$ . The magnitude of this contribution to the overall  $\Delta k_{I-FP}$  is small and applicable to large, poisoned containers, and it is suggested that applicants use the published number instead of re-calculating a value.

### 4.2.3 Neutron Capture and Transmutation Effect

The direct effect of cross section uncertainties upon  $k_{eff}$  is evaluated in [Wells 2005], but cross section uncertainties also propagate to isotopic content uncertainties. A study of isotopic effects [Gauld 2005] provides the relative sensitivity of isotopic contents to uncertainties in cross sections, and these sensitivities are used to scale the reactivity effects determined by [Wells 2005].

### 4.3 Isotopic Sensitivities

This section provides a description of the isotopic sensitivities of fission products and minor actinides.

The relative isotopic sensitivity of a given isotope to a change in cross section is expressed as:

$$S = (\Delta N/N)/(\Delta \sigma/\sigma)$$
 Equation 4-1

where

- S = Sensitivity of the concentration of isotope to uncertainty in cross section
- $\Delta N = Change in number density of isotope$
- N = Number density of isotope
- $\Delta \sigma$  = Change in cross section of isotope
- $\sigma$  = Cross section of isotope

A relative sensitivity of 1.0 would indicate that the concentration of an isotope is as sensitive as the cross section, i.e., a 10-percent change in cross section would result in a 10-percent change in

#### Isotopic Validation

the isotopic concentration. Fission product and minor actinide concentrations have sensitivities less than one because if the cross section were to decrease, the isotope would have fewer neutron capture reactions during irradiation in a reactor, which would result in a larger concentration of that isotope upon discharge from the reactor. Thus a neutron capture reaction has a negative sensitivity while precursor isotope decay has a positive sensitivity. Relative sensitivities for fission products are presented in [Gauld 2005]. The values of the relative sensitivities for each fission product and minor actinide are provided in Table 4-4 for each reaction, i.e., precursor isotope decay producing the isotope and neutron capture removing the isotope. The overall sensitivity for each isotope is provided by summing the reaction sensitivity values. Values for minor actinides were not provided in [Gauld 2005] and are set to the value of 1.0.

### 4.3.1 Linear Perturbation of Reactivity Effects

The reactivity effects of isotopic uncertainties are assumed to be linearly dependent upon the change in cross section of each isotope. The reactivity effect of isotopic uncertainty *for a given isotope* is:

$$\Delta k_{I} = S \times \Delta k_{C}$$
 Equation 4-2

where

 $\Delta k_{I} =$  Reactivity effect of isotopic uncertainty S = Relative sensitivity from Equation 4-1  $\Delta k_{C} =$  Reactivity effect of cross section uncertainty

## 4.4 Reactivity Effects of Isotopic Uncertainty

The reactivity effects of isotopic uncertainty are engendered by cross section uncertainty in the depletion calculations that calculate the isotopic changes to the fuel during irradiation in a reactor. Applying Equation 4-2 to the  $\Delta k$  value for each isotope of Table 3-6 and the sensitivities of Table 4-4 produces the reactivity effects of isotopic uncertainty presented in Tables 4-5 and 4-6 for PWR and BWR spent nuclear fuel, respectively. The reactivity effects of isotopic uncertainties, 5.2 percent for PWR and 2.5 percent for BWR, are less than the reactivity effects of cross section uncertainties, 15.2 percent and 14.1 percent, as expected, because the sensitivity coefficients are less than one (except for americium) and because of the negative reaction sensitivities of some of the nuclides.

## Table 4-4Isotope Relative Sensitivities

#### **Reactivity Effect of Cross Section Uncertainties Upon Isotopic Compositions**

	Principal															
	Isotope	Reaction <sup>e</sup>	S	Reaction <sup>e</sup>	S <sup>a</sup>	<b>S</b> <sub>Total</sub> <sup>b</sup>										
1	Mo-95	Zr-94	2.52E-04	Nb-94	4.14E-08	Zr-95	-4.56E-04	Nb-95	-9.96E-04	Nb-95m	-6.00E-06			Mo-95	-6.70E-02	-0.0653
2	Tc-99	Mo-98	4.17E-03	Tc-98	8.68E-09	Mo-99	-9.64E-05							Tc-99	-1.26E-01	-0.1217
3	Sm-149	Pm-147	2.79E-01	Nd-148	2.77E-02	Pm-148	8.77E-02	Pm-148m	6.21E-02	Sm-148	1.28E-02	Pm-149	-1.31E-02	Sm-149	-1.00E+00	-0.5176
4	Ru-101	Mo-100	3.16E-03	Ru-100	1.16E-03									Ru-101	-5.68E-02	-0.0525
5	Rh-103	Ru-102	5.26E-03	Rh-102	5.09E-09	Ru-103	-3.72E-03	Rh-103 (f,γ)	1.16E-13					Rh-103	-4.64E-01	-0.4550
6	Ag-109	Pd-108	1.20E-01	Pd-108(f,γ)	-2.16E-08	Pd-109	-1.06E-04	Ag-109(f,γ)	-2.15E-07					Ag-109	-3.85E-01	-0.2649
7	Nd-143	Ce-142	2.30E-03	Pr-142	5.21E-06	Nd-142	6.28E-04	Ce-143	-8.70E-05	Pr-143	-5.92E-03			Nd-143	-4.59E-01	-0.4501
8	Nd-145	Nd-143	2.56E-03	Ce-144	1.52E-03	Nd-144	1.07E-02							Nd-145	-1.53E-01	-0.1382
9	Sm-147	Nd-146	4.32E-03	Nd-147	-1.78E-02	Pm-147	-3.63E-01	Pm-147(f,γ)	2.10E-05					Sm-147	-3.30E-01	0.0551
10	Sm-150	Nd-148	1.57E-02	Pm-148	6.89E-02	Pm-148m	6.01E-02	Sm-149	-3.24E-03	Pm-150	-9.42E-06			Sm-150	-1.78E-01	-0.0301
11	Sm-151	Sm-149	-2.13E-03	Nd-150	1.42E-02	Sm-150	4.07E-01	Pm-151	-2.73E-03					Sm-151	-9.90E-01	-0.5639
12	Sm-152	Sm-150	2.37E-01	Sm-151	1.60E-02									Sm-152	-7.30E-01	-0.4770
13	Eu-151 <sup>d</sup>													Eu-151	-1	-1.0000
14	Eu-153	Sm-151	2.32E-02	Sm-152	3.19E-01	Eu-152m	1.54E-06	Sm-153	-6.51E-03					Eu-153	-5.52E-01	-0.2033
15	Gd-155	Eu-153	4.77E-01	Eu-154	-5.58E-02	Gd-154	1.74E-01	Eu-155	-7.77E-01					Gd-155	-1.01E+00	0.4738
16	Cs-133	Xe-132	1.49E-03	Xe-132(f,γ)	-7.16E-07	Cs-132	1.14E-10	I-133	-4.90E-05	Xe-133	-4.57E-03	Xe-133m	-5.68E-05	Cs-133	-1.79E-01	-0.1728
17	Np-237 <sup>d</sup>	Am-241	1													1.0000
18	Am-241 <sup>c</sup>	Pu-240	1													1 0000
10	$\Delta m_2 242 m^c$	Am 241	1													1 0000
19	A 040 <sup>C</sup>	AUT-241	1													1.0000
20	Am-243*	Am-242m	1													1.0000

#### Notes:

a. Sensitivities of an isotope to itself are always negative due to coupling of cross section and isotopic effects

b. Sensitivities of an isotope to neutron capture by parents may be positive or negative. Absolute value is summed due to independence of these nuclides

c. Sensitivities for minor actinides are conservative estimates applying a maximum value of 1.0

d. Sensitivity for Eu-151 is scaled from Gd-155, Sm-149, and Sm-151 isotope depletion sensitivities which have similar cross section magnitudes

e. Reactions are neutron capture and parent isotope decay to produce a principal isotope, and decay of the principal isotope

Source: ORNL/TM-2005/48, Gauld, I.C. and Mueller, D.E., "Evaluation of Cross-Section Sensitivities in Computing Burnup Credit Fission Product Compositions, August 2005

	(21-PWR Was		
	S <sub>Total</sub> *	Isotopic $\Delta k_{u}$	 Δk <sub>worth</sub>
Mo-95	-0.0653	-0.00002	-0.02
Tc-99	-0.1217	-0.00010	-0.1
Sm-149	-0.5176	-0.00067	-0.6
Ru-101	-0.0525	-0.00002	-0.02
Rh-103	-0.4550	-0.00034	-0.3
Ag-109	-0.2649	-0.00004	-0.04
Nd-143	-0.4501	-0.00047	-0.4
Nd-145	-0.1382	-0.00019	-0.2
Sm-147	0.0551	0.00005	0.05
Sm-150	-0.0301	0.00000	-0.004
Sm-151	-0.5639	-0.00036	-0.3
Sm-152	-0.4770	-0.00002	-0.02
Eu-151	-1.0000	-0.00023	-0.2
Eu-153	-0.2033	-0.00007	-0.1
Gd-155	0.4738	0.00038	0.4
Cs-133	-0.1728	-0.00006	-0.06
Np-237	1.0000	0.00303	2.9
Am-241	1.0000	0.00322	3.1
Am-242m	1.0000	0.00054	0.5
Am-243	1.0000	0.00060	0.6
		Percent Reactivity Effect	5.2

## Table 4-5PWR Reactivity Effect for Isotopic Uncertainties due to Cross Section Uncertainties

\* From Table 4-4

	(44-BWR Wa				
	S <sub>Total</sub> *	S <sub>Total</sub> * Isotopic Δk <sub>u</sub>			
Mo-95	-0.0653	-0.00001	-0.008		
Tc-99	-0.1217	-0.00005	-0.044		
Sm-149	-0.5176	-0.00148	-1.4		
Ru-101	-0.0525	-0.00005	-0.049		
Rh-103	-0.4550	-0.00029	-0.3		
Ag-109	-0.2649	-0.00015	-0.1		
Nd-143	-0.4501	-0.000428	-0.3		
Nd-145	-0.1382	-0.00010	-0.1		
Sm-147	0.0551	0.00006	0.1		
Sm-150	-0.0301	0.00001	0.007		
Sm-151	-0.5639	-0.00028	-0.3		
Sm-152	-0.4770	-0.00007	0.1		
Eu-151	-1.0000	-0.00016	0.2		
Eu-153	-0.2033	-0.00004	-0.039		
Gd-155	0.4738	0.00003	0.027		
Cs-133	-0.1728	-0.00005	-0.05		
Np-237	1.0000	0.00195	1.8		
Am-241	1.0000	0.00265	2.5		
Am-242m	1.0000	0.00022	0.2		
Am-243	1.0000	0.00027	0.3		
		Percent Reactivity Effect	2.5		

## Table 4-6BWR Reactivity Effect for Isotopic Uncertainties due to Cross Section Uncertainties

\* From Table 4-4

Table 4-5 shows that the PWR reactivity effect of isotopic uncertainties is 4.7 percent of the total reactivity worth of fission products and minor actinides, as compared to the 15.2 percent of the reactivity effect resulting from the cross section uncertainties (Table 3-5). The  $\Delta k_{I-FP(Capture)}$  is thus (5.2%/15.2% times the  $\Delta k_{C-FP}$  value of 0.017 in Table 3-7) equal to 0.0058. Similarly, the  $\Delta k_{I-FP(Capture)}$  for BWR is (2.5/14.1 times 0.015 in Table 3-7) equal to 0.0027.

## 4.5 Summary

The formation of fission product and minor actinide isotopes can be broken down into three separate processes so that the reactivity effect of the uncertainties of each process can be separately evaluated. The reactivity effects of the three sources of isotopic uncertainty are summarized in Table 4-7.

#### Table 4-7

Fission Product Isotopic Validation Reactivity Contributions

Fission Product Isotopic Validation Source of Uncertainty	PWR Value	BWR Value
Reactivity Effect of Fission Yield Curve	0.0046	0.0044
Reactivity Effect of Decay Constants	0.000375	0.000375
Reactivity Effect of Isotopic Uncertainty Caused by Uncertainty of Capture Cross Sections	0.0058	0.0027

# **5** FISSION PRODUCT BIAS CONFIRMATION

The validation of Actinide-Only Burnup Credit follows standard industry procedures and applies experimental data that define the isotopic contents for actinide isotope production and the reactivity effect of cross-sections. The availability of fission product data of this type is limited, so methodologies that use existing data describing the separate physical processes of isotopic production, and uncertainty data for cross sections, were developed. The classical "Propagation of Uncertainties" methodology is used to determine the reactivity bias factors. Confirmation of these bias values can be provided by the available fission product RCA data and critical experiments such as the Commercial Reactor Criticals (CRCs) and the (currently unavailable) French fission product criticals.

## 5.1 Commercial Reactor Criticals

The Commercial Reactor Criticals database consists of reactor restart and reactor startup physics tests for four reactors and 45 state points (Table 5-1). Startup physics testing may be performed in a PWR either hot or cold, and cold startups are particularly applicable to spent fuel storage and transport systems, which are cold when in a flooded condition. Reactor criticals involve detailed analyses of the isotopic contents of each assembly in the core for each cycle, and a reactivity is calculated for each restart. The reactivity calculation includes the uncertainties of both the isotopic calculations and the cross-section uncertainties of every isotope present in the reactor. The CRCs are termed "integral" experiments because both isotopic and cross-section uncertainties for all isotopes present are tested with a single reactivity measurement. Strong neutron absorbers with short half-lives, such as xenon, have essentially decayed before the restart so more long-lived isotopes dominate the calculations.

The CRCs have been shown [NUREG/CR-6951] to have very similar neutron spectra to spent fuel containers and transport casks, and are thus applicable critical experiments for burnup credit validation. For the purposes of confirmatory data, the bias and uncertainty of the reactor restarts are compared to the overall bias and uncertainty of fission product burnup credit.

Table 5-	1	
Tabulati	on of CRC	Reactivities

Case Name	<b>k</b> <sub>eff</sub>	σ	Case Name	<b>k</b> <sub>eff</sub>	σ
Crystal River 2	1.00156	0.00043	Crystal River 23	1.00108	0.00045
Crystal River 3	1.00867	0.00042	Crystal River 24	1.00331	0.00047
Crystal River 4	1.00305	0.00044	Crystal River 25	1.01073	0.00048
Crystal River 5	1.00267	0.00046	Crystal River 26	1.01154	0.00044
Crystal River 6	1.00662	0.00044	Crystal River 27	1.01113	0.00048
Crystal River 7	1.00686	0.00044	Crystal River 28	1.00055	0.00044
Crystal River 8	0.99922	0.00045	Crystal River 29	1.01222	0.00048
Crystal River 9	1.00481	0.00045	Crystal River 30	1.00534	0.00049
Crystal River 10	1.01265	0.00043	Crystal River 31	1.01968	0.00046
Crystal River 11	1.00096	0.00044	Crystal River 32	1.00108	0.00048
Crystal River 12	1.0173	0.00044	Crystal River 33	1.01232	0.00053
Crystal River 13	1.00423	0.00039	Three Mile Island 2	1.00048	0.00047
Crystal River 14	1.00784	0.00048	Three Mile Island 3	1.00443	0.00046
Crystal River 15	1.01418	0.00041	Sequoyah 2	1.00109	0.00046
Crystal River 16	1.00008	0.00043	Sequoyah 3	1.00679	0.00047
Crystal River 17	1.0075	0.00044	McGuire 2	0.99428	0.00043
Crystal River 18	1.00819	0.00045	McGuire 3	1.00013	0.00045
Crystal River 19	1.00824	0.00046	McGuire 4	0.99755	0.00049
Crystal River 20	1.01973	0.00047	McGuire 5	1.00565	0.00043
Crystal River 21	1.01584	0.00044	McGuire 6	1.00786	0.00047
Crystal River 22	0.99788	0.00044			

Note: Wells 2003, Table 4

The mean of the 45 state points is -0.007769 (underprediction) and the standard deviation is 0.003208, while the statistical uncertainty in the MCNP calculations is 0.000448. With a 95 percent confidence factor of 2.017 for the 45 state points, the bias plus uncertainty is -0.01430. Thus the combination of isotopic and reactivity calculated could underpredict the reactivity by as much as -0.0143.

## 5.2 Fission Product Data from Radiochemistry Assays

Limited fission product isotopic data are available from a number of sources, including actinide and fission product data from RCAs for the Bugey and Gravelines [EPRI 2008b]. These data can be used to confirm the accuracy of calculated isotopic inventories, and the associated reactivity uncertainty.

The RCA data for Bugey consist of samples taken from the fuel rod axial center for one rod removed from each of two different assemblies: rod H09 of assembly FGA54 and rod K11 of assembly FGC53. The assemblies are PWR assemblies without burnable poison assemblies. Isotopic samples were analyzed for the major actinides and cesium for rod H09, and for the major actinides, cesium, neodymium, americium and curium for rod K11. <sup>133</sup>Cs and <sup>145</sup>Nd are burnup credit isotopes, and the americium isotopes (especially <sup>241</sup>Am) are minor actinides that are important to burnup credit. Curium is not considered in these burnup credit calculations and was omitted from the confirmatory calculations. The lack of <sup>148</sup>Nd data for rod H09 prevented analysis of this RCA sample at this time.

The RCA data for Gravelines consists of samples taken from the fuel rod axial center for two rods, G07 and G11 of assembly FF06E2BV (E2), and one rod, J07, of assembly FF06E3BV(E3). The assemblies are PWR assemblies without burnable poison assemblies. All three isotopic samples were analyzed for the major actinides, americium, neodymium, cesium, curium, <sup>154</sup>Eu, <sup>106</sup>Ru, and <sup>144</sup>Ce. Curium, Eu, Ru, and Ce were omitted from the confirmatory calculations.

Analyses with the SAS2H computer code sequence showed reasonable agreement between calculated and measured isotopic contents for actinides, but unsatisfactory agreement for the minor actinides, neodymium, and cesium. The TRITON computer code sequence provides a much more accurate representation of the neutron flux in the sample rods, and only TRITON analyses are presented in this report. The TRITON calculations showed that the agreement between calculated and measured actinide data for Bugey has typical accuracy, with a  $\Delta k$  value of about one quarter of one percent. The agreement between calculated and measured actinide data for Gravelines showed excellent agreement, with a  $\Delta k$  value of a quarter of one percent for all three samples . In order to evaluate the fission product isotopic contents, the <sup>148</sup>Nd isotope was checked to insure that the calculated and measured contents are essentially equal. <sup>148</sup>Nd is a direct fission yield (no radioactive decay chains) and has a small neutron cross section, so it provides a very accurate measure of burnup. Comparisons between calculated and measured data for Bugey and Gravelines are provided in Tables 5-2 and 5-3, respectively.

Inspection of the data provided in Tables 5-2 and 5-3 show good agreement for the major actinides, and the  $k_{eff}$  values for measured and calculated major actinides and americium are provided in Tables 5-2 and 5-3. Note that the fission products are not included in these calculations because of the difficulties in assuring convergence of the small  $\Delta k$  values involved. (Convergence of a direct perturbation calculation requires substantial computer resources for this reason.)

Agreement between calculations and measurements of the cesium isotopes were not as satisfactory, resulting in nearly 30 percent differences for the important <sup>133</sup>Cs isotope. The data

#### Fission Product Bias Confirmation

for <sup>133</sup>Cs from all RCA samples is very consistent, ranging from 25 to 30 percent, which suggests that there is a real disagreement between the calculated and measured data. Note that the calculated values are less than the measured values, which is conservative since <sup>133</sup>Cs is a neutron absorber. Thus no action is needed – the computer code sequence simply underpredicts the content of the <sup>133</sup>Cs absorber in burnup credit calculations, and some of the benefit of this isotope is lost. This would not be the case, of course, if the computer calculations both underpredicted and overpredicted the measured isotopic content. The Bugey and Gravelines RCA data thus confirm the three americium isotopes, <sup>145</sup>Nd, and <sup>133</sup>Cs.

#### Table 5-2

#### Radiochemistry Assay Results for Bugey [(Calculated – Estimated)/Estimated]

Bugey Rod K11						
38.7			(C-E)/E			
GWd/MTU	RCA	TRITON	(Percent)			
u234	-1.162E-04	-1.27E-04	9.52			
u235	-5.512E-03	-5.16E-03	-6.30			
u236	-3.714E-03	-3.83E-03	3.07			
u238	-8.562E-01	-8.56E-01	0.00			
nd237	-4.655E-04	-4.57E-04	-1.94			
pu238	-1.831E-04	-1.89E-04	3.20			
pu239	-4.608E-03	-4.94E-03	7.12			
pu240	-2.290E-03	-2.40E-03	4.80			
pu241	-9.924E-04	-1.02E-03	2.87			
pu242	-6.247E-04	-6.68E-04	6.99			
am241	-3.591E-04	-3.54E-04	-1.52			
am242m	-8.764E-07	-7.94E-07	-9.37			
am243	-1.307E-04	-1.57E-04	19.88			
nd145	-6.906E-04	-6.96E-04	0.72			
nd148	-3.905E-04	-3.90E-04	-0.24			
cs133	-1.683E-03	-1.19E-03	-29.01			
cs134	-3.117E-05	-2.02E-05	-35.23			
cs135	-5.216E-04	-4.11E-04	-21.11			
cs137	-1.605E-03	-1.14E-03	-29.30			
Reactivity Comparison						
RCA Experiment TRITON Calculated						
1.07841	k∞	1.081				
0.00016	σ	0.00018				
	∆k/k <sub>RCA</sub>					

0.0024

Note: Negative sign in front of RCA and TRITON values indicate that the values are expressed in gram or gram fractions (software artifact).

## Table 5-3 Radiochemistry Assay Results for Gravelines [(Calculated – Estimated)/Estimated]

	Gravelines Rod G07 Gravelines R		Rod G11		Gravelines Rod J07						
38.2	isotopic CC	mparison	(C-E)/E	51 0	isotopic co	mparison	(C-E)/E	60.7	isotopic Co	mparison	(C-E)/E
GWd/MTU	RCA	TRITON	(Percent)	GWd/MTU	RCA	TRITON	(Percent)	GWd/MTU	RCA	TRITON	(Percent)
u234	-2.082E-04	-2.067E-04	-0.75	u234	-1.842E-04	-1.74E-04	-5.48	u234	-1.542E-04	-1.56E-04	1.24
u235	-1.256E-02	-1.256E-02	-0.07	u235	-7.711E-03	-7.70E-03	-0.11	u235	-4.953E-03	-4.96E-03	0.21
u236	-5.091E-03	-5.105E-03	0.27	u236	-5.785E-03	-5.76E-03	-0.44	u236	-6.019E-03	-6.01E-03	-0.14
u238	-8.472E-01	-8.472E-01	0.00	u238	-8.479E-01	-8.48E-01	0.00	u238	-8.475E-01	-8.48E-01	0.00
pu238	-1.826E-04	-1.608E-04	-11.94	pu238	-3.550E-04	-3.17E-04	-10.62	pu238	-5.320E-04	-4.81E-04	-9.61
pu239	-5.524E-03	-5.588E-03	1.17	pu239	-5.451E-03	-5.71E-03	4.71	pu239	-5.306E-03	-5.65E-03	6.52
pu240	-2.105E-03	-2.083E-03	-1.06	pu240	-2.653E-03	-2.66E-03	0.15	pu240	-2.970E-03	-3.01E-03	1.28
pu241	-1.173E-03	-1.105E-03	-5.81	pu241	-1.475E-03	-1.43E-03	-2.89	pu241	-1.535E-03	-1.52E-03	-1.26
pu242	-4.407E-04	-4.241E-04	-3.78	pu242	-8.279E-04	-7.97E-04	-3.73	pu242	-1.173E-03	-1.14E-03	-3.18
am241	-6.263E-04	-5.964E-04	-4.77	am241	-2.318E-04	-2.09E-04	-9.75	am241	-7.108E-04	-7.20E-04	1.29
am242m	-7.039E-07	-7.357E-07	4.53	am242m	-1.363E-06	-1.25E-06	-8.44	am242m	-1.858E-06	-2.01E-06	8.15
am243	-8.201E-05	-8.780E-05	7.05	am243	-2.177E-04	-2.14E-04	-1.80	am243	-3.119E-04	-3.51E-04	12.42
nd145	-7.318E-04	-7.302E-04	-0.22	nd145	-9.151E-04	-9.19E-04	0.37	nd145	-1.055E-03	-1.05E-03	-0.39
nd148	-3.938E-04	-3.880E-04	-1.47	nd148	-5.226E-04	-5.18E-04	-0.90	nd148	-6.335E-04	-6.22E-04	-1.79
cs133	-1.716E-03	-1.218E-03	-29.02	cs133	-2.174E-03	-1.55E-03	-28.64	cs133	-2.484E-03	-1.79E-03	-27.99
cs134	-5.463E-05	-3.141E-05	-42.50	cs134	-1.179E-04	-7.09E-05	-39.82	cs134	-1.372E-04	-8.40E-05	-38.79
cs135	-5.733E-04	-4.060E-04	-29.18	cs135	-7.726E-04	-5.47E-04	-29.18	cs135	-9.395E-04	-6.68E-04	-28.88
cs137	-1.734E-03	-1.180E-03	-31.92	cs137	-2.354E-03	-1.60E-03	-31.90	cs137	-2.765E-03	-1.89E-03	-31.71
	Reactivity (	Comparison	I		Reactivity C	Compariso	n		Reactivity C	Comparisor	า
RCA Expe	riment	<b>TRITON Ca</b>	lculated	RCA Expe	eriment	TRITON C	alculated	RCA Expe	eriment	TRITON Ca	alculated
1.22308	k∞	1.22485		1.1626	k∞	1.16848		1.07678	k∞	1.08725	
0.00018	σ	0.00018		0.00018	σ	0.00018		0.00017	σ	0.00017	
	∆k/k <sub>RCA</sub>				∆k/k <sub>RCA</sub>				∆k/k <sub>RCA</sub>		
	0.0014				0.0051				0.0097		

Note: Negative sign in front of RCA and TRITON values indicate that the values are expressed in gram or gram fractions (software artifact).

## 5.3 Fission Product Cross-Section Data

Limited fission product cross-section data is available, including several <sup>149</sup>Sm experiments with 4.738 wt% enriched fuel rods [NEA 2009], fission product benchmark critical experiments and the six French fission product critical experiment series [Anno 2001].

A confirmatory calculation of fission product reactivity uncertainty could be performed for <sup>149</sup>Sm because benchmark critical experiments using <sup>149</sup>Sm are available [NEA 2009 LEU-COMP-THERM-050]. The benchmark critical experiments for <sup>149</sup>Sm consist of a small central tank of <sup>149</sup>Sm solution surrounded by a larger tank containing UO<sub>2</sub> driver rods enriched to 4.738 wt%. A series of 11 experiments were performed, varying the <sup>149</sup>Sm concentration and the number of driver rods so that the critical water height also varied. Two additional experiments without neutron absorber in the small central tank were also performed, and five experiments were performed with dissolved natural boron in the central tank solution to verify the experimental conditions.

Confirmation of the Cs, Rh, and Sm isotopes can also be provided by a series of pseudo-fission product experiments performed in Japan using the STACY critical experiment facility [JAEA 2009]. These experiments are intended to support reprocessing using burnup credit with fission products and contain 6 percent uranyl nitrate solution to represent partly dissolved fuel. In this critical experiment set, the calculated/experimental ratio for <sup>133</sup>Cs is 1.06 or less, which confirms the 6 percent uncertainty used in this report. Similarly, the calculated/experimental ratio for <sup>103</sup>Rh is 1.02 or less, indicating that the 10 percent uncertainty used in this report is quite conservative. Elemental Cs, Rh, and Sm were used in the experiments, hence the term "pseudo-fission product". For Cs and Rh, the fission product of interest is the naturally occurring isotope, and the C/E results are directly applicable to the spent fuel form.

For samarium, the isotopic mixture of samarium isotopes in spent fuel is different from fresh fuel. In spent fuel with a fresh fuel enrichment of 4.0 wt% and a burnup of 40 GWd/MTU, <sup>149</sup>Sm (31.5 percent of total interaction rate) and <sup>151</sup>Sm (53.1 percent) dominate the total neutron interactions. In elemental samarium, <sup>151</sup>Sm does not occur, and <sup>149</sup>Sm (98.6 percent) dominates the total neutron interactions. Thus the pseudo-fission product experiment for samarium essentially represents the behavior of <sup>149</sup>Sm alone. The C/E results for samarium are 1.04 or less, and confirm the 10 percent uncertainty for <sup>149</sup>Sm used in this report.

For europium, <sup>153</sup>Eu dominates (86.5 percent) in spent fuel while <sup>151</sup>Eu (96.3 percent) dominates the total neutron interactions in the elemental form. Thus the pseudo-fission product experiment for europium essentially represents the behavior of <sup>151</sup>Eu alone. The C/E results for europium are 0.99 to 1.00, and confirm the 2 percent uncertainty for <sup>151</sup>Eu used in this report.

The four isotopes confirmed by the STACY critical experiments contribute 30 percent of the total reactivity worth of the 16 fission product and minor actinide isotopes evaluated in this report.

## **6** LOADING OPERATIONS AND BURNUP MEASUREMENTS

### 6.1 Present Requirements for Burnup Measurement

U.S. NRC Interim Staff Guidance - 8, Rev. 2 [ISG-8] ties acceptance of a burnup credit methodology to a verification by measurement of the burnup of each assembly before loading, with the requirement to adjust the verified reactor record burnup value by a combination of the uncertainties in the reactor value and the measurement. This approach is at odds with plant operators' contention that measurements represent an unnecessary detriment, in terms of impact on spent-fuel pool operations and costs, to the implementation of burnup credit, given

- 1. The very low probability of transportation accidents with the potential for re-flooding of the cask cavity, and
- 2. The very low conditional probability of a critical configuration assuming that re-flooding occurs.

### 6.2 Consequences of Misload with Fresh Fuel

The need for a burnup measurement for spent fuel prior to loading a storage or transport cask is dependent upon the possible consequences of a misload event. The consequences of a misload with fresh fuel could be significant, but administrative measures can be taken to insure that fresh fuel is not misloaded into a burnup credit cask. Further, an audit by independent personnel can be performed to determine the positions of fresh fuel assemblies prior to releasing a loaded cask from the spent fuel pool area. This audit is possible because there are only a limited number of locations for fresh fuel: the new fuel vault, the spent fuel pool, or the re-loaded reactor. The characteristics and positions of new fuel in a reactor core are established with a startup physics experiment, so a fresh assembly that was inadvertently left out of the core would be discovered. The new fuel vault is accessible without difficulty and any new fuel present can be inventoried. The final location, the spent fuel pool, would not normally contain fresh assemblies after the reactor restart unless there was a problem with an assembly, an expensive and important event, so the locations of new fuel in the pool are known and can be verified by visual examination. Therefore, misloading a burnup credit cask with fresh fuel and removing the cask from the loading area is not credible because it is preventable and detectable with proper audit procedures.

Procedures for a fresh fuel audit could be reactor specific due to differences in fuel handling procedures and reactor design. Reactor-specific procedures would allow generic procedures to be "tuned" to match the operating environment of a given reactor.

## 6.3 Consequences of Underburned Fuel

The possibility exists that some small fraction of the spent fuel loaded into a burnup credit cask could posses a lower, more reactive, burnup than specified by the cask burnup credit loading curve. Underburned fuel is spent fuel that has not achieved the burnup obtained from the reactor core-follow calculations used to plan reactor operations and reloading schemes. A study was performed to determine the magnitude of the effect of such a misload relative to the arbitrary margin for criticality safety (five percent). Results from this study are illustrated in Figure 6-1 that shows the increase in cask reactivity for a 32-PWR capacity burnup credit cask caused by the addition of substantially underburned assemblies. [EPRI 2003] The minimal consequences of loading a single underburned fuel assembly can be incorporated into the burnup credit loading curve if desired.



Figure 6-1 Cask Reactivity vs. Number of Misloads of Substantially Underburned Spent Fuel

# **7** SUMMARY

A practical methodology for burnup credit, based upon the NRC's ISG-8 Rev 2, has been developed. It supports enrichments up to 5.0 wt% <sup>235</sup>U initial and burnup beyond 50 GWd/MTU. A total of 14 actinides and 16 fission products are addressed. Burnable absorbers, both removable and fixed, are supported. Axial burnup profiles, both conservative and mean value, are provided for so that users do not have to select a particular profile from a database and then defend its conservatism. Both PWR and BWR axial burnup profiles and isotopic depletion models are available. Standard isotopic depletion codes, such as TRITON and CASMO, and standard criticality codes, such as MCNP and KENO, are employed. Isotopic depletion parameters are suggested based upon ORNL and Yucca Mountain reports.

The validation methodologies are all based upon standard methodologies, including extensions beyond the traditional RCA isotopic and critical experiment benchmark approaches. The classical "Propagation of Uncertainties" methodology is used to transform documented uncertainties for fission product isotope production and cross sections into equivalent reactivities for burnup credit validation. Confirmation of reactivity bias factors for fission products is provided by available data, including Commercial Reactor Criticals which validate the isotopic production and cross-sections together, and/or separate fission product RCAs and critical experiments.

Thus, all of the methods and validation required for full burnup credit are available. The methods are well-established and validation values for the ANSI/ANS 8-27-2008 standard for burnup credit are all available, based upon existing data.

Evaluations of the bias correction factors were performed with the results shown in Table 7-1.

Summary

	Bias Correction	PWR	BWR
Actinide-Only	$\Delta k_{I-Act}$	0.011	
	$\Delta k_{c-Act}$	0.008	
Fission Product	$\Delta k_{\text{I-FP(Fission Yield)}}$	0.0046	0.0044
	$\Delta k_{I-FP(Radioactive Decay)}$	0.000375	0.000375
	$\Delta k_{I-FP(Capture)}$	0.0058	0.0027
	$\Delta k_{_{C-FP}}$	0.017	0.015

#### Table 7-1 Bias Correction Factors

Note that the  $\Delta k$  values for fission products and minor actinides are of the same order of magnitude as the values for Actinide-Only burnup credit, even though the uncertainties of the fission product and minor actinide isotopes are substantially larger. This is because the reactivity worth fission products is much less than the reactivity worth of Actinide-Only burnup credit.

Using the equations shown in the "Propagation of Errors" (Section 2), the overall PWR bias correction factors for actinide-only and full, actinide plus fission product, burnup credit are thus  $\Delta k_{Act} = 0.014$  and  $\Delta k_{Act+FP} = 0.037$ . These values must be added to  $k_{eff,calculated}$  values for conservatism.

By comparison, the bias and uncertainty from the Commercial Reactor Critical state points is an underprediction of 0.0143, which is about half of the  $\Delta k_{Act+FP}$  value, so the methodology described in this Topical Report is conservative by a factor of 2<sup>1</sup>/<sub>2</sub> percent  $\Delta k$ .

This methodology also provides a means for applying recent fission product data, which are more suited to a confirmatory role than a means of calculating FP bias correction factors. One such resource is the STACY [JAEA 2009] pseudo-fission product experiments, which are intended to support reprocessing using burnup credit with fission products. These experiments confirm the cross-section uncertainties for <sup>133</sup>Cs, <sup>103</sup>Rh, <sup>149</sup>Sm, and <sup>151</sup>Eu used in this report. Together, these isotopes account for 30 percent of the total reactivity worth of the 16 fission products and minor actinides.

Radiochemistry data from the Bugey and Gravelines reactors are also suited to a confirmatory role. Four RCAs confirm the <sup>133</sup>Cs, <sup>145</sup>Nd, <sup>241</sup>Am, <sup>242m</sup>Am, and <sup>243</sup>Am isotopic uncertainties. These RCA samples also add to the Actinide-Only validation of the major actinides.

The assigned burnup value is obtained from reactor records, and a misload analysis shows that the misloading of one or more underburned assemblies does not compromise safety. Reactorsite-specific audit procedures can prevent and detect the misloading of a fresh assembly prior to completion of cask loading. The minimal consequences of loading a single underburned fuel assembly can be incorporated into the burnup credit loading curve if desired. Thus safety can be assured for burnup credit loading operations.

This report therefore concludes that the obstacles for implementation of fission product or "full" burnup credit have solutions, and full burnup credit can be safely implemented in future storage and transport cask systems.

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<sup>&</sup>lt;sup>3</sup> SCALE 6.0 includes the TSUNAMI and TRITON computer code sequences. The SAS2H computer code sequence was included in SCALE 5.1 and older versions.

# **A** BURNUP CREDIT – U.S. INDUSTRY GOALS

A set of goals for burnup credit were described in [EPRI 2002]. A practical burnup credit methodology must be focused on achievable goals that add value to the use of burnup credit.

- 1. Be practical, based on the U.S. Nuclear Regulatory Commission's Interim Staff Guidance 8 (ISG-8), as revised
- 2. Support initial enrichments up to 5.0 wt% U-235 without a loading offset
- 3. Include burnable absorbers
- 4. Provide standard axial burnup profiles
- 5. Allow for various cooling times
- 6. Include the addition of fission products
- 7. Allow for extension to high burnup (> 40 GWd/MTU)
- 8. Allow burnup measurements to be replaced by reactor records
- 9. Provide for BWR applications
- 10. Be based upon standard (consensus) depletion parameters
- 11. Provide standard benchmark methodologies
- 12. Support standard isotopic depletion codes

The first goal simply states that a burnup credit methodology must be practical to be of value to the U.S. commercial nuclear industry. An element of practicality is, to the greatest possible extent, to build upon accepted NRC position. Further, a practical methodology must be based upon existing data available to the industry.

The second goal, to increase the enrichment limit of ISG-8 Rev 1 to 5.0 wt% and remove loading offsets were achieved in Revision 2 of ISG-8.

The third goal, inclusion of burnable absorbers, has been accomplished through a number of studies performed at ORNL, especially [Wagner 2000].

The forth goal, provision of standard axial burnup curves, has been addressed in several ways. A reference that provides a database of axial profiles [YAEC 1997] was expanded and analyzed by [Scaglione 2003] to provide probabilistically conservative profiles and mean profiles.

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The fifth goal, allowance for various cooling times, has been addressed by the presentation to the Nuclear Waste Technical Review Board (NWTRB) "Direct Disposal of Dual-Purpose Canisters – Options for Assuring Criticality Control" [Machiels 2009].

The sixth goal, the addition of fission products, is key to the value of burnup credit. The number of fission products strongly influences the value of burnup credit, with  $k_{eff}$  decreasing as more isotopes are included. EPRI has selected the Principal Isotope list employed by the Yucca Mountain project [CRWMS M&O 1998] with the addition of Cs-133 that is used by the French program [Anno 2001].

The seventh goal, the extension of burnup credit to higher burnup, has been accomplished through ISG-8 Rev 2, which allows up to 50 GWd/MTU. Higher burnup do not appear to present any special problems and are addressed in this report by having bias/uncertainty terms that are burnup dependent.

The eighth goal, the use of Reactor Records for the assigned burnup of a spent fuel assembly, has been addressed [EPRI 2003].

The ninth goal, application of burnup credit to BWRs is addressed in this report. BWR burnup profiles are provided in [Huffer 2005] based upon the Yucca Mountain database.

The tenth goal, the provision of consensus isotopic depletion parameters has been provided by the selection of values from NUREG/CR-6761.

The eleventh goal, the use of standard benchmark methodologies, is provided for by the HTC experiments [NUREG/CR-6979] for uranium and plutonium isotopes and [Wells 2006] for fission products.

The twelfth goal, use of standard isotopic depletion codes, is shown by the use of TRITON (replacing SAS2H) in the EPRI methodology.

Thus, all of the industry goals for burnup credit can now be met. Improvements to the technical means for validation of burnup credit isotopics and fission product cross sections may be forthcoming based upon additional data and enhancements of the sensitivity tools of SCALE. Modifications to the methodology itself are at the discretion of the user.

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