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Review of Technical Issues Related to Predicting Isotopic Compositions and Source Terms for High-Burnup LWR Fuel

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Prepared by
I.C. Gauld, C.V. Parks

Oak Ridge National Laboratory
Managed by UT-Battelle, LLC
Oak Ridge, TN 37831-6370

D.D. Ebert, NRC Project Manager

Prepared for
Division of Systems Analysis and Regulatory Effectiveness
Office of Nuclear Regulatory Research
U.S. Nuclear Regulatory Commission
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ABSTRACT

This report has been prepared to review the technical issues important to the prediction of isotopic compositions and source terms for high-burnup, light-water-reactor (LWR) fuel as utilized in the licensing of spent fuel transport and storage systems. The current trend towards higher initial ^{235}U enrichments, more complex assembly designs, and more efficient fuel management schemes has resulted in higher spent fuel burnups than seen in the past. This trend has led to a situation where high-burnup assemblies from operating LWRs now extend beyond the area where available experimental data can be used to validate the computational methods employed to calculate spent fuel inventories and source terms. This report provides a brief review of currently available validation data, including isotopic assays, decay heat measurements, and shielded dose-rate measurements. Potential new sources of experimental data available in the near term are identified. A review of the background issues important to isotopic predictions and some of the perceived technical challenges that high-burnup fuel presents to the current computational methods are discussed. Based on the review, the phenomena that need to be investigated further and the technical issues that require resolution are presented. The methods and data development that may be required to address the possible shortcomings of physics and depletion methods in the high-burnup and high-enrichment regime are also discussed. Finally, a sensitivity analysis methodology is presented. This methodology is currently being investigated at the Oak Ridge National Laboratory as a computational tool to better understand the changing relative significance of the underlying nuclear data in the different enrichment and burnup regimes and to identify the processes that are dominant in the high-burnup regime. The potential application of the sensitivity analysis methodology to help establish a range of applicability for experimental data in code validation is also discussed and demonstrated.

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1 INTRODUCTION

The present trend in the commercial nuclear power industry is towards irradiating nuclear fuel to significantly higher discharge burnup through the use of higher initial ^{235}U enrichments, more complex fuel assembly designs that incorporate variable enrichments and burnable poisons, and more complex core loading and fuel management schemes. This trend has led to a situation where spent nuclear fuel (SNF) from many operating light-water reactors (LWRs) now extends beyond the area where experimental data are widely available to validate the computational methods employed to calculate spent fuel source terms for transportation and storage applications. Limited isotopic assay measurements on high-burnup fuel for pressurized-water reactors (PWRs) and boiling-water reactors (BWRs) are in progress or have been proposed. However, even assuming availability of these data in the future, there will likely be insufficient source-term-related data for high-burnup fuel in the near term to enable a comprehensive and reliable assessment of the calculational bias and uncertainty based on these measurements alone.

In view of the limited quantity of experimental data in the high-burnup regime that are likely to become available in the near future, it is important to understand the technical issues related to the analysis of high-burnup spent fuel and identify potential shortcomings of present analysis methods and/or data when applied to the high-burnup regime. This report has been prepared to review the relevant background information and technical issues important to the prediction of isotopic compositions and associated source terms utilized for licensing of spent fuel transport and storage systems for high-burnup LWR fuel. An understanding of the technical issues and challenges provides a basis for evaluating the merits and limitations of current analysis methods and nuclear data as applied to safety evaluations involving high-burnup fuel. Areas where further study may be warranted in order to reduce the calculational bias and uncertainty in spent fuel inventory predictions and source-terms characteristics are reviewed.

Section 2 of this report summarizes the publicly available experimental data that have been used to validate spent fuel isotopic inventory and source-term calculations in the United States and reviews the additional high-burnup experimental data that may become available in the near term. Section 3 reviews the generic issues and technical challenges in predicting isotopic compositions in high-burnup fuel as they relate primarily to the uncertainties in the key nuclear data and input variables that govern depletion and decay phenomena. Section 4 describes the high-burnup issues as they relate specifically to thermal, shielding, and subcriticality analyses for the licensing of spent fuel transport and storage systems. The characteristics that distinguish high-burnup from conventional-burnup spent fuel are reviewed in terms of decay-heat generation, gamma-ray sources, and neutron sources. Section 5 summarizes the high-burnup issues that need to be further investigated and resolved, and identifies areas where improvements in the computational methods and data may be warranted. Appendix A describes and demonstrates a sensitivity-based methodology proposed as a means of identifying the governing phenomena and associated key parameters and nuclear data important to predicting isotopic concentrations for high-burnup fuel. This methodology is being pursued as a tool to formally establish the range of application of experimental data and to help quantify the calculational biases and uncertainties in the high-enrichment and high-burnup area where there is likely to be a limited amount of experimental data to support code validation in the near term. An illustrative application of this methodology to evaluate the applicability of high-burnup spent fuel from the H. B. Robinson reactor for isotopic analysis is presented in Appendix B.

In a parallel effort to support the sensitivity work, ranking studies to identify the dominant nuclides and trends in the nuclide importance in high-burnup spent fuel have been performed and documented.[1] These studies relate to the areas of criticality safety, decay-heat generation, and radiation source terms for cask transport and interim storage applications, and provide some guidance on proposed validation approaches for the high-burnup regime.

1.1 Background

Validation of the computational methods and associated nuclear data currently used to predict isotopic compositions and radiological decay characteristics of SNF for transportation and storage applications in the United States has relied primarily on chemical assay data and decay heat measurements for the range of enrichments, assembly designs, and discharge burnups seen in the past. Examples of such validation studies from point-depletion methods, most commonly used to characterize SNF, are provided in Refs. 2–6. The recent trend towards the use of higher-initial-fuel enrichments has resulted in significantly higher-discharge burnups that extend beyond the available experiment data and place increasing reliance on the accuracy of computer code predictions. However, the validity of fuel characterization predictions established for the conventional burnup fuel should not be assumed to extend beyond the limits established by experimental benchmark data. In the absence of experiment data to confirm calculations, additional safety margins may need to be applied.

The limited quantity of experimental data in the high-burnup regime upon which code and data accuracy can be benchmarked has had a direct impact on the licensing of facilities used for transporting and storing spent fuel. Because of the lack of a sufficient isotopic assay database for high-burnup fuel, the Nuclear Regulatory Commission (NRC) has specified in Interim Staff Guidance 8 (ISG8)[7] that actinide-only burnup credit be limited to PWR fuel with a maximum assembly-average burnup of 40 GWd/t. Similarly, the limited amount of measured decay-heat data for high-burnup fuel in the United States has restricted the range of the NRC Regulatory Guide 3.54 for spent fuel heat generation to a maximum of 50 GWd/t for PWR fuel, and 45 GWd/t for BWR fuel.[8]

High-burnup fuel can present a number of technical challenges to the calculational methods and the nuclear data used to predict isotopic compositions and radiological decay properties. Obviously, the basic physics that govern the depletion and decay processes do not change as the assembly burnup is increased. However, there can be changes in the relative importance of the nuclear processes (nuclide production and decay chains) and data in different enrichment and burnup regimes that can directly impact the accuracy of predicted SNF characteristics. Increased uncertainty may also be introduced by approximations in the assembly modeling used to prepare accurate cross sections for the more complex assembly designs typically used with higher-enrichment fuel (e.g., variable-enrichment rods, integral burnable absorbers, and burnable poison rods) and the changes in the fuel characteristics associated with changing isotopic compositions.

One of the major contributors to the uncertainty in the calculated radiological properties or source terms in spent fuel can be the uncertainty in the calculated isotopic inventory. The accuracy of the predicted inventories is dependent to a large extent on the accuracy of the nuclear data parameters used to represent the basic neutron transmutation and decay chains and on the completeness of the chains in the depletion analysis model. As the assembly design and operating domains of LWR fuel change, so does the sensitivity of the predicted isotopic inventories to the underlying nuclear data. Any uncertainties in these data can become either more or less important to the prediction of individual nuclide compositions with changing enrichment and burnup. For instance, a preliminary sensitivity study performed in the course of this review indicates that the predicted concentration of ^{235}U is about three times more sensitive to changes in the ^{235}U fission cross section at 60 GWd/t as compared with 30 GWd/t (see Appendix A). Therefore, any error in the ^{235}U cross section will have a significantly larger adverse effect on the predicted ^{235}U inventory at the higher burnup.

The accuracy of predicted spent fuel radiological characteristics that involve aggregate fission products and/or actinides can also be affected by the changing fuel compositions that occur in high-burnup fuel by altering the importance of the individual radionuclides in a particular application. As an example, the contribution from ^{244}Cm to the decay heat for 30-GWd/t fuel after a 5-year cooling represents about 2% of the total. However for 60-GWd/t fuel the contribution from ^{244}Cm dramatically increases to about 20% of the total decay-heat generation. Consequently any potential errors in the predicted concentration of ^{244}Cm or decay data associated with ^{244}Cm will have a much greater effect on the accuracy of the decay-heat prediction at high burnup than lower

burnup, where the contribution from ^{244}Cm is much less. The total neutron source at discharge is also observed to increase nearly exponentially with burnup, but the gamma-ray source increases linearly with burnup.[9] Therefore, the relative importance of the neutron source in shielding analyses of spent fuel cask designs will increase with burnup, and any potential error in the predicted neutron source will take on increasing importance (i.e., larger errors) in the calculation of total dose rate.

1.2 Objectives

The objectives of this report are to provide a review of the technical issues that are important to the accurate prediction of source terms in the high-enrichment and high-burnup regimes and the areas that are most likely to compromise the accuracy of existing computational methods. This review document is intended to provide a brief background and description of the technical issues being explored and their potential importance to high-burnup spent fuel analysis, to describe sensitivity-based analysis methodologies currently being developed to identify the important nuclear data parameters, and to present some preliminary results from the sensitivity techniques being used.

In the future, deficiencies in the current modeling methods will be reviewed with the objective of providing guidance on methods limitations and applicability, and the use of appropriate bounding analysis values where appropriate. Identifying the dominant high-burnup issues and computational areas that would benefit most (e.g., reduced calculational uncertainties) from the upgrading of methods and/or data is a key goal of this task. As part of this effort, a reference document providing the dominant radionuclides important to thermal, shielding, and criticality analysis in the high-burnup regime will be compiled. Areas where methods and data development would reduce calculational uncertainties, and methods to make better use of the limited amount of existing validation data in the high-burnup regime, are also being explored.

2 REVIEW OF EXPERIMENTAL DATA

This section reviews the experimental data currently available for the validation of spent fuel isotopic characterization, decay-heat generation, and neutron and gamma-ray source-term predictions. In addition, potential sources of new data that could become available to support high-burnup fuel validation in the near future are identified. The review of experimental data is important to establishing the assembly design and operating regimes that are adequately represented by experiments, those regimes that are under-represented, and areas where no experimental data exist.

2.1 Isotopic-Assay Data

The spent fuel isotopic assay data that have been used in the most recent and most comprehensive point-depletion code validation efforts in the United States include PWR[2,3,4] fuel assays with a maximum burnup of 46 GWd/t and BWR fuel[5] assays with a maximum burnup of 34 GWd/t. The assay data for PWR fuel contain only one measurement with an enrichment above 3.4-wt % ^{235}U. For BWR fuels the maximum enrichment of the assayed samples is 2.9-wt % ^{235}U. A 1998 review[10] of available assay measurements identified and recommended several sources of public domain data (Yankee Rowe, Obrigheim pellet assays, and Garigliano reactor fuels) that were not used in the most recent validation studies.[2-5] Although these measurements do not extend the enrichment or burnup range covered by the data previously used, the additional data should provide a better statistical interpretation of the code biases and uncertainties in the conventional enrichment and burnup regimes and their incorporation into the validation database is recommended. Several assay experiments identified in Ref. 10 were not recommended for use in code validation (e.g., TMI-2, Genkai-1, Mihama, and Sena), primarily due to either incomplete design and operational data or suspected problems with some of the experimental results.

A summary of the recommended isotopic assay data for PWR and BWR reactor fuels is given in Tables 1 and 2. The tables list the fuel enrichment range, burnup range, the approximate number of usable samples, and the number of total fission products and fission products important to burnup credit, uranium and plutonium isotopes, and curium isotopes (increasingly important to the spent fuel properties in the high-burnup regime) that were measured in each of the experimental programs. Some limited information on the fuel assembly design (presence of integral burnable gadolinium poison rods or burnable poison rods) is also indicated.

The number of existing isotopic assay measurements for BWR assembly designs is significantly less than that currently available for PWR assemblies, both in terms of the number of different assemblies and the number of assay samples available. In addition, fission product measurements, particularly for the important burnup credit fission products, are extremely limited for PWR fuel and are almost nonexistent for BWR fuel. Consequently, the acquisition of data from new programs is seen as important to extending the enrichment and burnup regimes, and providing needed fission product measurements for all regimes.

With the current trend towards higher-enrichment and higher-burnup fuel, the acquisition of additional experimental data to support code applications in this regime is seen as a high priority in the United States and other countries with viable nuclear programs. Currently, several programs are in progress to obtain these data in the United States, and several international programs from which isotopic assay data could be acquired in the near term. Most of these programs are associated with burnup-credit activities. The existing data and potential new sources of data that could be acquired, or will likely become available in the next few years, are also listed in Tables 1 and 2. The programs that could provide sources of new assay data are described briefly in the following sections.

Table 1 Existing recommended sources and potential sources of PWR isotopic assay data

Reactors (Program)	Lattice type	Enrichment (wt % ^{235}U)	Burnup range (GWd/t)	No. of usable samples	Assembly absorbers		No. of nuclides measured[a]		
					BPRs[b]	Gd	F.P.[c]	U + Pu	Cm
Available PWR data									
Calvert Cliffs-1	CE 14 × 14	2.45 – 3.04	18.7 – 46.5	9	X		24/11	9	1
H. B. Robinson-2	W 15 × 15	2.56	16.0 – 31.7	6	X		2/1	7	0
Obrigheim - assembly	14 × 14	3.13	25.9 – 30.3	10			9/0	7	2
Trino Vercelles	W 15 × 15	2.7 – 3.9	3.4 – 36.9	15			6/0	8	2
Turkey Point-3	W 15 × 15	2.56	30.5 – 31.3	5			0	9	0
Obrigheim - pellets	14 × 14	2.83, 3.0	15.6 – 36.9	15			9/0	7	2
Yankee Rowe, I-IV	18 × 18	3.40	1.32 – 32.34	20			0	10	2
Yankee Rowe, V	18 × 18	2.90	7.55 – 14.05	12			0	10	2
Total				92					
Potential PWR data									
TMI-1 (DOE)	W 15 × 15	4.0, 4.65	30 – 51.3	5	X	X	18/14	9	0
H. B. Robinson[e] (NRC)	W 15 × 15	2.9	73 (peak)	N/A[d]	N/A	N/A	N/A	N/A	N/A
REBUS	17 × 17	3.8, 5.0	30 – 60	N/A	N/A	N/A	23/14	6	4
Gosgen[e] (ARIANE)	15 × 15	3.5, 4.1	30 – 59.7	3			31/16	9	4
LWR-PROTEUS	N/A	3.5, 4.1	36 – 82	N/A	N/A	N/A	29/15	9	4
Bugey-3 (Fr)[f]	17 × 17	2.1, 3.1	19 – 38	N/A			14/13	9	>2
Fessemheim (Fr)	17 × 17	2.6, 3.1	27 – 60	N/A	X		14/13	9	>2
Gravelines 2 + 3 (Fr)	17 × 17	4.5	25 – 62	N/A	X		14/13	9	>2
Tihange-1 (Fr)	15 × 15	3.1	10 – 40	N/A			14/13	9	>2
Japan (High-burnup UOX measurements)	N/A	3.8	60.2	2	N/A		39/10	9	4

[a] Approximate number of measured fission products (F.P.), major actinides (U + Pu), and curium (Cm) isotopes for each sample.

[b] BPR = Burnable poison rod.

[c] Second value represents the number of fission products important to burnup credit.

[d] Data not known or not available.

[e] Assay data from reconstituted assemblies (see discussion in Sect. 2.1.2 and 2.1.3).

[f] French PIE Program.

Table 2 Existing recommended sources and potential sources of BWR isotopic assay data

Reactor data (Program)	Lattice type	Enrichment (wt % ^{235}U)	Burnup range (GWd/t)	No. of usable samples	Assembly absorbers BPRs[b]	Assembly absorbers Gd	No. of nuclides measured[a] F.P.[c]	No. of nuclides measured[a] U + Pu	No. of nuclides measured[a] Cm
Available BWR Data									
Cooper	GE 7 × 7	2.94	18.9 – 46.5	6		X	6/1	9	1
Grundremminger	6 × 6	2.53	14.4 – 27.4	8			4/0	8	1
JPDR	6 × 6	2.60	2.2 – 7.0	21			12/0	9	2
Garigliano	8 × 8	2.41	4.2 – 13.8	17			3/0	7	1
Total				52					
Potential BWR Data									
French PIE Program	GE 9 × 9	3.95	20 – 40	N/A[d]	N/A	N/A	14/13	9	>2
Quad Cities-1 (DOE)	N/A	3.0, 3.8	60 – 77	7		X	18/14	9	0
Limerick-1 (NRC)	9 × 9	3.6	54 – 57	N/A	N/A	N/A	N/A	N/A	N/A
LWR-PROTEUS	N/A	~ 4.0	< 75	N/A	N/A	N/A	29/15	9	4
Dodewaard[e] (ARIANE)	6 × 6	4.94	57	1		X	31/16	9	4

[a] Approximate number of measured fission products (F.P.), major actinides (U + Pu), and curium (Cm) isotopes for each sample.

[b] BPR = Burnable poison rod.

[c] Second value represents the number of fission products important to burnup credit.

[d] Data not known or not available.

[f] Assembly contained two MOX fuel rods.

2.1.1 DOE Measurements

Under contract with General Electric and Argonne National Laboratory, the U.S. Department of Energy (DOE) Yucca Mountain Site Characterization Office is seeking to obtain measured assay data for PWR and BWR fuel with burnups greater than those readily available in the United States to support a disposal criticality analysis methodology. These data, obtained from high-burnup fuel assemblies from Three Mile Island Unit 1 (TMI-1) and Quad Cities Unit 1 (QC-1) would extend the burnup range of the existing PWR database beyond 50 GWd/t, and the BWR database beyond 70 GWd/t. Experimental results for some of the fuel samples have been released in draft form, and some reactor physics data are still being compiled. Final results and analyses are expected by March 2001.

2.1.2 NRC Measurements

Two fuel assemblies are being considered for radiochemical isotopic analysis by the NRC to provide additional validation data for high-burnup fuel. The candidate assemblies are from the H. B. Robinson PWR reactor and

Limerick-1 BWR reactor, with maximum burnups of about 70 GWd/t and 57 GWd/t, respectively. A sensitivity-based study of the applicability of the H. B. Robinson samples to the validation of isotopic depletion methods was performed by the Oak Ridge National Laboratory (ORNL) to address concerns over the relatively low enrichment of the fuel (2.9-wt %) with respect to the burnup, and consequently the degree to which these samples would be representative of typical high-burnup fuel. The study suggested these samples would be applicable and valuable to high-burnup fuel validation (see Appendix B). More recent information on the H. B. Robinson fuel, however, indicates that the assemblies were reconstituted to achieve an artificially high burnup. The candidate fuel rods were removed from the original assembly and placed in a second sub-assembly containing fresh fuel with 3.85-wt % enrichment and 10 wt % Gd_2O_3 burnable poison rods, potentially compromising the value of isotopic assay data from this fuel for use in code validation. The reconstitution can dramatically alter the neutronic environment of the candidate assay fuel rods during their exposure in a way that is not representative of that experienced by typical high-burnup commercial reactor fuel. As a result, acquisition of assay data from the H. B. Robinson fuel is no longer *highly* recommended, and further evaluations are required to assess the potential value of these samples to code validation. Although the significant reconstitution of the fuel may preclude use of these samples for formal validation purposes, the assay data may provide a formidable benchmark for multidimensional depletion analysis methods, which are more capable of modeling the assembly reconfiguration.

2.1.3 ARIANE Program

The ARIANE program led by Belgonucleaire has performed extensive isotopic content measurements for many important burnup credit nuclides for MOX and some UO_2 fuel samples. The experimental data relevant to commercial LWR fuel from the ARIANE Program include comprehensive assay measurements of three samples for Gosgen PWR UO_2 fuel with enrichments of about 3.5 and 4.1 wt %, and burnups of between about 30 and 60 GWd/t. Isotopic data are also available for a high-burnup Dodewaard BWR UO_2 sample with an initial enrichment of 4.94-wt % ^{235}U and burnup of approximately 57 GWd/t. This sample (designated DU1) represents the highest enrichment data currently available for BWR fuel. Preliminary code benchmarks using the DU1 sample have been reported.[11] However, the Dodewaard 6 × 6 BWR assembly contained two MOX fuel rods and has been excluded from consideration in this review for this reason. Given the relatively small amount of new data likely to become available for high-burnup BWR fuel, it may be worthwhile assessing the appropriateness of the DU1 UO_2 measurements since the MOX rods were not situated close to the assayed UO_2 rod.

A recent detailed review of the Gosgen assay data found that the higher enrichment samples (GU3 and GU4) were obtained from a fuel rod irradiated in a reconstituted assembly. Consequently, the value of these fuel samples for validating spent fuel isotopic predictions for commercial spent fuel may be limited, and further evaluation of the effects from the reconstitution is needed.

The ARIANE data are currently classified as proprietary and publication of the detailed experimental data will be restricted for a period of two years after completion of the program. Publication of results would be limited to quantities such as the calculated/experiment (C/E) ratios during this period. The ARIANE Program is nearing completion (expected in September 2000), and publication restrictions could be lifted as early as October 2002.

2.1.4 French PIE Program

The French post irradiation examination (PIE) programs offer a potentially large quantity of isotopic assay data for PWR fuel in the high-enrichment and high-burnup range.[12,13] The experimental data are considered to be of high quality with low experimental uncertainties. The data from the French programs are proprietary. The enrichment and burnup regimes are not significantly different from that available from other identified sources, with the notable exception of the data from the Gravelines program that includes assays for 4.5-wt % enrichment fuel with a burnup range from about 25 to 62 GWd/t. These data would significantly extend the enrichment and

burnup range covered by experimental data beyond that provided by the ARIANE or the TMI-1 data. Additional isotopic assays for 3.95-wt % enrichment BWR fuel with burnups up to about 40 GWd/t are currently in progress.

2.1.5 REBUS and LWR-PROTEUS Programs

Isotopic analyses of spent PWR fuel will be performed as part of the REBUS program[14] (Belgonucleaire) and Phase II of the LWR-PROTEUS program[15] (PSI). These programs are proprietary. NRC is currently a participant in REBUS and will have access to the isotopic assay data. The schedule for reporting the commercial UO_2 radiochemical analysis results from REBUS is late 2002 to early 2003. The LWR-PROTEUS Phase II measurements are scheduled for completion in July 2001.

The proposed list of actinides and fission products is similar in both programs and comprehensive in terms of representing the major burnup credit nuclides. REBUS includes PWR fuel with enrichments of 3.8 wt %, 50 -60 GWd/t, and 5.0 wt %, 30 GWd/t. LWR-PROTEUS includes PWR fuel with 3.5 wt % and burnups extending to 82 GWd/t, and 4.1 wt % and 36 GWd/t, and BWR fuel with 4 wt % and up to 75 GWd/t.

2.1.6 Japanese Measurements

Several recent measurements on high-burnup PWR UO_2 samples have also been reported by the Japanese.[16] These data include extensive actinide and burnup-credit fission product isotopic inventory measurements for relatively high-enrichment (3.8-wt %) and high-burnup (62-GWd/t) fuel and would be a valuable addition to the database. Other measurements involving destructive assays of PWR with enrichments up to 4.5-wt % ^{235}U and 40 to 45 GWd/t are reportedly in progress. The availability of these measured data is unknown and is currently being explored.

2.1.7 Summary of Experimental Assay Data

In general, the available isotopic assay data correspond to older fuel assembly designs and are limited to 40 GWd/t and 3.5 wt %. Much of the available isotopic data are for the major actinides (uranium and plutonium), whereas the more recent measurements generally include extensive fission product data, particularly for those fission products important to burnup-credit programs. Historical fuel assembly burnup data up to 1993 and projected data up to 2005 were obtained from the LWR Characteristics Database to estimate current and near-term experimental data needs. The results are illustrated in Figure 1 (each data point represents a group of more than 100 fuel assemblies) and indicate the typical maximum discharge burnups likely to be encountered in normal reactor operations. The data for PWR and BWR assemblies are very similar and suggest that for 5-wt % fuel the expected discharge burnup will be about 65 GWd/t.

Figures 2 and 3 illustrate the enrichment and burnup regimes covered by the available assay measurements for PWR and BWR fuels respectively (from Table 1 and Table 2), and show the areas represented by potential new sources of data. Even with the new data currently being measured or planned for measurement, the number of chemical assays for fuel in the high-burnup regime is significantly less than that in the more conventional (lower)-burnup regime and a statistical analysis of code validity based solely on the assay data in this regime could prove challenging. Other options that can provide a technical basis for establishing predicted calculational bias over extended design and operating domains, in conjunction with new experimental data as they become available, are currently being explored. Sensitivity-based methods (discussed in Appendix A) are currently being utilized as a means of identifying the key parameters and governing phenomena that are important to predicting isotope compositions and radiological decay properties in high-burnup spent fuel and to better understand code behavior and the calculational bias associated with high-burnup spent fuel. Ultimately the aim is to combine the limited amount of high-burnup assay measurements likely to become available in the near term with sensitivity and

uncertainty methods to provide a reliable and relevant approach to establishing and extending the range of application of the available experimental data to support code validation.

2.2 Decay-Heat Generation Data

The available decay-heat measurements on spent fuel assemblies in the United States extend to a maximum of about 3.4-wt % ^{235}U enrichment and 40 GWd/t for PWR fuel, and a maximum of 2.5-wt % ^{235}U and 28 GWd/t for BWR fuel. A summary of the experimental decay-heat data for the Point Beach 2 and Turkey Point 3 PWR fuel assemblies and Cooper BWR assemblies is provided in Table 3. These measurements were used as the primary basis for establishing the range of application and the uncertainties and biases for the NRC decay heat Regulatory Guide 3.54 (Ref. 8). The data listed in Table 3 represent a relatively small sampling of the decay heat data publicly available.[17] However, the data in Table 3 have been reviewed and are judged as high quality with relatively well documented assembly design and reactor operating history data that are necessary for code validation studies.

The enrichment and burnup regime represented by the decay-heat measurements is very similar to that currently available for isotopic assay measurements. However, the assembly cooling times, an important parameter to decay heat predictions, are limited to less than 10 years. The decay heat in this regime is dominated by contributions from a limited number of fission products (e.g., ^{90}Y, ^{137m}Ba, ^{134}Cs). As cooling times extend beyond 10 years, the accuracy of current computational methods becomes increasingly uncertain due to the changing radionuclide importance, and the high level of agreement observed between calculations and experiments[6] should not be assumed to extend beyond the regime covered by measurements. Similarly, some adverse impact on the accuracy of predicted decay-heat levels may be expected due to increased importance of the actinides as the burnup of spent fuel increases. This issue is discussed further in Sect. 4.

The prospects for acquiring additional decay-heat measurements for high-burnup fuel in the near term are limited. No new experiments are known to be either planned or under way within the United States. The Swedish Nuclear Fuel and Waste Management Co., SKB, is conducting calorimetric measurements of decay heat for PWR and BWR spent fuel assemblies in pools at the Swedish Central Interim Storage Facility for Spent Nuclear Fuel, CLAB. As part of the Swedish program, measurements on these same assemblies will be repeated at intervals in the future (several decades), and assemblies of different design may be added to the study. The burnup levels of the Ringhals 2 and 3 (15 × 15 and 17 × 17 design) PWR assemblies measured to date in the CLAB facility range from 19.7 to 51 GWd/t, and decay times vary from about 6 to 19 years. The burnup of the Ringhals (8 × 8) BWR assemblies ranged from 21 to 38 GWd/t, with decay times from about 4 to 15 years.

The Swedish decay-heat data represent a potentially significant extension of the experimental database for the PWR and BWR fuel compared to data currently available in the United States. This extension of the experimental database, combined with the potential for expanding the measurements to higher-burnup fuel, different fuel assembly designs, and the prospect of acquiring data for longer cooling times, would make the Swedish data valuable to code validation efforts.

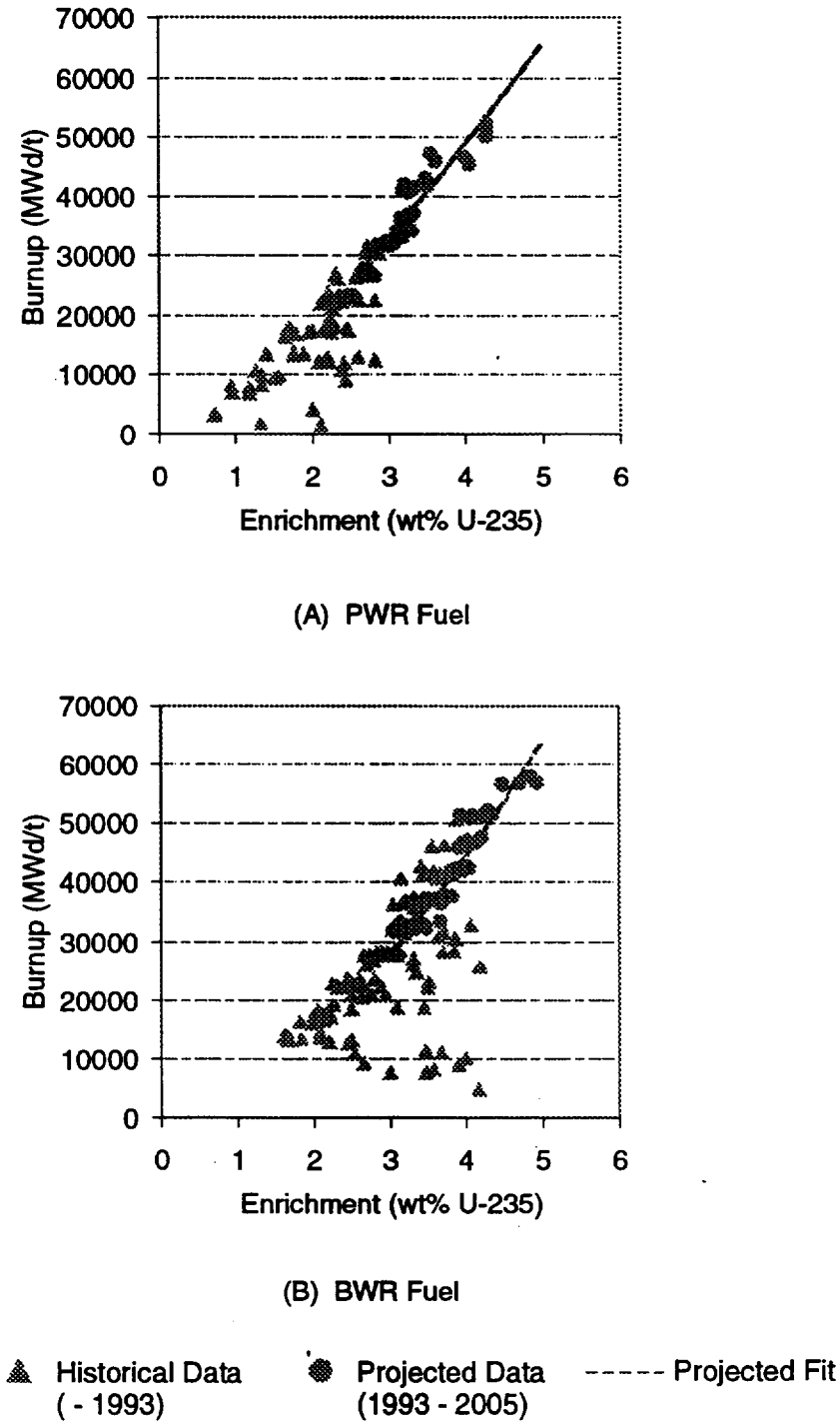

Figure 1 Historical and projected LWR spent fuel discharge burnup

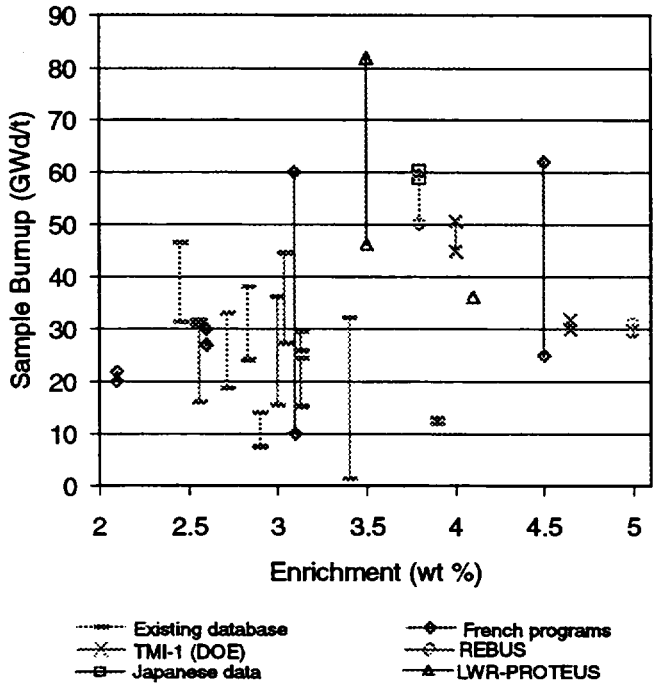

Figure 2 Existing and potential new isotopic assay data for PWR fuel

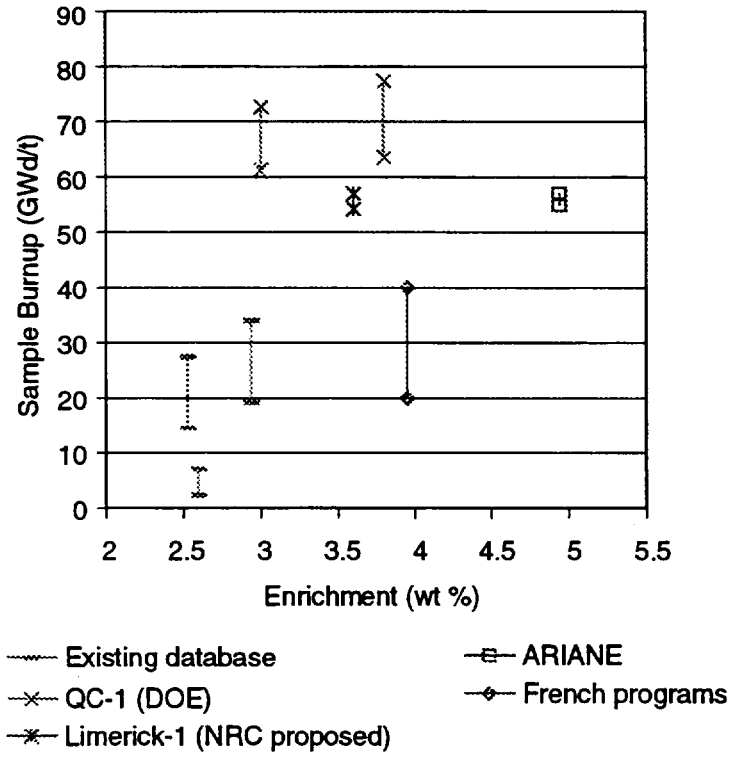

Figure 3 Existing and potential new isotopic assay data for BWR fuel

Table 3 Recommended decay-heat validation data currently in use

Reactors (type)	Lattice type	Enrichment (wt % ^{235}U)	Burnup range (GWd/t)	Cooling time (years)	No. of measure-ments[a]	Assembly absorbers BPRs[b]	Gd
Point Beach Unit 2 (PWR)	W 14 × 14	3.397	31.9 – 39.4	4.5	8		
Turkey Point Unit 3 (PWR)	W 15 × 15	2.559	24.8 – 28.2	2.4 – 5.7	6		
Cooper (BWR)	GE 7 × 7	1.1, 2.5	11.7 – 27.6	2.3 – 7.0	25		X

[a] Represents the decay heat measurements selected for the validation exercise used to support the decay heat Regulatory Guide 3.54.
[b] BPR = Burnable poison rods.

2.3 Radiation Source Data

Relatively few benchmark quality experiments are available in the public domain for validating the accuracy of predicted neutron and gamma-ray source terms used in shielding analyses. The majority of the data currently available consists of measured dose rates outside of shielded facilities (e.g., storage casks). Therefore, these experiments represent integral benchmarks that provide a measure of the combined ability to calculate the isotopic inventories and associated radiation source terms, plus the ability to calculate the dose rates through the shielded configurations. Because the relative importance of the spent fuel isotopic compositions on the predicted dose rate is strongly influenced by the shield configuration (e.g., shield materials, thicknesses, etc.), the applicability of the results for validation may also be restricted to the same types of shielding configurations, materials, assembly operating history, and cooling times as those used in the benchmark.

Pacific Northwest Laboratories (PNL) and Idaho National Engineering Laboratory (INEL) performed a series of neutron and gamma-ray dose rate measurements involving several different cask designs and configurations as part of a cask performance testing program.[18–21] These tests provide integral shielding benchmarks involving 15 × 15 PWR assemblies with enrichments up to about 3.2-wt % ^{235}U and a maximum burnup of 35 GWd/t. Cooling times vary from about 1 to 14 years. A review of the neutron and gamma-ray dose rates reported by PNL and INEL, summarized in Ref. 22, indicates a relatively large and consistent discrepancy between the PNL and INEL dose-rate measurements, with many data points differing from between 20 and 50%. The applicability of these integral benchmarks for explicitly validating the radiation source terms may be difficult to establish, due in part to the apparent large uncertainty in the measurements, and since discrepancies between calculations and measurements may be attributed to errors in the measurements, source-term calculation, or the shielding analysis used to predict the dose rate. However, these experiments represent some of the best benchmarks currently available to validate computational methods used for cask shielding assessments.

Neutron and gamma-ray measurements have also been reported for the French TN-12 transport cask[23] and used in a code-benchmarking exercise sponsored by the Office of Economic Cooperation and Development (OECD), Paris. The gamma-ray measurements involved 3.1-wt % ^{235}U enrichment 17 × 17 PWR fuel assemblies with a burnup of about 17 GWd/t, and cooling times from 1 to 3 years. Neutron measurements involved fuel assemblies having a burnup of about 36 GWd/t and cooling times of about 2 years. The cask design is steel with an outer resin neutron shield, with a capacity for 12 assemblies. The detailed comparisons of the calculated source terms from participants in the benchmark provide a good review of the current computational methods. The agreement

in the predicted total neutron source was generally good; howeve, variations in the contributions from individual actinides (other than ^{244}Cm) were observed, suggesting differences in the nuclear data used in the various calculations. In addition, discrepancies in the gamma-ray source are attributed to differences in the gamma-ray yield data from the various libraries. The low-to-moderate burnups of the assemblies used in this benchmark; however, may limit the applicability of the measurements to validating high-burnup fuel.

Several dose rate measurements have also been reported for Cooper BWR 7 × 7 assemblies[24] with an enrichment of 2.5-wt % ^{235}U and average assembly burnup of about 26 GWd/t, residing in an REA 2023 (steel and lead shield) cask. Again, the relatively low burnup of these assemblies may make it difficult to apply the results to validation of high-burnup fuel.

Experiments performed at the General Electric Morris Operation (GE-MO) facility[25] involved a series of gamma-ray dose rate measurements for PWR and BWR fuel assemblies in air and water. The PWR measurements involved assemblies with initial enrichments from 2.26- to 4.0-wt % ^{235}U and a maximum burnup of 40.2 GWd/t. The BWR assemblies had an enrichment of 2.13 wt % and maximum burnup of only 5.1 GWd/t. The cooling time of these assemblies ranged from about 2 to 10 years. These data are particularly valuable to source-term validation since they utilize relatively high-burnup fuel (PWR assemblies only), and the calculated results do not involve the potentially large uncertainties associated with transport calculations through shielded facilities (e.g., casks). It is recommended that the GE-MO measurements be reviewed to assess their applicability to high-burnup SNF source-term validation.

No other available sources of neutron or gamma-ray validation data have been identified in this review. Given the lack of relevant experimental data in the high burnup regime, alternative validation methods need to be explored. A potential avenue for validating radiation source-term predictions may be through "separate-effects" studies that would combine the accuracy of the predicted concentrations for the dominant radioisotopes in high-burnup fuel (established by radiochemical assay measurements) with separate assessments of the accuracy of the radiation emission yields and spectra for these nuclides. This approach is discussed further in Sect. 4.

3 GENERIC ISSUES RELATED TO ISOTOPIC PREDICTION

One of the major contributors to the uncertainty, in the calculation of radiological properties for spent fuel, can be the uncertainty in the calculated isotopic inventory. This section discusses some of the generic technical issues important to the prediction of isotopic inventories and some of the perceived challenges to the calculational methods and nuclear data as applications tend towards increasingly higher fuel enrichments and burnup.

Calculational uncertainties can be classified as:

1. uncertainty in the actual system parameters being modeled (e.g., assembly power history, initial fuel compositions, unknown reactor conditions, value of the fuel discharge burnup, etc.);
2. uncertainty due to modeling approximations (e.g., geometry approximations, simplification of the power history, etc.); and
3. uncertainty attributed to the calculational methods and data used by the codes.

This review only attempts to address uncertainties related to the calculational methods and data, and modeling approximations as they affect the accuracy of the flux solution used to calculate the neutron reaction rates that define the transmutation process in the depletion analysis.

The accuracy of depletion analysis calculations depends on the completeness of the burnup and decay chains in the calculational model, and on the accuracy of the nuclear data used to predict the neutron reaction (transmutation) and decay rates. The nuclear data required for depletion calculations can be divided into two classes of data: (1) problem-independent data, and (2) problem-dependent data. The problem-independent data include the nuclear decay data such as half-lives, decay modes, branching fractions, energy release per decay (used to calculate decay heat), and fission product yields. These data reflect basic nuclear properties that remain invariant for any fuel design, operating regime, or application. The uncertainties in these data are dependent only on the accuracy of the data evaluations (measurements). The problem-dependent data are primarily the neutron reaction rates that define the transmutation processes such as neutron capture and fission. The reaction rates are, mathematically, the product of the neutron flux and cross section, both of which are energy-dependent quantities. Although the energy dependence of isotopic cross sections does not change (i.e., they are intrinsic properties of the individual isotopes), the neutron flux spectrum may be very problem dependent and time dependent. The assembly design and reactor operating parameters, particularly those parameters that influence the neutron flux spectrum such as material compositions, geometry, poisons, temperatures, etc., can have a significant effect on the reaction rates. Therefore, the production of accurate reaction rate information (reaction rates or effective cross sections) requires that the flux spectrum in the fuel is calculated accurately. Consequently, uncertainties in the transmutation rates can be attributed to both the basic energy-dependent cross-section evaluations (measurements) and the uncertainty in calculating the problem-dependent and burnup-dependent neutron flux spectrum used to obtain the reaction rate data used in a depletion calculation.

3.1 Reaction Rates

The ability to generate problem- and time-dependent reaction rates that accurately represent the fuel assembly design parameters and reflect changes in the assembly environment due to burnup and reactor operating conditions is extremely important to the accurate prediction of the spent fuel isotopic compositions. Some of the most significant advancements in improving the accuracy of point-depletion codes, widely used to predict spent fuel characteristics, have been in the application of improved reaction cross sections by using reactor physics codes to calculate the neutron energy spectrum in the fuel to properly weight the energy-dependent cross sections

used in the depletion calculations.[26] The main sources of potential uncertainty in the reaction rates as applied to depletion analysis calculations are discussed in this section. These include (1) the basic cross-section evaluations (measurements), (2) resonance cross-section shielding issues, and (3) fuel assembly modeling issues and flux solver methods used to generate the reaction rate information ultimately used in the depletion calculations.

3.1.1 Nuclear Data Evaluations

Currently, a large number of sources of evaluated nuclear cross-section data are available. These sources include the ENDF/B-V and –VI files, European JEF-2 files, Japanese JENDL-3 files, Chinese CENDL-2 files and the Russian BROND-2 data files. Even though these data files use common cross section evaluations for some nuclides, they also contain independent evaluations for others. In the United States, the ENDF/B data have typically been applied as the preferred source of basic cross-section data. However, although these evaluations represent some of the most up-to-date information available, they are not always complete. For example, the ENDF/B-V evaluations for ^{154}Eu and ^{155}Eu contained incomplete resonance cross-section data that resulted in significant errors in the predicted concentrations for ^{154}Eu, ^{155}Eu, and ^{155}Gd. This deficiency in the basic data led to an overprediction in the ^{154}Eu concentration using the deficient data and was partially responsible for a factor of 2 overprediction in the gamma dose rate for the MC-10 cask benchmark.[20] An earlier study of the effects of nuclear data uncertainty of fission products reactivity worth[25] found that there were significant differences in the calculated absorption fractions for the resonance absorbing nuclides using different evaluated nuclear data files JENDL-2, ENDF/B-V, and JEF-1, particularly for ^{133}Cs, ^{135}Cs, ^{99}Tc, ^{103}Ru, ^{108}Pd, and ^{155}Eu.

Comparisons of the predicted ^{244}Cm concentration using ENDF/B-IV- and –V-based data also suggest significant improvements using the newer cross-section evaluations (ENDF/B-V). It is therefore recommended that the cross-section evaluations important to the production of the dominant nuclides in high burnup fuel applications be reviewed against alternative data, wherever possible, as a means of identifying potential deficiencies in the nuclear data.

3.1.2 Resonance Cross-Section Issues

As the burnup of SNF increases, so do the concentrations of the longer-lived fission products and the majority of the actinides in the fuel. As the concentrations of highly absorbing nuclides increase, resonance capture and fission cross-section resonance shielding effects become larger. Resonance shielding effects include self-shielding (e.g., the influence of ^{238}U on itself) and mutual shielding effects (i.e., the effect of overlapping resonances between different nuclides). The ability to accurately correct resonance cross sections for shielding effects is important to the accuracy of fuel characterization analyses due to the high importance of cross sections to depletion analyses. Resonance cross-section issues are unique to multigroup transport methods.

A previous study investigating fission product resonance shielding effects for high conversion (designed for high plutonium production) LWRs[27] demonstrated the increased importance of self-shielding for high-burnup fuel on the predicted absorption rates and number densities. Even though this study focused on high-conversion fuel assembly designs, which have a much harder neutron spectrum and consequently higher resonance absorption rates than typically observed in conventional LWR fuel, the findings may have applicability to high-burnup studies since the increasing use of burnable absorbers and the high absorption rates that occur at high burnup also result in a hardening of the spectrum compared to conventional LWR burnup regimes. The same study also found that accounting for mutual shielding effects caused by resonance overlap between uranium and plutonium isotopes (e.g., ^{238}U) and several of the highly absorbing resonance fission products including ^{150}Sm, ^{151}Sm, ^{109}Ag, ^{95}Mo, and ^{133}Cs resulted in significant changes in the fractional absorption rates for these nuclides. Other computational benchmarks (calculations only) have shown an increase in the dispersion in physics code results with increasing plutonium content,[28] which is attributed to the quality of the cross-section data and insufficient resonance shielding calculations (neglecting mutual shielding effects). These latter effects resulted in significant

differences in the calculated atom densities for ^{242}Pu and its progeny, ^{243}Am and ^{244}Cm. The effect of resonance overlap in mixed absorbers was also studied[29] for PWR fuel with a relatively low burnup of 26 GWd/t. Although the study indicated that predicted plutonium and uranium concentrations did not appear to be highly sensitive to resonance interference effects at this burnup (predicted ^{240}Pu concentration was affected by about 1%), the potential effect at higher burnup could be larger.

Most of the codes currently used for depletion analysis treat resonance self-shielding of actinides and fission products but do not handle resonance overlap effects. For example, the NITAWL code is used to calculated resolved resonance cross sections and BONAMI is used to calculate self-shielded cross sections in the unresolved range for many libraries used for point-depletion analyses. These codes perform resonance self-shielding for both the actinides and fission products; however, mutual shielding effects are not taken into account. These effects are not currently perceived as a major issue to high-burnup fuel since most calculational methods now employed treat resonance self-shielding effects, and mutual (interference) shielding is believed to be of relatively minor importance even in the high-burnup LWR fuel regime. However, the perceived low importance of overlap effects needs to be demonstrated.

It is recommended that the importance of resonance cross-section shielding and resonance interference effects in the high-burnup regime be reviewed for both actinides and fission products. Any potential increase in the cross-section uncertainty attributed to possible self-shielding and mutual shielding effects should be investigated and quantified. This investigation could be accomplished using more rigorous calculational methods such as the CENTRM code[30] that can assess the effects of resonance overlap.

3.1.3 Modeling Issues

The increasing complexity of LWR assembly designs associated with higher initial enrichment fuel challenge the ability of physics codes to model the assemblies and accurately calculate the neutron flux and reaction rate information applied in the depletion analysis. The use of variable enrichment fuel rods within an assembly, burnable poison rods, and integral burnable poisons place increasing demands on the geometry capabilities of the physics codes used to model these assemblies. In addition, expanded operating regimes such as higher operating temperatures, higher specific powers, larger void fractions, and more complex and heterogeneous core loading patterns all combine to challenge computational methods.

The ORIGEN2 and ORIGEN-S codes are used extensively worldwide for spent fuel characterization due to their ability to characterize spent fuel by explicitly representing the concentrations of over 1600 individual nuclides. These codes obtain weighted cross sections for depletion analysis using different approaches. Extended-burnup PWR (50 GWd/t) and BWR (40 GWd/t) cross-section libraries, developed for the ORIGEN2 code, used detailed multidimensional physics models to simulate axial and radial depletion effects, axially varying moderator density, soluble boron, and burnable poisons within the assembly and used a complete fuel management cycle.[31] However, these libraries are based on predefined assembly design characteristics and operating conditions. Burnup-dependent cross sections are updated for only a limited number of actinides. As fuel assembly designs incorporate more advanced design features, these predefined assumptions may become less applicable and the cross sections generated with these models become less accurate. The ORIGEN-S code, as used within the SCALE code system, has the ability to utilize problem-dependent cross sections generated using a simplified one-dimensional neutronics model of the assembly to calculate the neutron flux spectrum (with a capability to approximate some two-dimensional effects). However, as assembly designs become more complex it can be an increasing challenge to accurately calculate the flux spectrum and thus prepare appropriate assembly cross sections using the simplified 1-D model in the SCALE system.[32]

The ability to accurately predict the reaction rates for a fuel assembly is dependent on the flux solver capabilities of the specific physics code used to model the assembly, with true multidimensional codes being more capable of explicitly modeling some of the more complex assembly design features than 1-D, or quasi 2-D codes. As the

fuel assembly designs and fuel management schemes become more complex, the ability of the physics methods to represent the neutronic environment (flux spectrum) of these assemblies in the reactor needs to be considered as a potentially large source of additional uncertainty for higher-enrichment and high-burnup fuels. The uncertainties in the neutron reaction rates associated with modeling complex fuel assembly designs with the present physics methods is perceived as a key issue needing resolution due to the well-established importance of reaction rates (or effective cross sections) to the accuracy of isotopic predictions. This issue is currently being investigated using 2-D calculational methods to estimate the magnitude of the uncertainties caused by the effects of modeling approximations involving various levels of assembly complexity, and ultimately estimate the importance on the predicted isotopic compositions. Some initial studies in this area have been reported for BWR fuel assemblies.[30, 33]

Another modeling-approximation issue is related to the use of point-depletion methods such as the ORIGEN codes to simulate the assembly-average fuel compositions. Such methods require that any heterogeneity, in the initial fuel compositions (e.g., variable-enrichment fuel rods), be homogenized and represented as average fuel parameters, and assumes all fuel in the assembly depletes at the same average rate. This method differs from more rigorous multidimensional depletion methods that allow an explicit representation of the different fuel regions (rods) of an assembly and allow depletion to proceed at the rate governed by the calculated local flux and power levels in the assembly. The accuracy of the point-depletion model for assembly calculations hinges on the assumption that the variation in isotopic concentrations with burnup is linear, or near-linear. The potential bias due to the point-depletion approximation depends to a large extent on the assembly heterogeneity and the burnup gradient across the assembly. The largest potential adverse effects are perceived to be for the higher actinides, which can exhibit very nonlinear production behavior with increasing burnup. Consider for example two fuel rods, one with a 30% higher burnup than the other. Given a nuclide that exhibits an exponentially increasing concentration with burnup, it is easy to demonstrate that the average concentration in the two rods will be greater than that predicted on the basis of the average burnup for the two rods. Although this effect is believed to be a minor issue for high-burnup LWR fuel calculations due to the relatively uniform fuel rod burnup in an assembly, it may be worthwhile considering the effect in code benchmarking exercises. Further studies are recommended to address the potential for larger calculational bias due to point-depletion representations of the complex and heterogeneous fuel assembly designs associated with higher-enrichment fuel.

A similar, and potentially larger, modeling effect has been observed for neutron source calculations that are based on the average axial assembly burnup.[34] The neutron source at the peak axial burnup position is underpredicted by scaling the average neutron source by the axial peaking factor since the neutron source term (dominated by ^{244}Cm) is nonlinear with burnup (varies approximately as the burnup to the fourth power).

3.2 Nuclear-Decay and Fission-Yield Data

Nuclear-decay data encompass the problem-independent data that describe the radioactive decay processes. These data include decay constants (half-lives), decay modes, branching fractions, and energy release per decay. Within this class of data are the fission product yields since these data are also largely problem-independent. In general, these data have a relatively small associated uncertainty and are not expected to be key parameters affecting the accuracy of depletion calculations in the high-burnup regime (relative to lower-burnup fuel). Decay constants are generally more important at the longer cooling times associated with long-term permanent spent fuel disposal where even small changes in the decay constants can lead to large changes in spent fuel isotopics.

A review of the nuclide half-lives[35] from ENDF/B-VI indicates that the measurement uncertainty is typically less than 1%. However, some nuclides have much larger uncertainties. As an example, a revision to the half-life of ^{79}Se in 1997 resulted in a change by more than a factor of 30 from previously used values. The new evaluation for ^{79}Se still has an uncertainty of about 18%. Decay data uncertainties need to be reviewed with respect to their effect on the predicted concentrations and activities of the dominant nuclides to the high-enrichment and high-burnup regimes.

3.3 Review of Isotopic Prediction Issues

The ability to obtain accurate reaction rates or cross sections for depletion analysis is perceived to be a key issue in accurately predicting isotopic compositions in high-burnup spent fuel. Although the accuracy of the basic cross-section evaluations (e.g., ENDF/B) is likely a significant factor related to the accuracy of predicted isotopics, it is not an issue unique to high-burnup applications since the cross-section libraries applied in high-burnup depletion analyses are typically the same as for conventional-burnup fuel. Therefore, the potential effect of cross-section evaluation uncertainty in the high-burnup regime lies in the changing sensitivities or importance to the data, where increasing sensitivity on a cross section with large uncertainty may adversely effect the accuracy of isotopic predictions relative to conventional-burnup fuel.

A potentially larger issue, and one directly related to high-burnup spent fuel studies, is the challenge of modeling increasingly complex assembly geometries with 1-D flux solvers typically applied for spent fuel characterization. The effects of averaging the assembly compositions and cross sections for uniform initial enrichments is perceived as a relatively minor issue to predicting isotopic compositions. Assembly averaging is only seen to be potentially important for assembly designs with variable-enrichment fuel pin splits or designs that employ burnable poison-rod arrangements that are not easily represented by 1-D physics code models. Resonance cross-section shielding issues are similarly perceived to be of relatively low importance as a high-burnup issue. However, these issues will be reviewed and their relative importance quantified in future studies.

4 ISSUES RELATED TO SPECIFIC APPLICATIONS

This section discusses the technical issues important to the specific application areas of decay-heat generation, neutron and gamma-ray source terms, and subcritical neutron multiplication (burnup-credit applications) in the high-burnup regime. These issues are addressed with respect to the cooling times generally characteristic of spent LWR fuel transportation and storage systems (typically 5 to 100 years).

The technical issues related to the accuracy of predicted isotopic compositions in high-burnup spent fuel are discussed in Sect. 3. More often the calculation of spent fuel isotopic inventories are an intermediate step towards calculating fuel characteristics and decay properties (e.g., decay-heat generation, radiation source terms, net criticality effects) which are based on a summation of the individual isotopes in spent fuel and their associated responses. The uncertainty in calculations involving aggregate nuclides includes the uncertainty in the predicted isotopic inventory and the uncertainty in the nuclide response (e.g., neutron and gamma-ray emission data, energy release per decay, etc.). As spent fuel tends towards higher enrichments and higher discharge burnups, the fuel compositions can change dramatically, resulting not only in changes in the magnitude of the response being calculated, but also in the nuclides that contribute most significantly to the spent fuel properties. The changes can adversely affect the accuracy of the predicted results by shifting the nuclides of importance away from those that may be well predicted to those, which may have high uncertainty.

To assist in identifying the dominant nuclides and changes in the characteristics of SNF with increasing burnup, nuclide importance rankings have been generated for both high-burnup PWR and BWR fuels for each of the application areas, including criticality safety, decay-heat generation, and neutron and gamma-ray source terms in Ref. 1.

4.1 Decay-Heat Generation

The relative contribution of the fission products plus activation products (e.g., ^{60}Co) and actinides to the total decay heat for 20-GWd/t (3.0-wt %) and 50-GWd/t (4.5-wt %) spent fuel, obtained from Ref. 36, are shown in Figures 4 and 5 for cooling times from 2 to 10,000 years. These figures illustrate the increased relative importance of actinide decay heating for high-burnup spent fuel and the increased total thermal decay power at higher burnup for all cooling times. The relative importance of the individual actinides and fission products in high-burnup spent fuel are compared in Figures 6 and 7. The decay heat rankings are given in Table 4 for the two burnups (20 GWd/t and 50 GWd/t) and their corresponding enrichments. The rankings show that the ^{134}Cs and ^{154}Eu fission products exhibit increasing relative importance with burnup since they are produced indirectly from ^{133}Cs and ^{153}Eu neutron capture, respectively, rather than as direct-yield fission products. All other fission products exhibit decreasing relative importance with burnup due primarily to the increased importance of the actinides relative to the fission products. A study to evaluate trends in nuclide importance performed as part of this review show that the most dramatic changes in the relative nuclide importance for high-burnup fuel (5-year cooling time) occur for ^{244}Cm and to a lesser extent, ^{238}Pu and ^{134}Cs. Figures 8 and 9 illustrate the trends in the relative contributions of the dominant radionuclides to the total decay heat with increasing burnup (from 30 to 60 GWd/t) for typical PWR fuel for 5- and 100-year cooling times.

Figure 8 shows that after a 5-year cooling time, the contribution from ^{244}Cm to the total decay heat increases from about 2% at 30 GWd/t to over 20% at 60 GWd/t. Therefore, any potential errors in the predicted ^{244}Cm inventory will have a much larger adverse effect on the predicted decay heat at high burnup than lower burnup, where the contribution from ^{244}Cm is much less. Figure 9 shows that after a 100-year cooling time the contribution from ^{238}Pu increases from about 10% of the total at 30 GWd/t to about 25% at 60 GWd/t. The contribution from ^{241}Am exhibits a corresponding decrease in importance over the same range.

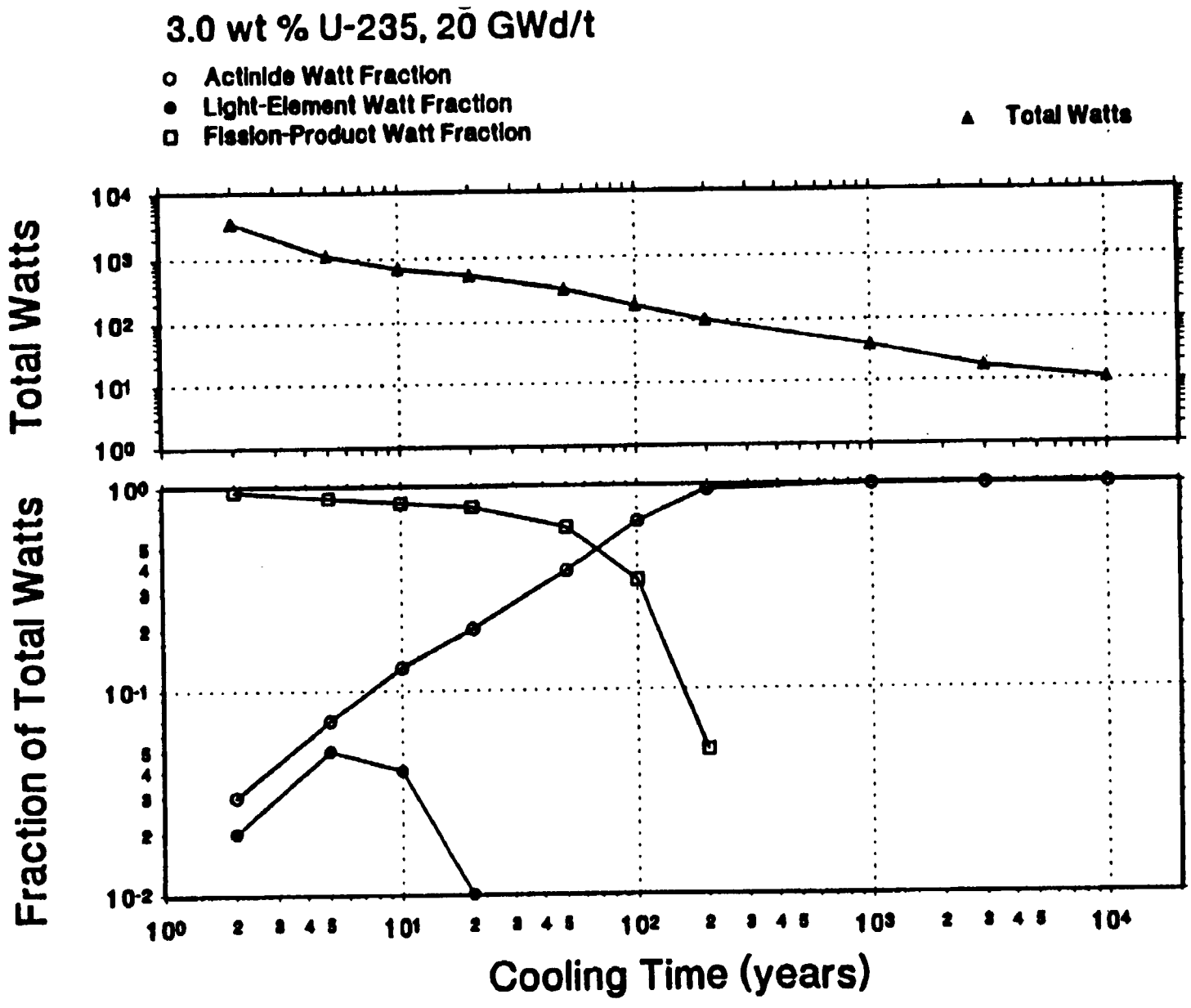

Figure 4 Actinide and fission product total decay heat fractions for 20-GWd/t spent fuel (from Ref. 36)

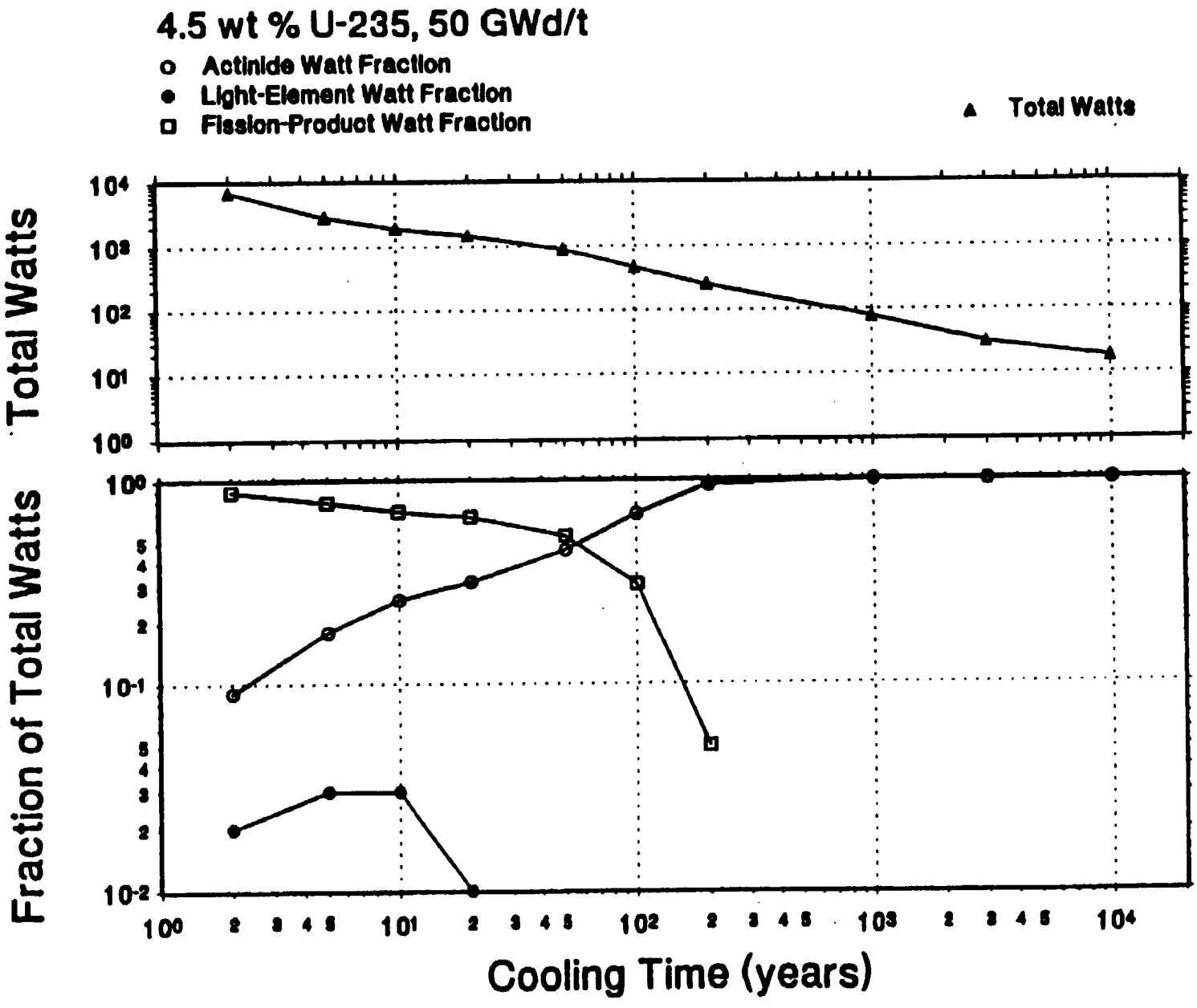

Figure 5 Actinide and fission product total decay heat fractions for 50-GWd/t spent fuel (from Ref. 36)

Table 4 Decay-heat rankings of actinides, fission products, and light elements with greater than 1% of total decay heat at 5 and 10,000 years (from Ref. 36)

	5 years		10,000 years	
Nuclide	20 GWd/t 3.0 wt %	50 GWd/t 4.5 wt %	20 GWd/t 3.0 wt %	50 GWd/t 4.5 wt %
Actinides				
Pu-238	1 (2)[a]	2 (7)	-	-
Am-241	2 (2)	3 (2)	-	-
Pu-240	-	-	2 (33)	2 (41)
Pu-239	-	-	1 (65)	1 (54)
Cm-244	-	1 (7)	-	-
Am-243	-	-	-	3 (3)
Fission products and light elements				
Y-90	1 (23)	1 (19)	-	-
Ba-137m	2 (20)	2 (18)	-	-
Cs-134	3 (11)	3 (17)	-	-
Rh-106	4 (10)	4 (7)	-	-
Pr-144	5 (9)	7 (4)	-	-
Cs-137	6 (6)	5 (5)	-	-
Sr-90	7 (5)	6 (4)	-	-
Co-60	8 (5)	8 (3)	-	-
Eu-154	9 (1)	9 (2)	-	-

[a] Percentage contribution to the total decay heat levels.

In addition to considering the changing relative importances of the dominant isotopes, the accuracy of decay-heat predictions in high-burnup fuel must also account for the potential changes in the accuracy of the predicted inventories of the dominant isotopes in the high-burnup range compared with the conventional-burnup range. For a nuclide that exhibits increased importance in a regime where the ability to accurately predict that nuclide is decreasing, the net adverse effect on the accuracy of the predicted quantity will be compounded. The uncertainties in the basic nuclear-decay data (decay constants and recoverable energy per decay) should also be considered in any estimation of the biases and uncertainties in regimes with little or no experimental data. These uncertainties are available in the evaluated nuclear data files.

The limited amount of measured decay-heat data for high-burnup fuel readily available in the United States has restricted the range of the NRC Regulatory Guide 3.54 for spent fuel heat generation to a maximum of 50 GWd/t for PWR fuel and 45 GWd/t for BWR fuel. The potential for larger uncertainties in the predicted inventories in high-burnup SNF due to changing sensitivities to nuclear data and/or increased uncertainties in the nuclear cross sections caused by modeling approximations (see Sect. 3) need to be considered. Any extrapolation beyond the current limits of the regulatory guide (in the absence of a sufficient quantity of measured data) must consider the changing relative nuclide importance at higher burnup, particularly the dramatic increase in the importance of ^{244}Cm and other actinides.

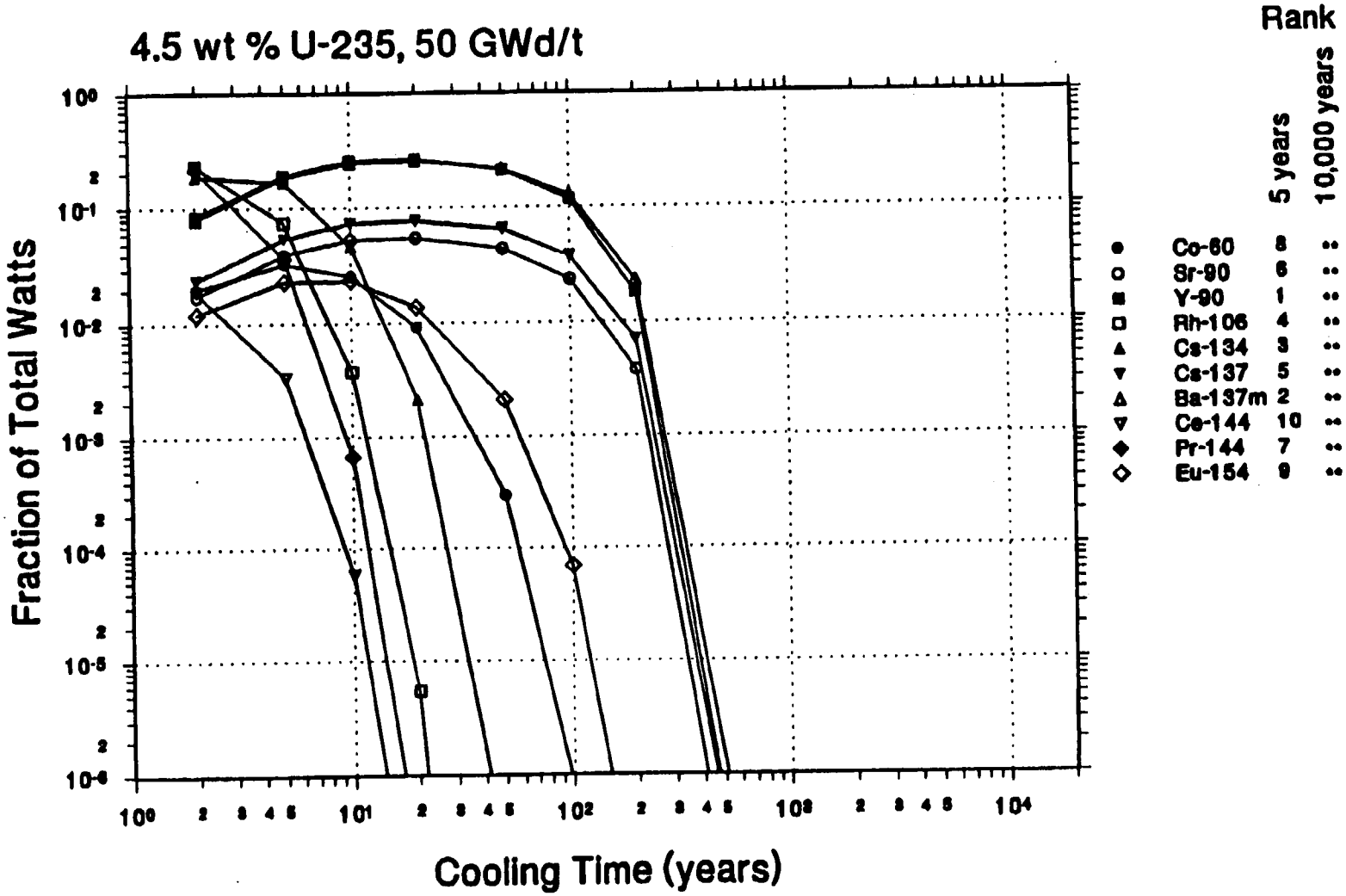

Figure 6 Fission product and activation product decay-heat generation for 50-GWd/t spent fuel (from Ref. 36)

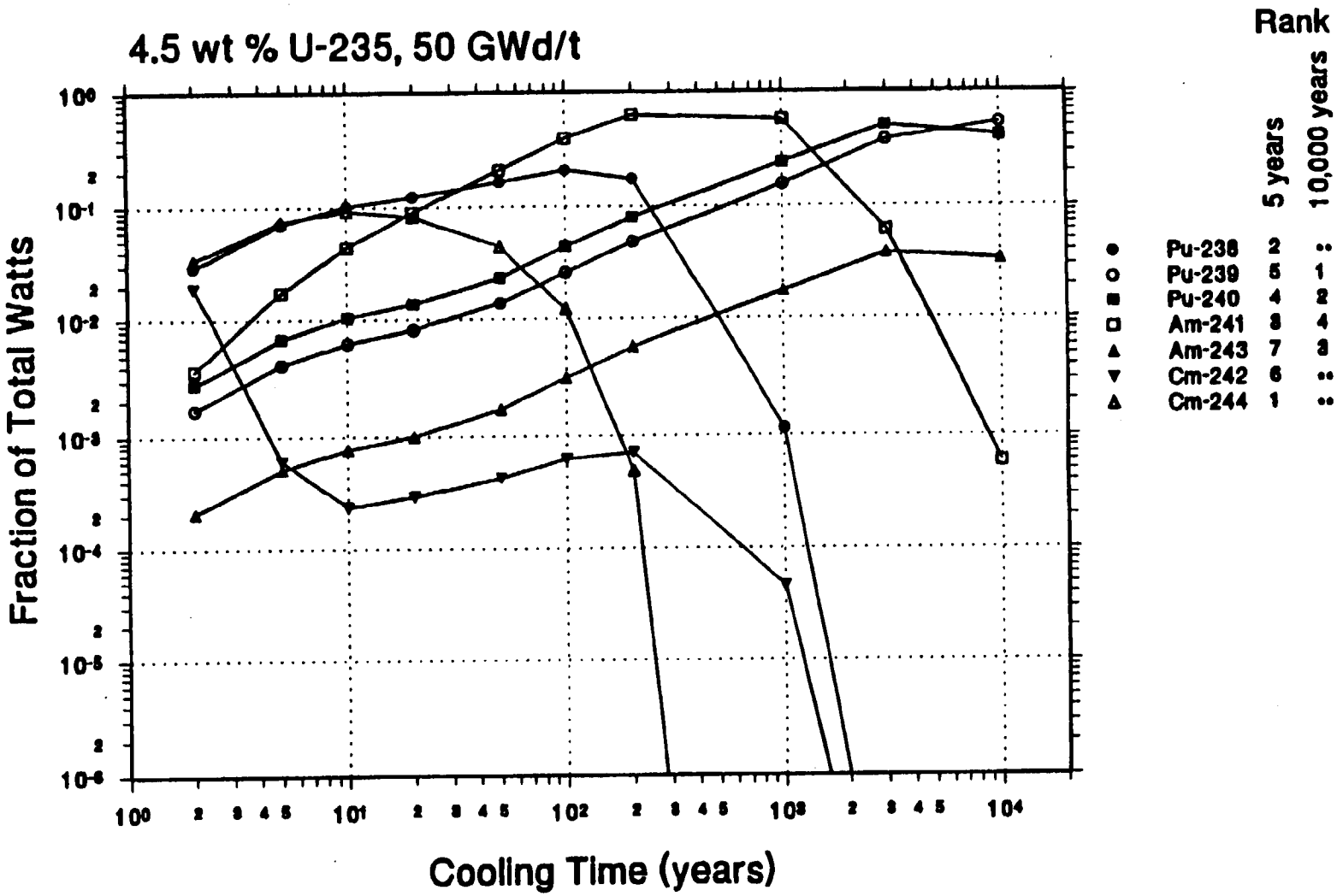

Figure 7 Actinide decay-heat generation for 50-GWd/t spent fuel (from Ref. 36)

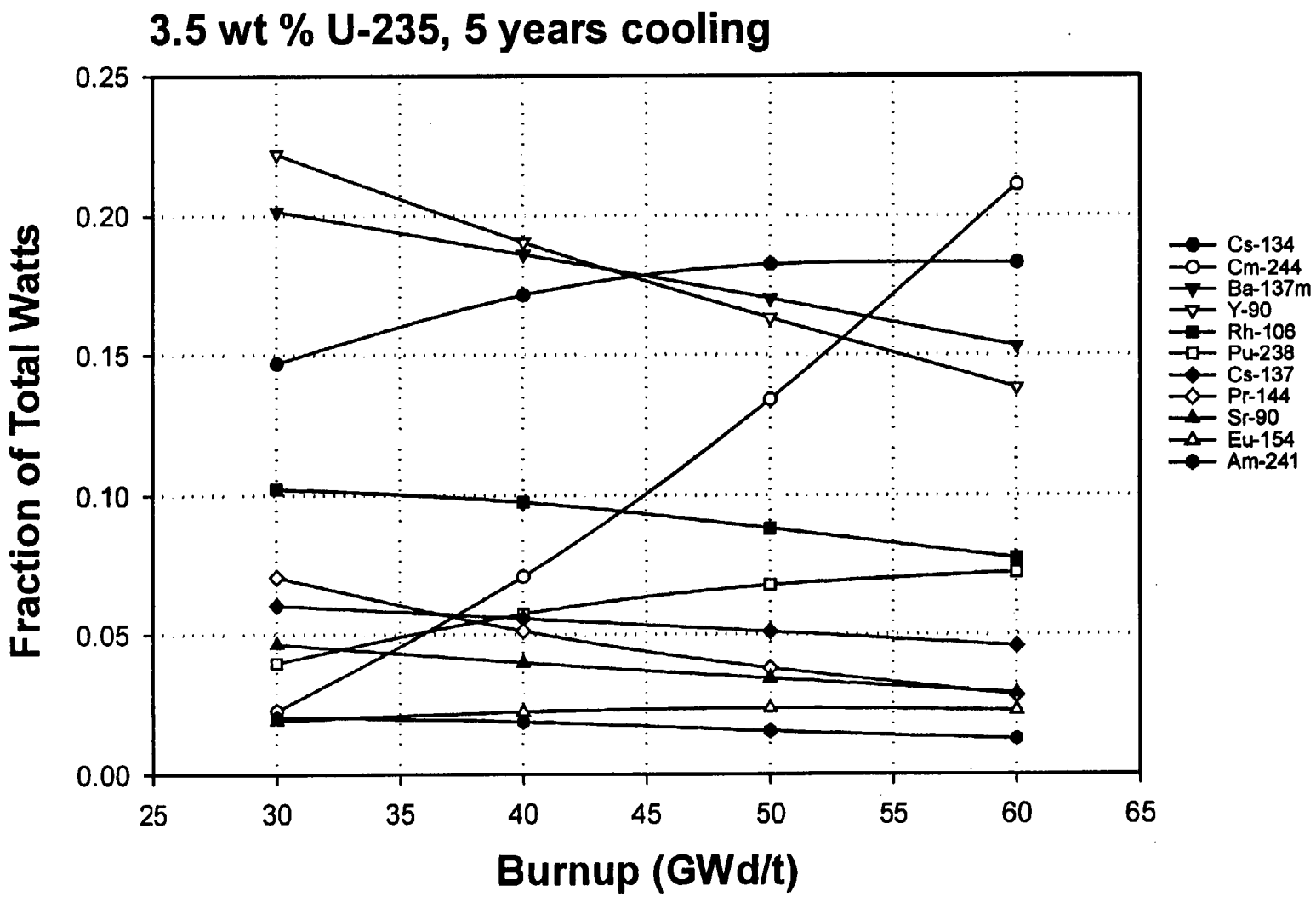

Figure 8 Total decay-heat fractions after a 5-year cooling time as a function of burnup

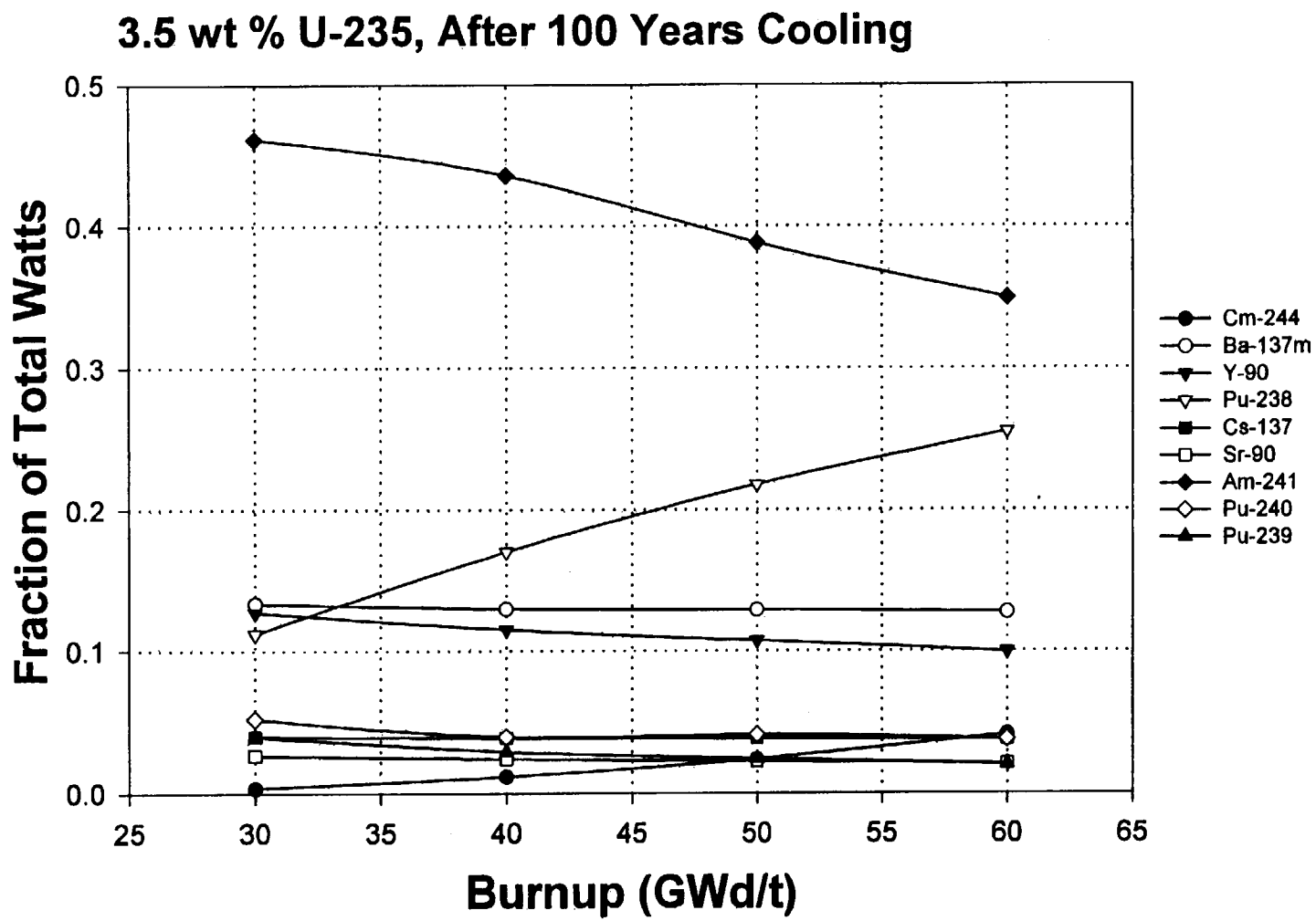

Figure 9 Total decay-heat fractions after a 100-year cooling time as a function of burnup

4.2 Radiation Source Terms

The accuracy of predicted neutron and gamma-ray source terms in high-burnup spent fuel is dependent on both the ability to predict the concentrations of the dominant actinides and fission products, and the accuracy of the radiation source characteristics (photon emission yields and energy spectra) for the dominant nuclides in the particular application. The gamma rays (photons) generally represent the primary component of the total dose rate outside of shielded spent fuel transportation and storage facilities for conventional-burnup fuel. Gamma rays are emitted directly by the radionuclides in spent fuel during decay (primary gamma rays), and are also emitted by the interaction of neutrons in the fuel and the surrounding shielding structures (secondary gamma rays). Figure 10 illustrates the relative contribution of the primary gamma rays, neutrons, and secondary gamma rays as a function of cooling time for 20-GWd/t (3.0-wt %) spent fuel for the TN-24 (iron cask with outer neutron resin shielding) dry storage cask. Figure 11 illustrates the results for 50-GWd/t (4.5-wt %) spent fuel. The dominant fission products and actinides that contribute to the primary gamma dose rate for three different cask shield materials are ranked in Table 5. The ranking is based on the relative importance of the nuclides to the total dose rate for 20-GWd/t and 50-GWd/t spent fuel at a 5-year cooling time.

Note that these results include a significant contribution from ^{60}Co, which is produced from the activation of structural material (Inconel grid spacers and end-fittings) associated with the fuel assembly. The contribution from ^{60}Co is highly dependent on the initial cobalt impurity levels. The ranking of ^{60}Co in Table 5 assumed a relatively high impurity level typical of the older fuel assemblies. Newer assemblies are expected to have much lower impurity levels and will therefore exhibit significantly lower dose-rate contributions from activation products. The calculation of activation product sources (structural activation) requires a modified calculational approach than for spent fuel (fission products and actinides) since the activated structures typically reside in a significantly different neutronic environment and flux level than that experienced by the fuel. It is recommended that the burnup dependence of structural material activation be studied using more realistic impurity levels that reflect the newer assembly designs.

Figs. 10 and 11 illustrate the dramatic increase in the importance of the neutron source (and consequently also the secondary gamma rays) to the total dose rate for high-burnup fuel at cooling times of interest to transport and cask storage analyses. For a burnup of 20 GWd/t the neutron and secondary gamma-ray dose rates are of low importance for cooling times up to about 200 years. However, at 50 GWd/t the neutron dose rate becomes important after only 10-years cooling, and actually becomes the dominant component of the dose rate after 30 years. This dominance is attributed to the rapid buildup of the plutonium, americium, and curium actinides (neutron sources) at high burnup. The contribution of ^{244}Cm to the total dose rate for the various cask designs exhibits about a factor of 10 increase from 20 to 50 GWd/t (see Table 5). The dominant fission product nuclides, ^{134}Cs and ^{154}Eu, also show increasing importance with burnup.

4.2.1 Photon Data Libraries

The gamma-ray source spectra used in spent fuel shielding analyses are generated using the calculated isotopic inventories in the fuel and gamma-ray emission data (photon yields and energies). These gamma-ray data may be in the form of line data containing the individual gamma energies and intensities, or as multigroup data that represent the average intensities in an energy-group interval. Inaccuracies in the photon data, or incomplete library data, can have a direct and significant impact on the accuracy of the radiation source terms.

As spent fuel tends towards increasingly higher burnup, the relative importance of the dominant radionuclides in shielding applications can change significantly (see Table 5). Consequently, the accuracy of the photon data for nuclides exhibiting increasing importance in the high-burnup regime needs to be reviewed to ensure that potential

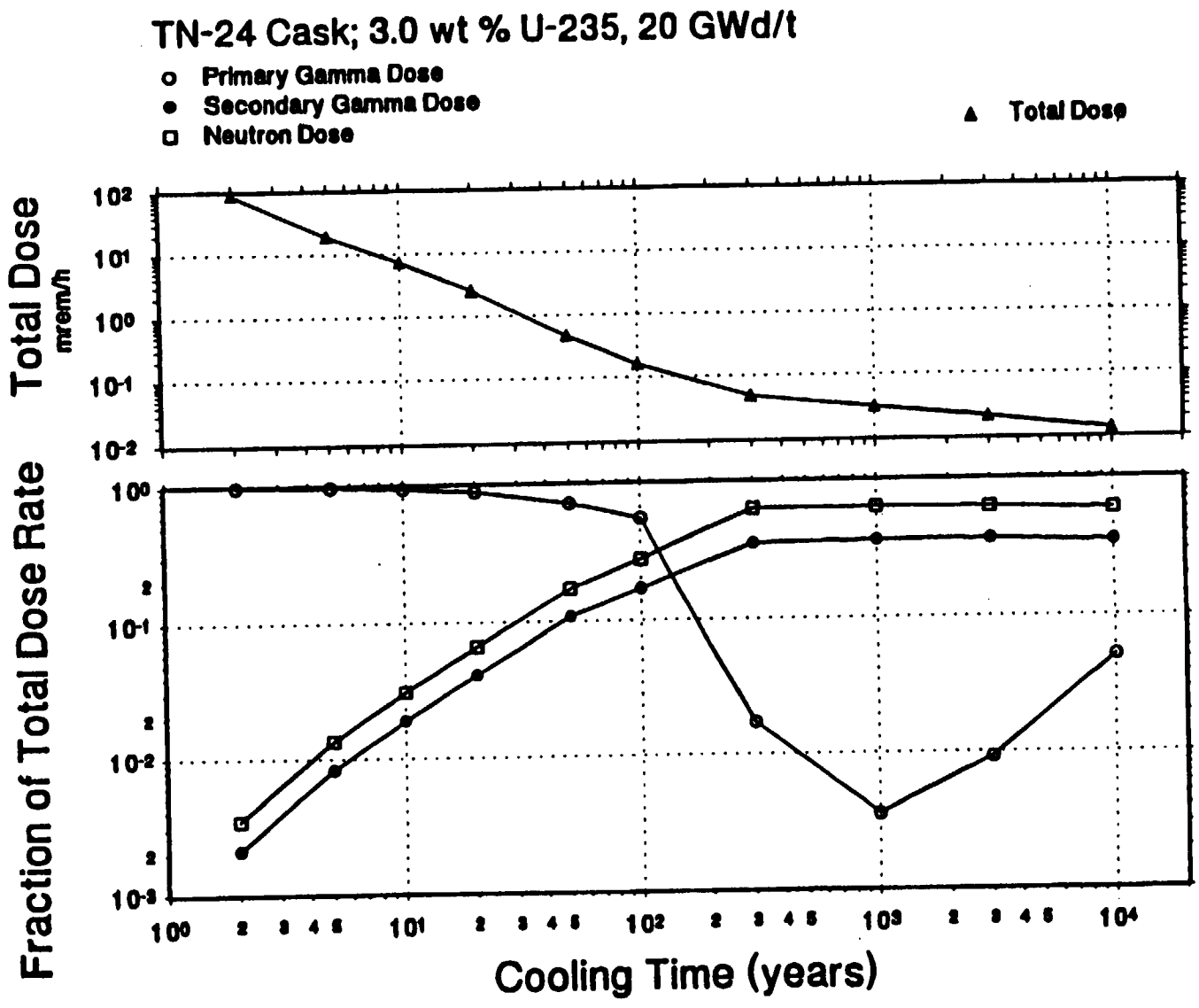

Figure 10 Neutron and gamma-ray dose rates and dose fractions for 20-GWd/t spent fuel in TN-24 dry transport cask (from Ref. 36)

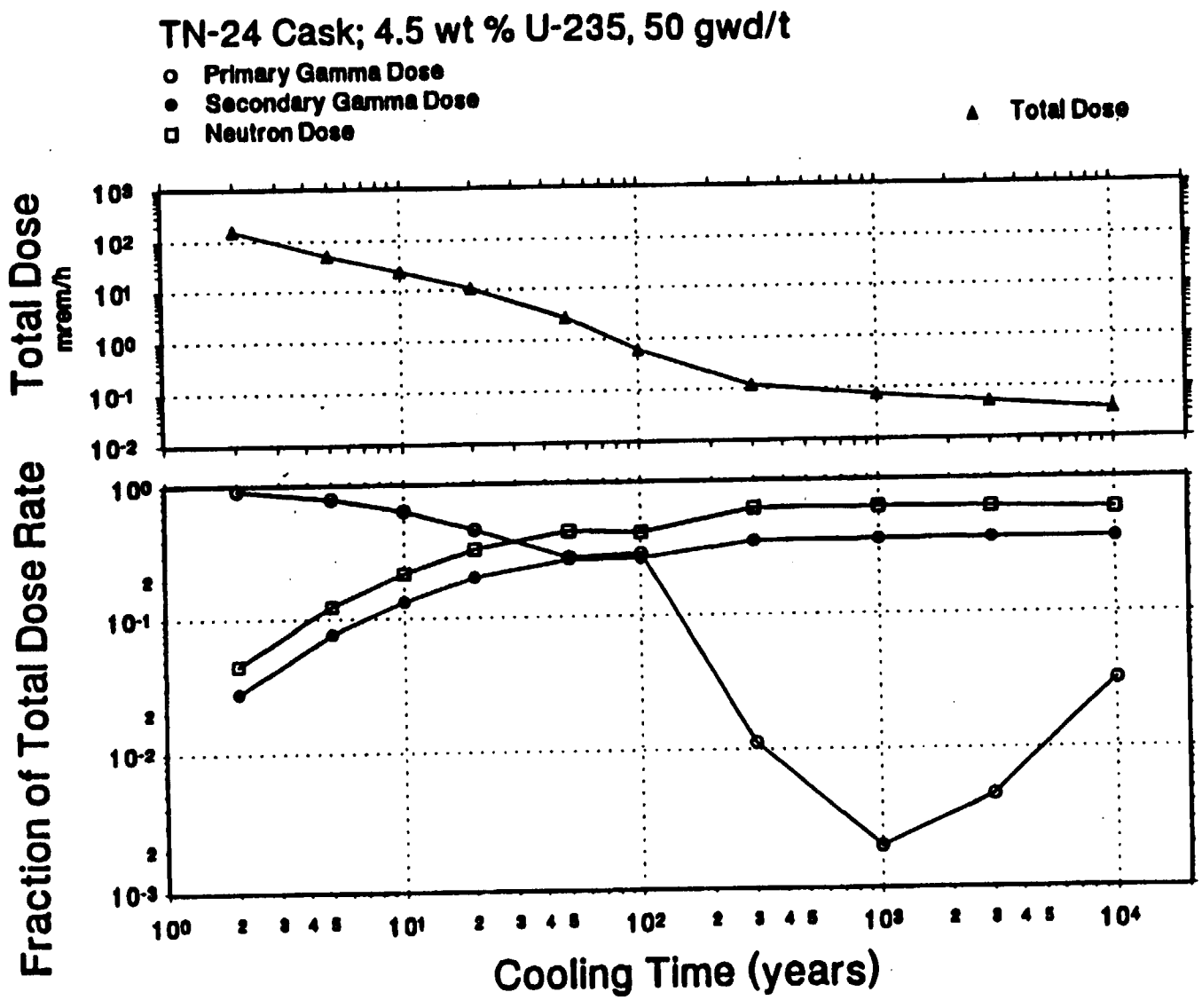

Figure 11 Neutron and gamma-ray dose rates and dose fractions for 50-GWd/t spent fuel in TN-24 dry transport cask (from Ref. 36)

Table 5 Shielding rankings nuclides greater than 1% of total dose at a 5-year cooling time (from Ref. 36)

	Iron cask[a]		Lead cask[a]		Concrete cask[a]	
Nuclide	20 GWd/t 3.0-wt %	50 GWd/t 4.5-wt %	20 GWd/t 3.0-wt %	50 GWd/t 4.5-wt %	20 GWd/t 3.0-wt %	50 GWd/t 4.5-wt %
Co-60	1 (49)[b]	1 (33)	1 (56)	1 (40)	1 (50)	1 (39)
Pr-144	2 (19)	3 (8)	2 (17)	3 (8)	3 (12)	5 (6)
Cs-134	3 (11)	2 (15)	3 (10)	2 (16)	2 (14)	2 (23)
Rh-106	4 (9)	5 (6)	4 (8)	5 (6)	5 (7)	6 (6)
Eu-154	5 (4)	4 (7)	5 (4)	4 (8)	6 (5)	3 (10)
Ba-137m	6 (3)	6 (3)	-	-	4 (9)	4 (9)
Y-90	7 (1)	-	6 (1)	-	7 (1)	7 (1)
Cm-244	1/-/-[c](1/-/-)[d]	1/1/-(12/7/-)	1/-/-(2/-/-)	1/1/-(19/1/-)	-/-/-(-/-/-)	1/1/-(2/2/-)

[a]Gamma shields consist of 27-cm steel, 12.7-cm lead, and 50-cm concrete for the iron, lead, and concrete casks.
[b]Percentage contribution to the total dose.
[c]Rankings with respect to neutron/primary gamma/secondary gamma dose rates. The /-/ symbol indicates all contributions less than 1%.
[d]Percentage contribution from each isotope to the total dose, listing neutron/primary gamma/secondary gamma separately.

errors in the photon evaluations (that may not be evident in the lower-burnup regime) do not become a large source of error at high burnup.

One of the most extensive photon libraries[37] available in the United States was developed for the ORIGEN code in 1979 using ENSDF (Evaluated Nuclear Structure Data File) nuclear data.* The photon data evaluations in this library are therefore well over 20 years old. The photon libraries currently in use need to be evaluated to address the adequacy of the database for the nuclides important in the high-burnup regime and identify potential improvements that can be achieved using more up-to-date evaluated photon data currently available in ENSDF. Specifically, the photon library data for the dominant fission product nuclides that exhibit increasing relative importance in high-burnup fuel (e.g., ^{134}Cs and ^{154}Eu) should be reviewed against newer photon yield data. Improvements in the predicted gamma-ray source over the entire LWR fuel enrichment and burnup regime may also be achieved by updating other high-importance radionuclides.

4.2.2 Neutron-Source Data

Neutron sources in spent fuel are generated primarily from spontaneous fission neutrons and from (α,n) reactions in the fuel. The (α,n) component results when alpha particles (α), emitted by actinide decay, interact with light elements in the matrix (lithium, boron, oxygen, etc.) and is therefore highly dependent on the compositions of the medium containing the α-emitters. In addition to these primary sources of neutrons in spent fuel, neutrons are generated when primary neutrons cause secondary fissions in the spent fuel (subcritical multiplication) which in turn release subsequent fission neutrons (~2.4 neutrons/fission). Therefore as the neutron multiplication factor-k_{eff}, for a storage cask increases, so does the contribution from the secondary fission neutrons (source multiplication effect).

* ENSDF is produced by the International Nuclear Structure and Decay Data Network and is maintained at the National Nuclear Data Center, Brookhaven National Laboratory.

The neutron source (neutron/s) emitted from typical spent PWR fuel with discharge burnups of 27 and 60 GWd/t are illustrated in Figs. 12 and 13 for cooling times up to 10,000 years. The figures show the increased relative importance of spontaneous fission neutrons relative to the (α,n) source at the higher burnup. For 60-GWd/t fuel the spontaneous fission neutron source completely dominates the source over the cooling time of interest to spent fuel transport and storage. Previous work[9] suggests that the total neutron source at discharge increases nearly exponentially with burnup. The impact of the increased relative importance of the neutron source to the total dose rate (after 5-year cooling) outside of several storage cask designs (attributed mainly to ^{244}Cm) is evident in Table 5.

The nuclear data required to calculate the neutron production rate from spontaneous fission include the spontaneous fission half-life, the average neutron yield per fission, and the concentrations for each of the contributing actinides. The spectrum of the spontaneous fission source is provided by ORIGEN-S using the spontaneous fission spectra for ^{242}Cm and ^{244}Cm and the calculated concentrations for these two actinides. This spectrum is then renormalized to the total neutron source from all spontaneous fission nuclides. A similar procedure is used to calculate the (α,n) source and spectra using the concentrations and (α,n) neutron spectra for ^{238}Pu, ^{242}Cm and ^{244}Cm. The ORIGEN2 code calculates the neutron source but does not provide the associated energy spectrum.

It is recommended that the calculational procedures, nuclear data, and assumptions presently used to calculate the neutron source and energy spectra be reviewed to assess the adequacy of the current methods and data for the actinide compositions associated with high-burnup fuel. Any potential improvements in these calculations that can be achieved using more up-to-date data should also be investigated given the significantly greater contribution of neutron sources to the total dose rate for high-burnup fuel. Particular emphasis should be placed on evaluating the spontaneous fission sources due to their increased relative importance to the total neutron source at high burnup.

4.3 Criticality Safety

The recent issuance of Interim Staff Guidance 8 (ISG8) by the NRC (Ref. 7) allows criticality safety analyses for commercial PWR spent fuel storage and transport casks to take partial credit for the decrease in reactivity of spent fuel as a result of irradiation. ISG8 currently recommends that the application of burnup credit be restricted to actinides only (no fission products), limits burnup credit to PWR fuel having an enrichment below 4.0 wt % without a loading offset penalty and up to 5.0 wt % with an offset penalty, and only considers assemblies that have not used burnable absorbers. The guidance also limits the maximum amount of burnup credit to that available in PWR fuel with an assembly-average burnup of 40 GWd/t or less.

A review and summary of the phenomena and technical issues relevant to burnup credit in storage and transportation casks is provided in Ref. 38. Based on previous burnup-credit studies, the actinides and fission products listed in Table 6 are considered to be most important to dry storage and transport cask criticality safety analyses. The relative worth of these nuclides will vary to some degree, depending on the enrichment, burnup, assembly design, and cooling time, but the important nuclides will remain the same.

Table 6 Most important nuclides in criticality calculations

^{234}U	^{235}U	^{236}U	^{238}U	^{238}Pu	^{239}Pu
^{240}Pu	^{241}Pu	^{242}Pu	^{241}Am	^{243}Am	^{237}Np
^{95}Mo	^{99}Tc	^{101}Ru	^{103}Rh	^{109}Ag	^{133}Cs
^{143}Nd	^{145}Nd	^{147}Sm	^{149}Sm	^{150}Sm	^{151}Sm
^{151}Eu	^{152}Sm	^{153}Eu	^{155}Gd		

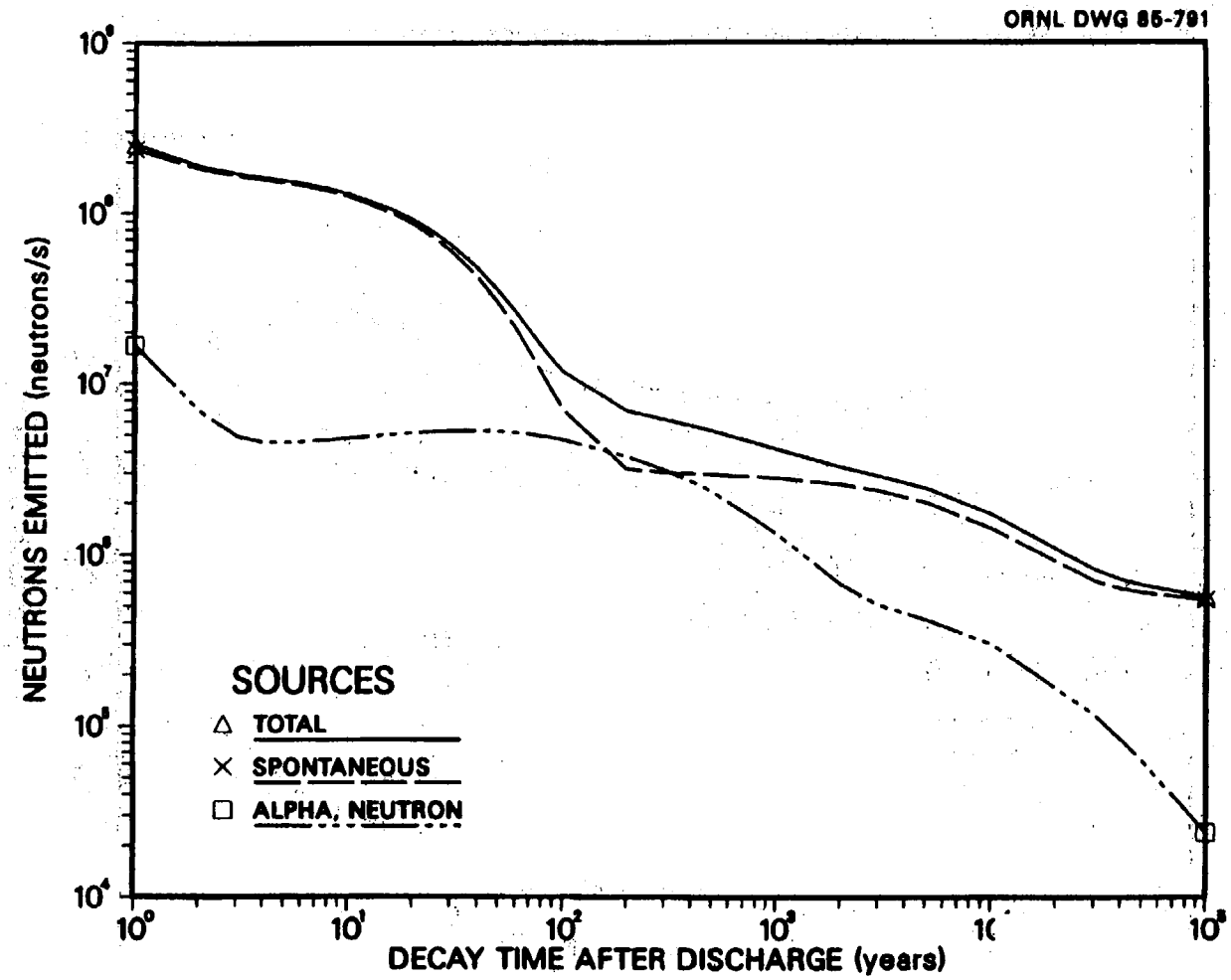

Figure 12 Neutron sources from 1 metric ton (t) of PWR spent fuel; 27.5 GWd/t (from Ref. 9)

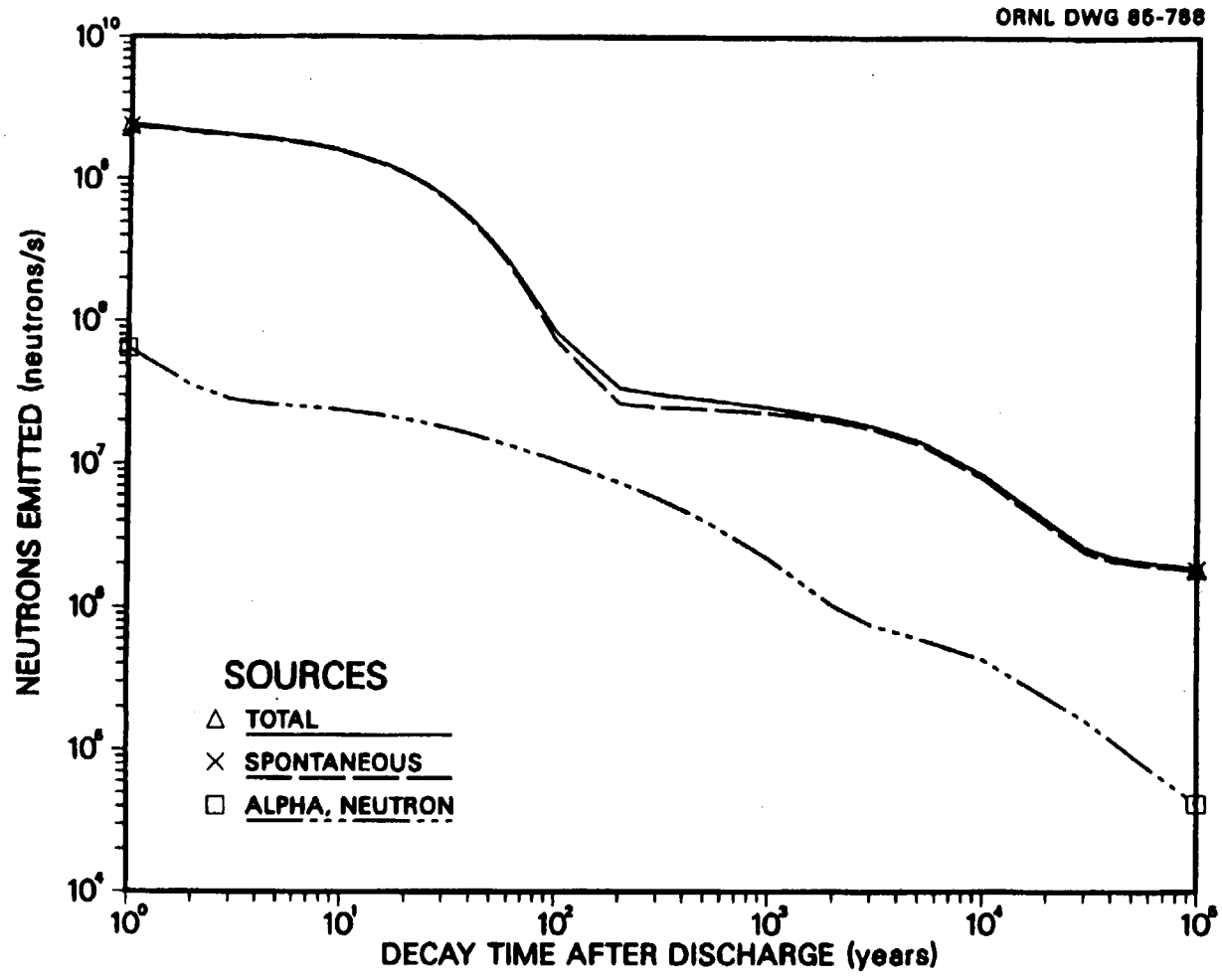

Figure 13 Neutron sources from 1 metric ton (t) of PWR spent fuel; 60 GWd/t (from Ref. 9)

The approach to burnup credit for storage and transport casks proposed in the United States has been to qualify the accuracy of the calculated isotopic predictions for the important burnup-credit nuclides using radiochemical assay data, and to qualify separately the accuracy (bias) in the calculation of the system k_{eff} by validating against critical experiments. An important consideration in the application of burnup credit to criticality safety is establishment of appropriate biases and uncertainties for the predicted isotopic compositions.

The burnup limitation of 40 GWd/t in ISG8 is based largely on the absence of experimental isotopic assay data to support code validation beyond this burnup (see Sect. 2). The isotopic validation data currently available for PWR fuel in the United States are limited for the most part to the actinides in Table 6. Even though several experiments included a fairly extensive number of fission product measurements, only the Calvert Cliffs assay data included measurements for many of the important burnup-credit fission products in Table 6. For BWR fuel, the number of burnup-credit fission products with measured data is severely restricted, and the isotopic measurements are effectively limited to just the actinides in Table 6. The limited quantity of fission product data is a major reason that ISG8 allows burnup credit for only the actinides.

The potential acquisition of high-burnup PWR and BWR fuel assay measurements in the near term, discussed in Sect. 2, would add valuable experimental data for validating depletion analysis codes used for predicting the concentrations of the burnup-credit actinides. However, even with the addition of potentially new data there will still be a relatively small amount of data from which to establish code bias and uncertainty in the high-enrichment and high-burnup regime.

The relative importance of actinide and fission product nuclides to the negative worth (absorptions) of PWR fuel with a burnup of 50 GWd/t, taken from Ref. 34, is illustrated in Figs. 14–16 as a function of cooling times from 5 to 10,000 years. The figures indicate that the majority of the neutron absorption is attributed to only a few actinides, and individually, fission products contribute much less to neutron absorption. In terms of the aggregate importance, fission products contribute about 15 to 20% of the total absorptions.

4.4 Review of Application Issues

Many of the technical issues related to the prediction of isotopic compositions in high-burnup fuel described in Sect. 3 are directly relevant to the application area of decay heat generation, radiation source terms, and criticality safety. In spent fuel criticality safety applications (burnup credit) the accuracy of isotopic predictions and the assigned calculational biases are key factors in accurately predicting the reduction in reactivity in spent fuel. The sensitivity-based methods described in Appendix A are being explored as a means of better understanding and quantifying the trends in biases in the predicted isotopic concentrations of the nuclides important to each of the application areas over the entire enrichment and burnup regime. This information, combined with the limited amount of assay data that will likely become available in the near future from national and international programs (see Sect. 2), may make it possible to establish a technical basis for extending the enrichment and burnup restrictions currently recommended in NRC guidance documents.

Sensitivity analyses are also being pursued as a method to establish similarity of different spent fuel systems by evaluating the dependence on the underlying nuclear data that influence spent fuel compositions (e.g., cross section, decay data, etc.). Regimes having a high degree of similarity suggest that validation data obtained in one spent fuel regime may be highly applicable to other regimes since the basic processes and nuclear data parameters that govern the spent fuel compositions as they affect the response of the fuel are largely the same. These methods are also being used to study the significant differences between the burnup and enrichment regimes to identify which parameters are important to predicting the key nuclides are not sufficiently measured by the available data, and to identify where additional isotopic validation data may be needed.

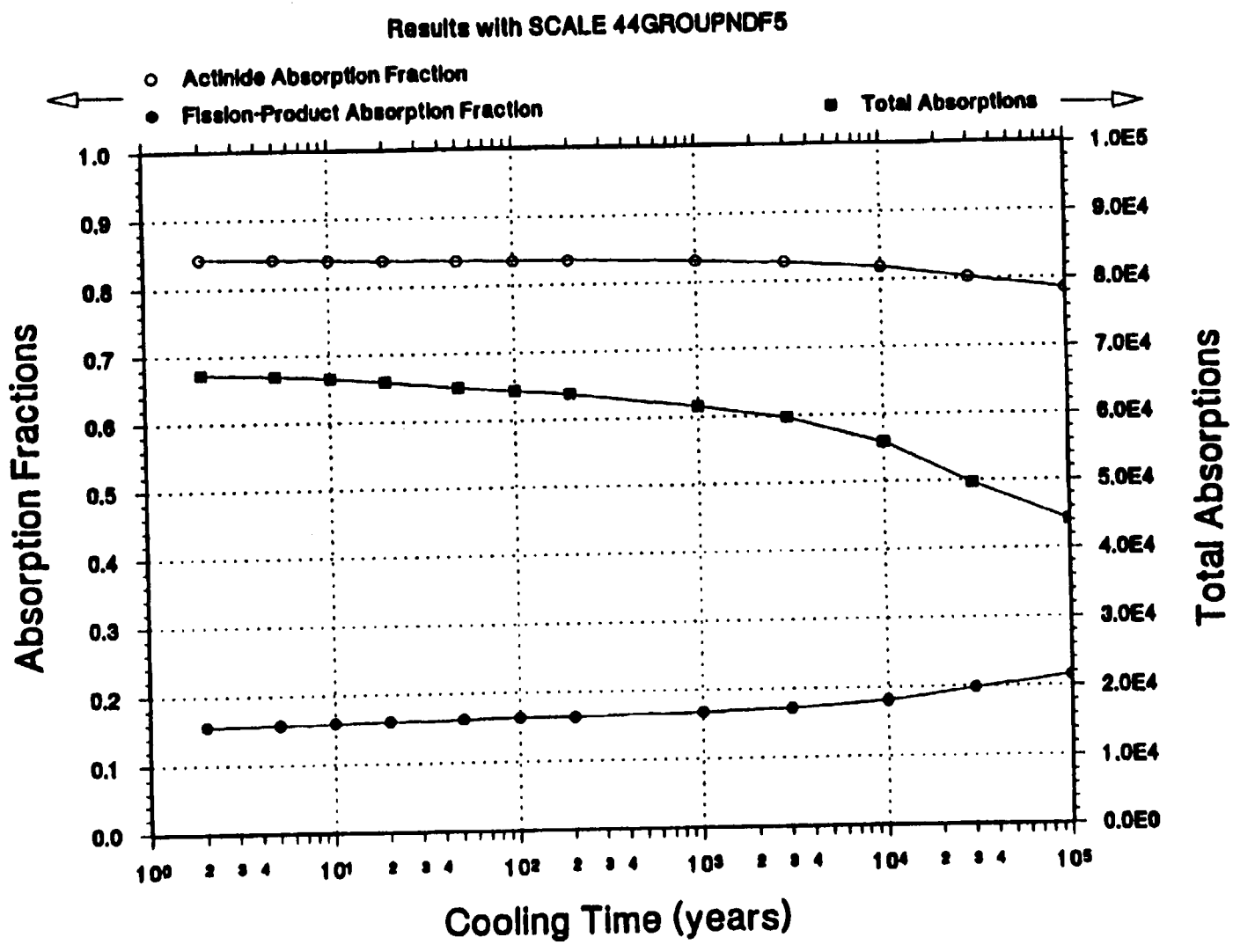

Figure 14 Fraction of neutron absorptions versus cooling time for 4.5-wt %-enriched PWR fuel having a burnup of 50 GWd/t (from Ref. 34)

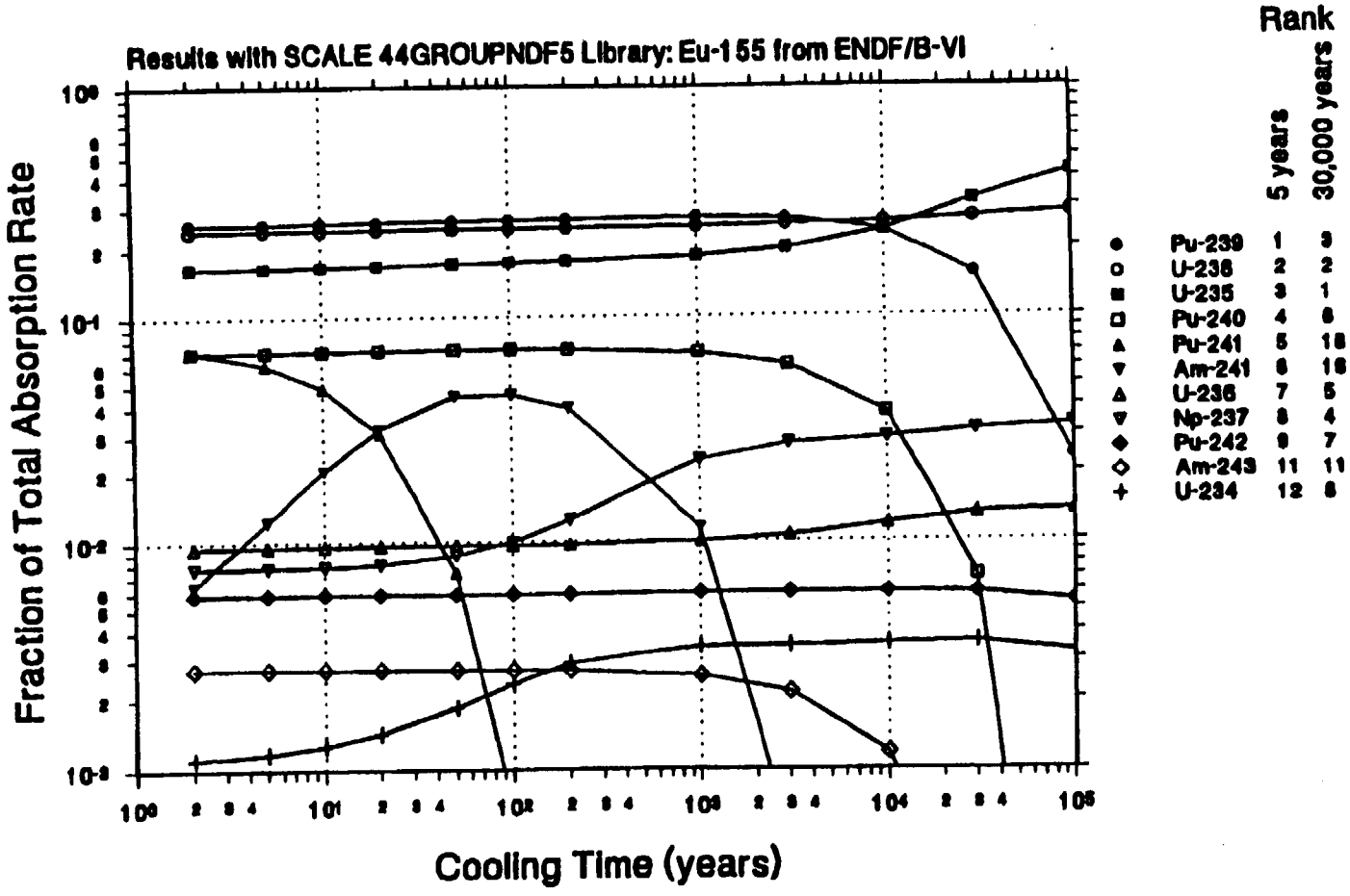

Figure 15 Fraction of neutron absorptions by major actinides at various cooling times for 4.5-wt %-enriched PWR fuel having a burnup of 50 GWd/t (from Ref. 34)

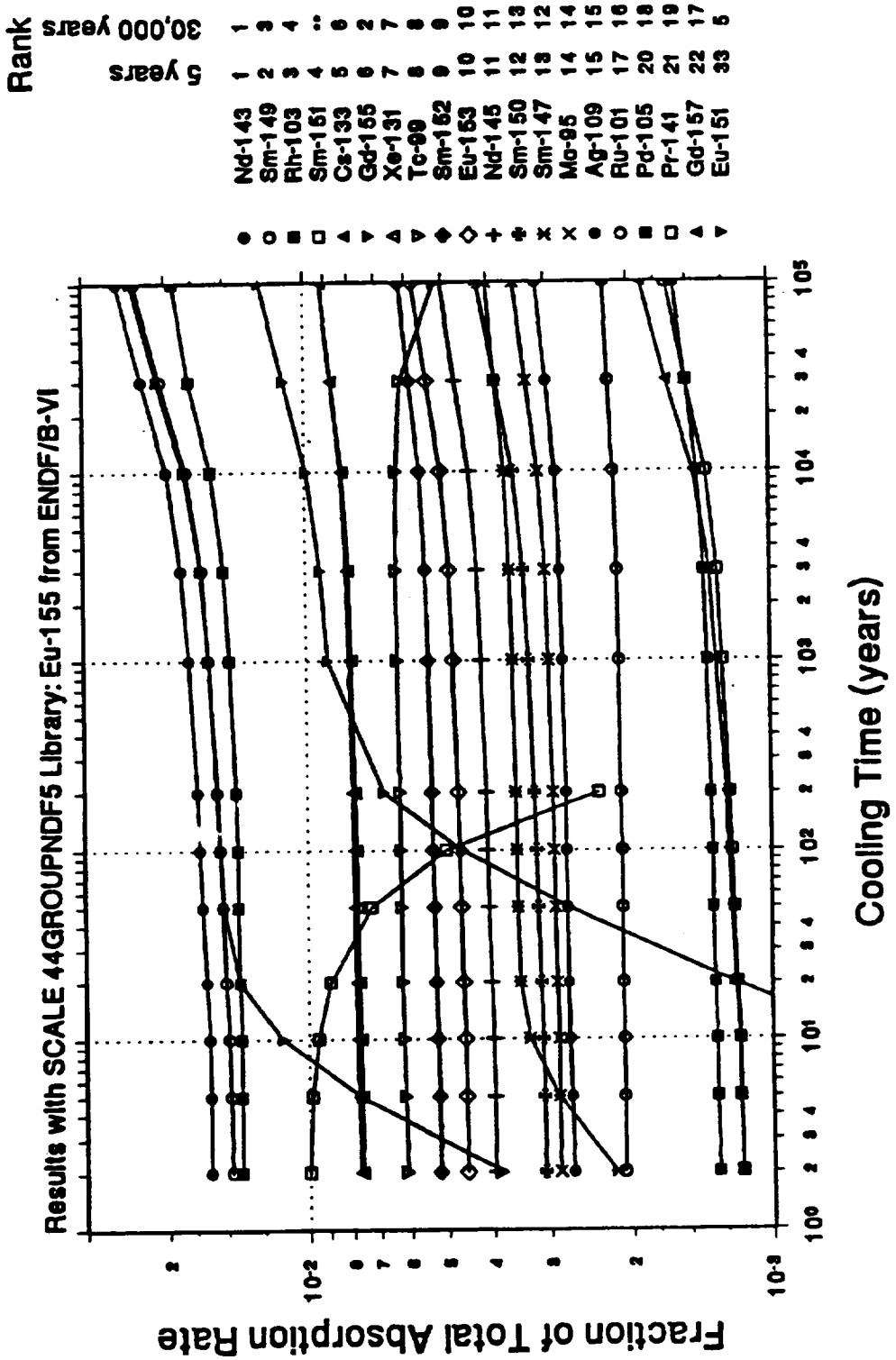

Figure 16 Fraction of neutron absorptions by major fission products at various cooling times for 4.5-wt %-enriched PWR fuel having a burnup of 50 GWd/t (from Ref. 34)

In parallel with sensitivity studies, importance rankings of the key radionuclides in each application area have been established in high-burnup spent fuel.[1] These rankings are designed to facilitate validation efforts related to the prediction of spent fuel isotopics and radiological decay properties of high-burnup spent fuel by identifying the nuclides that have the greatest impact in the application areas of interest and illustrate the variation in the nuclide importance with burnup. A review of the finding from this study indicates that relatively few radionuclides not already being measured in new burnup-credit chemical assay programs are needed to provide a relatively comprehensive characterization of high burnup spent fuel for decay heat and source-term application areas. Also, several key radionuclides exist as parent-daughter pairs (e.g., ^{137}Cs–^{137m}Ba, ^{90}Sr–^{90}Ym ^{144}Ce–^{144}Pr). The study suggests that the measurement of fewer than ten additional radionuclides would be sufficient to represent the isotopes that are responsible for most of the response in all applications considered here. These data could provide a means of validating the codes in the respective areas without having to initiate new experimental programs to measure decay heat and radiation spectra from high-burnup SNF.

5 PROPOSED RESEARCH AREAS

This report has reviewed some of the perceived technical issues related to the accuracy of current calculational methods and nuclear data for predicting spent fuel isotopic inventories and source terms in the high-enrichment and high-burnup regimes. This section summarizes the key issues identified in this review that are important to understanding the data parameters and phenomena important to high-burnup fuel, briefly discusses the work performed to date, and identifies areas that need further review to address possible shortcomings in current calculational methods and data and to reduce computational uncertainties. No priority or ranking has been assigned to these areas since their relative importance to code performance in the high-burnup regime has not been fully quantified.

Experimental Assessment Activities

- Add the low- and moderate-enrichment and burnup isotopic assay data from the Yankee-Rowe, Obrigheim (pellet assays), and Garigliano reactor fuels to the current isotopic validation database.
- Investigate the applicability of the new high burnup isotopic assay data from DOE measurements and the ARIANE Dodewaard (DU1) and Gosgen measurements for commercial spent fuel validation and incorporate the results into the validation database as appropriate.
- Continue sensitivity-based methods development (described in Appendix A) as a means of establishing a technical basis for the applicability of assay data, and better understanding and quantifying the trends in code bias and uncertainties for higher-enrichments and higher-burnup regimes.
- Re-analyze the existing isotopic benchmarks using current methods and cross-section libraries, and update the existing validation reports.
- Provide analysis and consulting support to the planned decay-heat measurements in Sweden. Pursue acquisition of more extensive decay-heat data for higher-enrichment and higher-burnup fuels for a wider range of assembly designs and cooling times.
- Review the GE-MO assembly radiation level measurements and evaluate the applicability of these experiments to the validation of source-term predictions for high-burnup SNF.

Nuclear Data and Modeling Activities

- Extend the nuclide importance ranking studies to include a wide range of enrichments and burnups, and includes a comparison of PWR and BWR assemblies. This task was identified as an essential first step in assessing the uncertainties associated with high-burnup spent fuel by considerably reducing the number of potentially important actinides and fission products that need to be studied. This activity is largely complete with the extension of the previously nuclide ranking study[34] published in Ref. 1.
- Perform an intercomparison of different calculational methods (1-D and 2-D flux solvers) to assess the merits of multidimensional methods as applied to calculating isotopic inventories for advanced fuel assembly designs, and higher-enrichment and high-burnup fuel.
- Compare nuclear decay and cross-section evaluations from several independent sources for the most important nuclides in each of the application areas. This would likely involve approximately 20 to 30 nuclides.
- Investigate the importance of resonance cross-section shielding issues in high-burnup fuel depletion.
- Evaluate the importance of the point-depletion approximation to model high-burnup fuel assembly characteristics. This includes the effects of assembly modeling approximations required for some complex

assembly designs (e.g., fuel homogenization), and the effects attributed to point depletion (using an average assembly burnup to represent fuel compositions).

- Review accuracy and completeness of the photon emission data for the key nuclides in shielding. Significant improvements in the calculated gamma-ray sources that could be achieved by upgrading the photon data of other dominant nuclides in shielding analyses should also be considered.
- Review the accuracy and completeness of the neutron source data and spectral data. Identify data and processes important to the production of ^{244}Cm, a nuclide that exhibits a dramatic increase in importance for decay heating and shielding in the high-burnup regime. Accurately predicting ^{244}Cm is a computational challenge due to the large number of nuclide precursors leading to its production, and identifying high-burnup issues that could adversely affect the accuracy of present calculational data and methods needs to be reviewed further.
- Evaluate the important structural activation products (mainly ^{60}Co) in high-burnup fuel assemblies using impurity levels that are representative of the newer assemblies.

6 SUMMARY AND CONCLUSIONS

As fuel assembly designs and operational limits evolve to allow higher initial enrichments and higher discharge burnups, the current models and nuclear data used to predict the isotopic compositions and source terms need to be reevaluated. The source terms of interest are those utilized in license reviews, including thermal (decay heat), shielding, confinement, and subcriticality analysis related to the transport, storage, and disposal of spent nuclear fuel. This report reviews some of the background issues, presents the perceived challenges to the calculational methods, and identifies initial areas for study. The technical issues related to the accuracy of predicted isotopic inventories and associated source terms for high-burnup LWR fuel are currently being investigated to determine the adequacy of predicted source terms and provide technical guidance on the appropriate models, analysis methods, and data that can be used.

Due to a projected lack of a sufficiently comprehensive experimental database for high-burnup fuel, a sensitivity-based methodology is currently being explored as a tool to assist in identifying the key parameters and governing phenomena important to accurately predicting high-burnup spent fuel properties. It is hoped that sensitivity-based methodologies can be used to identify important areas of potential concern, to better understand the impact of known uncertainties on responses of interest, to assist in reducing uncertainties in regimes of measured experimental data, and to estimate uncertainties appropriate for use beyond the regime of measured data. This methodology appears to be promising, and further work in applying these methods to support the high-burnup spent fuel program is proceeding with emphasis in the following areas: (1) apply sensitivity coefficients to assist in the development of a technical basis for establishing the range of application for experimental data, (2) help identify areas that are currently deemed to be underrepresented by experimental data, and (3) apply sensitivity techniques to obtain a better interpretation of code uncertainties and biases in regions with a limited amount of data.

In parallel with the sensitivity-based studies, a review of computational methods and modeling issues is recommended to assess the potential impact of the more complex LWR fuel designs, extended operating regimes, and core loading schemes associated with modern commercial reactor operations, which is not addressed by the sensitivity methods. An investigation of modeling issues will specifically address the ability of existing methods to accurately represent the increasingly complex fuel designs used with higher enrichment and high burnup fuel. Combining these two approaches will provide a comprehensive method to assess code performance in extended fuel regimes.

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APPENDIX A

APPLICATION AND DEMONSTRATION OF SENSITIVITY-BASED METHODS TO DEPLETION ANALYSIS

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APPLICATION AND DEMONSTRATION OF SENSITIVITY-BASED METHODS TO DEPLETION ANALYSIS

Since the 1970s perturbation methods have been developed and applied as a means of determining the sensitivities of the input variables and data parameters on the calculated quantities for applications of interest. Use of these methods can provide code developers and users with valuable information and insight into the physical behavior of a calculational model by identifying the governing phenomena and key parameters for which accurate values are required.

A.1 Background

Sensitivity-based techniques, combined with uncertainty information for the data parameters (e.g., cross sections and decay constants), are currently being explored as a potential basis for estimating calculational uncertainties and biases in the high-burnup spent fuel regime, where there is currently limited experimental isotopic assay data for code validation. As fuel enrichments and discharge burnup extend beyond the presently validated regime, the accuracy of the calculational methods and data used to predict the isotopic compositions and spent fuel properties becomes increasingly uncertain. Even with the acquisition of new data likely to become available in the near term, the number of chemical assay measurements in the high-burnup regime is significantly less than that in the conventional (lower)-burnup regime, and a statistical analysis of code uncertainty based solely on limited data in the high-burnup regime could prove challenging.

This appendix briefly describes the development and application of a sensitivity-based methodology to depletion analysis calculations. Since November 1999, ORNL has been developing and testing a modified version of the ORIGEN-S code with the capability of providing sensitivity information. Sensitivity techniques are being used to identify the key nuclear data parameters and governing phenomena important to fuel depletion and decay analysis to better understand code behavior and calculational bias associated with different enrichment and burnup regimes. Ultimately, the aim is to be able to combine the limited amount of high-burnup measurements likely to become available in the near term with sensitivity and uncertainty methods to provide a reliable and relevant approach to establishing and extending the range of application of available experimental data used to support code validation.

A.2 Previous Studies

Sensitivity analysis has been successfully applied to depletion analysis codes in the past.[1,2,3] An adjoint capability using perturbation theory was developed for the ORIGEN isotope generation and depletion code by Williams and Weisbin[1] and used to calculate sensitivity profiles and uncertainties for the parameters of the time-dependent nuclide concentrations. Worley and Wright[2] subsequently applied a different methodology to the ORIGEN2 code to obtain time-dependent relative sensitivities using the GRESS code system that adds derivative-taking and derivative-propagating capabilities to FORTRAN code. More recently, the French have applied and tested sensitivity and uncertainty methods to estimate the total uncertainty in fission product decay heat predictions that are attributed to the uncertainties in the basic nuclear decay data.[4]

A.3 Sensitivity Techniques

The accuracy of spent fuel depletion analysis methods depends to a large extent on the accuracy of the nuclear data and decay parameters important to the prediction of the dominant isotopic inventories for the response type and application regime of interest. For burnup-credit applications that involve fissionable systems (i.e., spent fuel) the response of interest is the neutron multiplication factor, k, and the key isotopes are the dominant neutron absorption and fissionable nuclides. The data parameters of greatest importance are the cross sections (e.g., neutron capture, fission, etc.) and decay data (e.g., half-lives, branching fractions, etc.) that define the isotopic generation and depletion rates and consequently the final concentrations of the key isotopes in the spent fuel.

Depletion codes such as ORIGEN use immense quantities of nuclear-decay data and cross-section data to represent the behavior of more than 1600 individual nuclides. Sensitivity methods provide a means to identify the nuclear-decay data, and cross sections are contributing most significantly to a calculated quantity, identify the dominant production and decay chains, determine trends in the different production chain importance with changing operational conditions such as burnup, and allow quantitative estimates of the uncertainty in the calculated quantities attributed to uncertainties in the basic nuclear data to be made. Sensitivity analysis methods are also being explored and applied as a potential method for extrapolating the bias and uncertainty established for conventional-burnup fuel to the higher-enrichment and higher-burnup regimes. This is accomplished by assessing the degree of similarity between different spent fuel systems based on the sensitivity coefficients (importances) of the underlying properties of a system – the basic nuclear reaction and decay parameters for fuel in the low- and high-burnup regimes as they relate specifically to the nuclides of importance to the prediction of k. A response that exhibits very similar dependencies to the underlying nuclear data and reaction/decay processes in two separate systems involving spent fuel with different enrichment and burnup combinations will likely have similar uncertainties or biases in the calculated response since any biases attributed to the nuclear data will have a similar effect on the response in both systems. Conversely, two systems that have significantly different fuel compositions, or exhibit different sensitivities to the underlying data and processes, may be expected to have biases and uncertainties which are largely unrelated to one another. For example, a regime where fission products are the dominant contributor to the response will exhibit highest sensitivities to the processes and nuclear data that influence the concentrations of the dominant fission products. Comparing these sensitivities with a system where actinides are the dominant contributor to the response will show large differences in the parameter sensitivities since the important data parameters that affect the response in the respective regimes are different.

A.4 Implementation

Recently, ORNL has applied the GRESS 3.0 (Gradient Enhanced Software System) code system[5,6] developed in the 1980s to obtain sensitivity coefficients for the nuclide densities and time-dependent radiological properties calculated by the ORIGEN-S code. GRESS employs a FORTRAN precompiler that automatically processes existing programs and adds derivative-taking capabilities by repeated application of the calculus chain rule. The procedure allows the derivative of any real floating-point variable used in the code to be calculated with respect to any other variable or data parameter. Partial derivatives are calculated for each assignment statement in the code and propagated based on the arithmetic operations performed. The partial derivatives are the normalized first derivatives of the output variable with respect to the input variables or data parameters. The relative (normalized) sensitivities are derived from the derivatives by taking the product of the derivative and the parameter value and dividing by the response value. The relative sensitivities represent the change in the calculated value with respect to a change in the data parameter, and therefore provide a direct measure of the parameter importance to that application. To first order, a sensitivity of 1.0 means that a 1% change in an input or data parameter will cause a 1% change in the result.

The derivatives calculated by GRESS ($dR/d\alpha$) represent the change in a response, R (e.g., nuclide number density or neutron multiplication factor), with respect to a parameter, α, on which the response depends (e.g., a cross section or decay constant). The sensitivity coefficient, S, is defined as

$$S = \frac{dR/R}{d\alpha/\alpha},$$

where R is the response of interest, and α is the data parameter of interest.

The main advantage of derivative propagating codes, such as GRESS, is that it will operate on existing ANSI standard FORTRAN 77 coding and generally require few changes in the original coding to make the source compatible with the GRESS precompiler. After the enhanced source with derivative-taking capabilities has been generated using the precompiler, it is linked with FORTRAN and C language GRESS subroutines using a standard FORTRAN compiler. The resulting enhanced program will execute identically to the original source program with the option of also calculating derivatives.

The alternative approach to sensitivity analysis using adjoint theory generally requires extensive knowledge of the model equations and often requires a considerable implementation and code development effort. The advantage of the adjoint sensitivity methodology, however, is that response sensitivities can be obtained with respect to all parameters in the model using a single forward mode and reverse mode (adjoint) calculation. A limitation in the GRESS methodology is that the practical number of parameter sensitivities that can be obtained in a single calculation is about 200. For complex models involving large quantities of data, such as depletion analyses, this restriction requires that the user have some a priori knowledge about the subset of data parameters that are most likely to have the greatest impact on the calculated results. Ranking studies, such as those described in Refs. 7 and 8, are currently being applied to determine the nuclides that are dominant in each of the application areas to reduce the number of nuclides and input data parameters that need to be considered in the sensitivity analysis. Sensitivities for larger classes of data parameters (>200) are currently being obtained by repeated sensitivity calculations.

A.5 Sensitivity Parameter Development

The sensitivity of a calculated response to each nuclide, reaction type, and decay parameter represents a potentially large amount of information which is too large to be of general use. Therefore, a method of obtaining a global measure of the similarity between two systems is being investigated using the combination of individual data parameters sensitivities that are most important in the calculations. The parameter type described here is based on the so-called E-parameter, recently applied at ORNL to assess the degree of similarity between different fissionable systems for criticality safety validation studies.[9,10] The E-parameter is defined as follows:

$$E = M^{-1} \sum_{j=1}^{N} \sum_{i=1}^{R} S_{aij} S_{eij},$$

where

$$M = \{ \sum_{j=1}^{N} \sum_{i=1}^{R} (S_{aij})^2 \sum_{j=1}^{N} \sum_{i=1}^{R} (S_{eij})^2 \}^{1/2}$$

and S is the sensitivity coefficient of a calculated result to a specific reaction cross section or decay parameter R and nuclide N, and the subscripts a and e represent the two different systems (or, in this case, fuel with different enrichments and/or operating- and cooling-time regimes) of an *application* and *experiment*. An experimental

system may refer to spent fuel with an enrichment and burnup regime for which measured data are available or calculational biases are well established. The application system refers to a spent fuel regime for which there are no experimental data or perhaps limited experimental data. The E-parameters provide a global measure of similarity between two different systems and have limits of 0 (no similarity) and 1 (identical systems). The E-parameters, as defined here, currently do not include data uncertainty information and therefore are equivalent to assuming all uncertainties are of equal magnitude.

A second parameter type being evaluated is the T-parameter, which provides a measure of the relative importance of an individual parameter, or subgroup of parameters, in one system compared with another system. The T-parameter is similar in many respects to the E-parameter and is most easily defined in terms of the partial E-parameter, or dE, defined as follows:

$$dE = M^{-1} S_a S_e ,$$

where M is the normalization factor defined above and S_a and S_e are the sensitivity coefficients for an individual parameter in the application and experimental systems respectively. The T-parameter is effectively the ratio of the partial E-value of the parameter in the two different systems (above) to the partial E-value of the parameter with respect to the same system, and is defined as:

$$T = M^{-1} S_a S_e^{-1} \sum_{j=1}^{N} \sum_{i=1}^{R} S_{aij}^2 ,$$

where the parameter provides a measure of the relative importance of a parameter in an application system with respect to the experimental system and has a value of 1.0 for a parameter that has the same relative importance in both systems, <1.0 if the parameter is less important in the application regime, and >1.0 if it is more important in the application regime. Parameters with T-values greater than about 0.95 may be considered reasonably similar in the two systems or regimes. That is, the relative importance of the parameter in the application is at least 95% of that in the reference experimental system.

A.6 Actinide Inventory Sensitivities

As a demonstration of the application of sensitivity-based methods to depletion analyses, the enhanced version of ORIGEN-S, with derivative-taking capabilities added by GRESS, was used to calculate sensitivity coefficients for the key nuclear-decay data and cross-section parameters in a depletion analysis study. The focus of the demonstration was on the predicted concentrations of the major actinides, namely the uranium and plutonium isotopes, plus ^{241}Am (a burnup-credit isotope) and ^{244}Cm (an important decay heat and neutron source). The sensitivities of the calculated atom number densities (concentrations) for these nuclides to the nuclear data parameters applied in the depletion model were generated as a function of increasing burnup. The trends in the relative sensitivities with burnup provide a measure of the changing importances of the parameters to the calculated value. Sensitivities that increase in magnitude with burnup indicate that any uncertainties or bias in that parameter will have a larger effect on the results at higher burnup than at lower burnup. Conversely, decreasing sensitivities suggest that any errors in the data will play a reduced role in the calculated result.

At this point nuclear data uncertainties have not been incorporated into the methodology. Combining relative sensitivity trends, such as those presented here with uncertainty data (e.g., cross sections, fission yields, decay data), can provide additional information that may be useful for estimating trends in the uncertainty and bias associated of predicted concentrations or response.

In this demonstration the sensitivity profiles were calculated for a generic LWR fuel assembly having an initial enrichment of 3.0-wt % ^{235}U. The depletion calculations were performed using a nominal power of 35 MW/t and extended to a maximum burnup of 75 GWd/t. The results presented here are only intended as a demonstration to illustrate typical trends in the sensitivity coefficients for several important data parameters with burnup. Relative sensitivity profiles are illustrated for several of the nuclear data parameters important to depletion analyses, including the decay constants (half-lives), neutron-capture cross sections, fission cross sections, and (n,2n) cross sections.

A.6.1 Decay Constants

The relative sensitivities of the major actinide concentrations to the decay constants (λ) in the actinide decay chains are generally small (e.g., < 0.1). The notable exception is the importance of the decay constant of ^{241}Pu on the calculated inventory of ^{241}Am. The relative sensitivity of this parameter is about 1.0 near the beginning of the cycle, and exhibits only a gradual decrease to 0.98 at 75 GWd/t. Therefore, even though the relative sensitivity to this parameter is high, no adverse impact on the accuracy of the calculated ^{241}Am inventory with increasing burnup is associated with the decay constant.

The trends in the relative sensitivity of the predicted ^{235}U concentration at discharge to the decay constants of the major actinides is shown in Figure A.1 for illustrative purposes. The decay constant of ^{239}Np is dominant for most actinides due to its importance in the holdup of ^{239}Pu production in the chain $^{239}U \rightarrow {}^{239}Np \rightarrow {}^{239}Pu$, although the magnitude of the sensitivity is still relatively small (<0.02). The positive sensitivity indicates that an increase in the decay constant of ^{239}Np will result in an increase in the predicted ^{235}U concentration, and this sensitivity increases nearly linearly with burnup. An increase in the ^{239}Np decay constant (decrease in half-life) will result in an increased rate of ^{239}Pu production and a higher ^{239}Pu concentration, and therefore a higher ^{239}Pu fission rate. This higher ^{239}Pu fission rate in turn reduces the ^{235}U fission rate required to maintain a fixed power level, and consequently leads to a higher ^{235}U density. Conversely, an increase in the decay constant of ^{241}Pu, a major fissile isotope, to ^{241}Am, will result in a lower fissile plutonium concentration and consequently require a higher rate of ^{235}U fission to maintain a given power level, resulting in a lower ^{235}U concentration.

These examples are provided for illustrative purposes only. In general, the sensitivity of the predicted actinide concentrations to the decay constants is relatively small. Also, the values of the important decay constants are generally well known (low uncertainties). Therefore, these parameters are considered to be of relatively low importance to the overall uncertainties in depletion calculations. In general, the decay constants take on increasing importance during decay (not considered in this demonstration) since radioactive decay is the only process important to the buildup and decay of radionuclides.

A.6.2 Capture Cross Sections

The relative sensitivities of the predicted actinide concentrations considered in this study to the capture cross sections (σ_γ) of the burnup chain actinides exhibit significant variations as a function of burnup and are of a larger magnitude than the sensitivities observed for decay constants. As two illustrative examples, the sensitivity profiles for ^{239}Pu and ^{244}Cm production are shown in Figure A.2 and Figure A.3, respectively. The sensitivity profiles for the predicted ^{239}Pu concentration are only shown for the capture cross sections of ^{238}U and ^{239}Pu. The cross sections for all other actinides in the actinide chains have relative sensitivities less than 0.02 and are therefore not shown. Figure A.2 illustrates the relatively large and positive sensitivity of the ^{239}Pu concentration to the ^{238}U capture cross section (about 1). The sensitivity to the ^{239}Pu capture cross section is about –0.3, with the negative value indicating a reduction in the predicted concentration due to increasing the capture cross section. Both of these results are fairly intuitive.

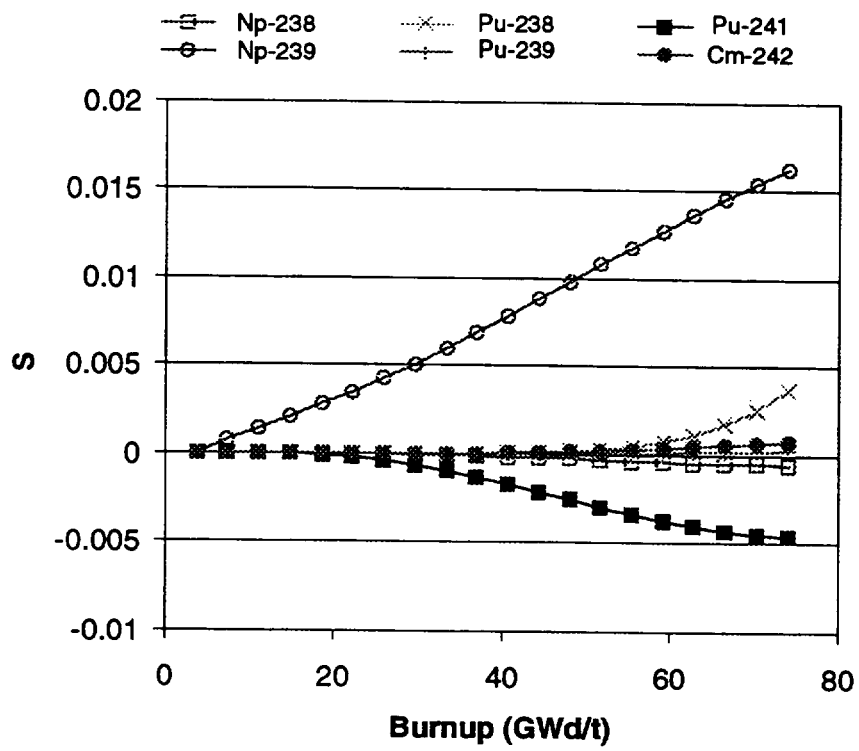

Figure A.1 Relative sensitivity, S, of predicted ^{235}U concentration on decay constants as a function of burnup

The relative sensitivity of the predicted ^{244}Cm concentration to the actinide capture cross sections (precursors to ^{244}Cm) is illustrated in Figure A.3. The ^{244}Cm concentration is very sensitive to a relatively large number of cross-section parameters, which is attributed to the large number of capture and decay precursors of ^{244}Cm. The sensitivity trends are also much less intuitive than in the previous example for ^{239}Pu. The figure illustrates that the ^{244}Cm concentration is highly sensitive to the capture cross sections of the major production chain nuclides including ^{239}Pu, ^{240}Pu, ^{241}Pu, ^{242}Pu, and ^{243}Am. However, the negative sensitivity to the ^{238}U capture cross section over much of the burnup range is counter-intuitive since an increase in the capture cross section might be expected to increase the ^{244}Cm. However, the increased production of the fissile isotopes ^{239}Pu and ^{241}Pu caused by an increase in ^{238}U capture ultimately results in a reduction in the neutron flux as the concentration of the fissile plutonium isotopes increases, caused by the large fission cross sections for the plutonium isotopes relative to ^{235}U. As the plutonium isotopes build up, the same total fission rate can be maintained with a lower flux.[†] The reduction in flux ultimately offsets the initial increased plutonium production (caused by an increase in the ^{238}U capture cross section) by reducing the flux (and therefore the reaction rates) responsible for the capture process that produce ^{244}Cm.

Note that the high sensitivity of ^{244}Cm to the ^{243}Am cross section (Figure A.3), and absence of a significant sensitivity to the ^{243}Cm cross section indicates that the production of ^{244}Cm is largely via the chain $^{243}Am \rightarrow {}^{244}Am \rightarrow {}^{244}Cm$.

[†] The flux computed by ORIGEN-S is based on the time-dependent compositions, the fission and capture cross sections, and the energy per fission. For fix-power calculations the computed flux, ϕ, is that required to produce the required power, P, given the relationship $P = 1.6 \times 10^{-19}\ \Sigma\, Q_{ij} N_i\, \sigma_{ij}\, \phi$, where N_i is the density of nuclide i, Q_{ij} is the recoverable energy of nuclide i and reaction j, and s is the reaction cross section. Consequently, an increase in the effective fission cross section will cause a corresponding decrease in the flux.

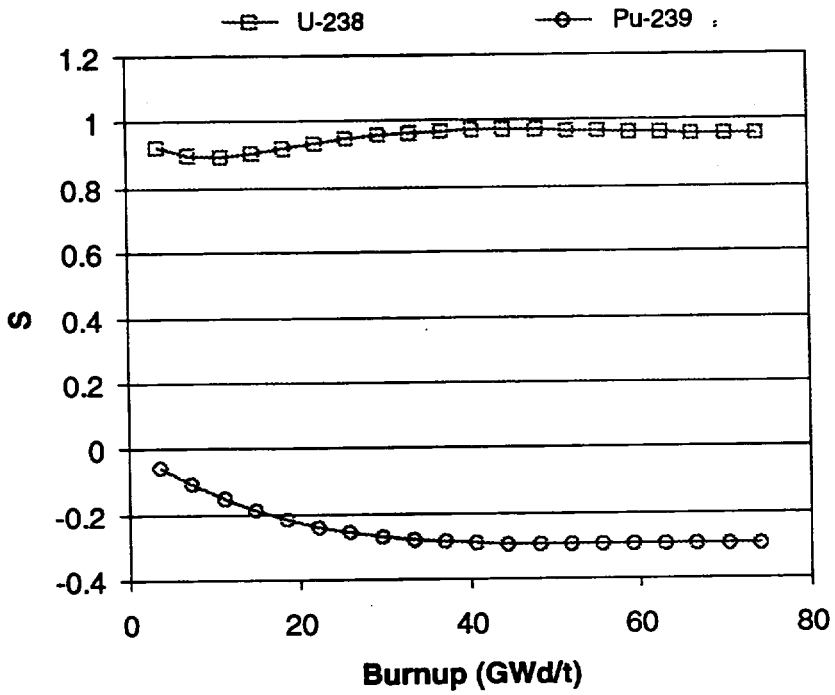

Figure A.2 Relative sensitivity, S, of predicted ^{239}Pu production to capture cross sections as a function of burnup

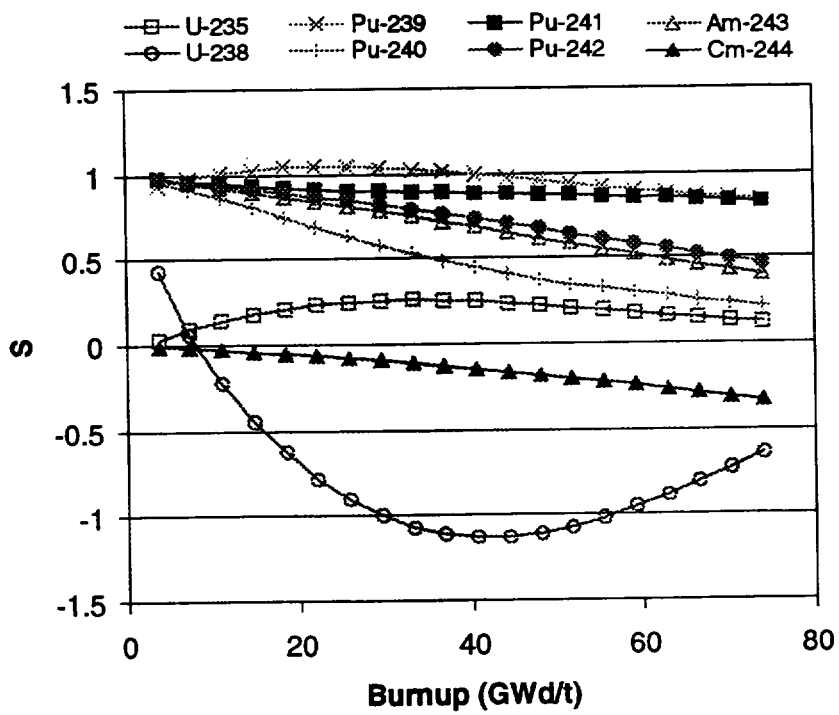

Figure A.3 Relative sensitivity, S, of predicted ^{244}Cm production to capture cross sections as a function of burnup

Although the observed behavior of the ^{244}Cm number density on the ^{238}U cross section is somewhat of an artifact of the calculational model, which employed a fixed power to define the irradiation history (rather than fixed flux), this model is commonly used in depletion calculations and thus represents the type of code behavior and parameter sensitivity expected in typical applications.

A.6.3 Fission and (n,2n) Cross Sections

The relative sensitivities of the predicted actinide concentrations to the fission cross sections (σ_f) of the actinides in the actinide burnup chains exhibit significant variations as a function of burnup. The predicted concentrations of the major fissile actinides ^{235}U, ^{239}Pu, and ^{241}Pu exhibit some of the largest sensitivities. For illustrative purposes the sensitivity profiles for ^{241}Pu production to the fission cross section of the major actinides are shown in Figure A.4. The profiles for ^{241}Pu are typical in shape of many of the plutonium isotopes studied. The sensitivity of the predicted ^{241}Pu concentration to the ^{239}Pu and ^{241}Pu fission cross sections exhibits an increasing negative sensitivity with increased burnup, but remains nearly constant for a burnup above about 40 GWd/t. The sensitivity to the ^{235}U fission cross section exhibits a very large negative value initially, decreasing in sensitivity with increasing burnup. The sensitivity to the ^{235}U cross section is related to the flux normalization discussed previously.

The sensitivity of the predicted concentration of ^{235}U to the fission cross sections shown in Figure A.5 indicates a trend towards an increasingly negative relative sensitivity with increased burnup, the direct result of the effect the fission cross section has on the concentration and the decreasing ^{235}U concentration with burnup (which increases the relative effect).

The only actinides exhibiting a significant sensitivity to the (n,2n) reaction cross sections were ^{238}Pu production from ^{238}U and ^{234}U production from ^{235}U. The relative sensitivity profiles of the predicted ^{234}U concentration to (n,2n) reactions are illustrated in Figure A.6. In general the sensitivities above 40 GWd/t remain relatively small (<0.01). However, these sensitivities must be combined with the uncertainties in the (n,2n) cross sections, which are considerably larger than for the dominant reactions, such as capture and fission, to assess the net impact of the changing sensitivities in the high-burnup regime.

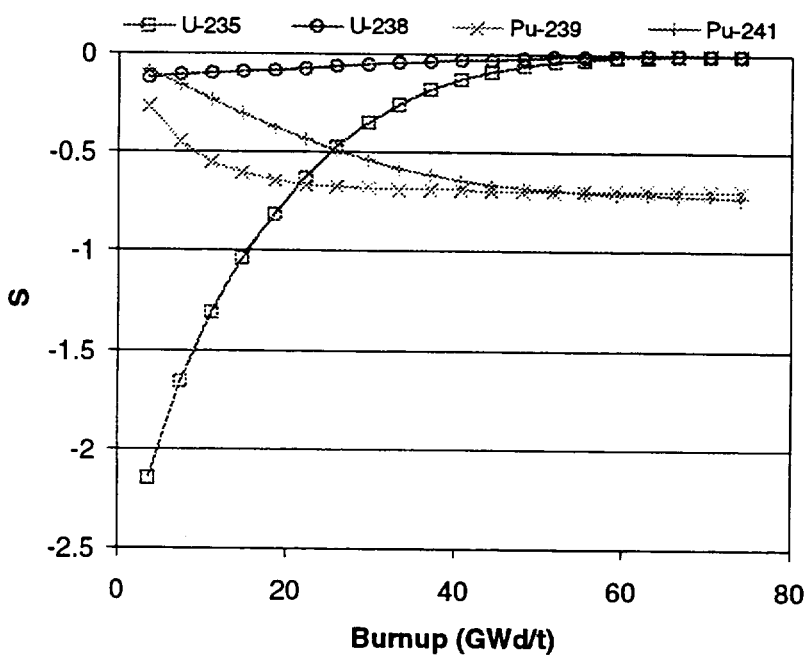

Figure A.4 Relative sensitivity, S, of predicted ^{241}Pu production on fission cross sections as a function of burnup

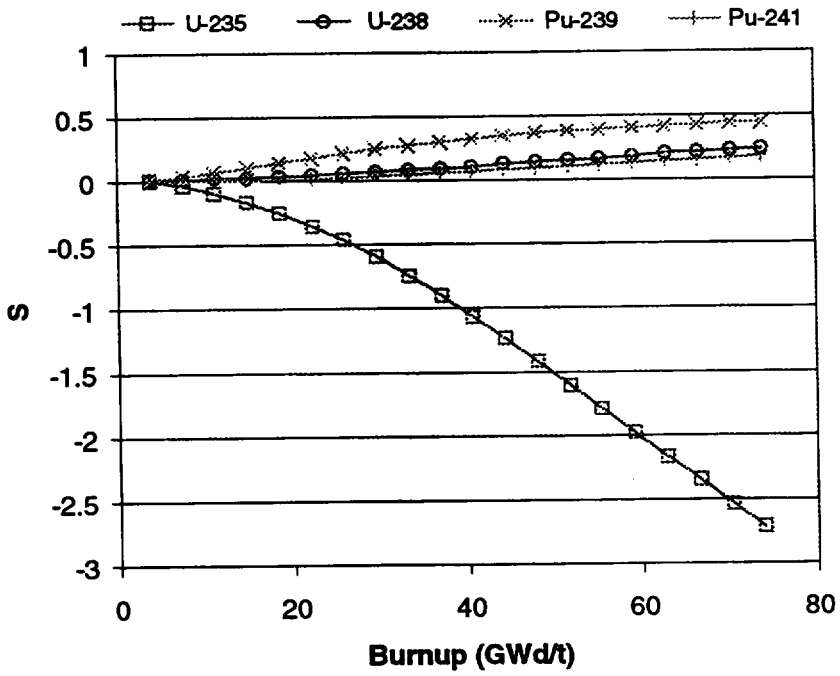

Figure A.5 Relative sensitivity, S, of predicted ^{235}U concentration on fission cross sections as a function of burnup

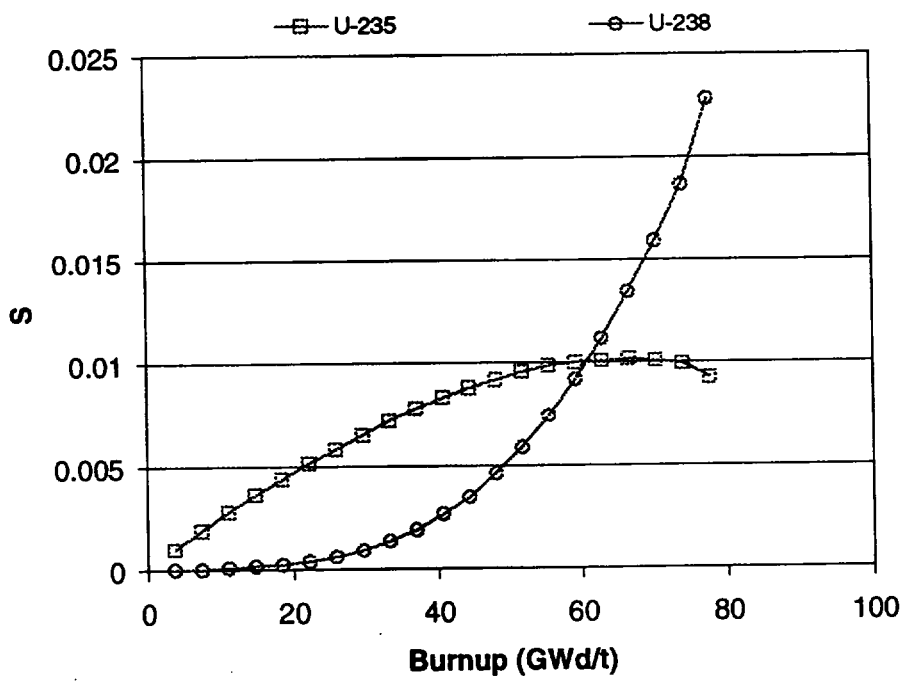

Figure A.6 Relative sensitivity, S, of predicted ^{234}U concentration on (n,2n) reaction cross sections as a function of burnup

A.7 Neutron Multiplication Factor Sensitivities

Sensitivity methods were also applied to investigate the degree of similarity of spent fuel having different enrichment and burnup combinations in terms of the importance of the neutron cross sections on the k_∞ of the system. The cross sections influence the k_∞ of spent fuel in two ways: (1) by affecting the compositions of the fuel during irradiation, and (2) by defining the rate of neutron absorption and fission in the spent fuel for the away-from-reactor environment.[‡] The sensitivities presented in this study include *only* the influence of actinide and fission product concentrations in the fuel. In total, sensitivities were obtained for 116 individual nuclide- and reaction-type combinations, which represent the key data parameters influencing spent fuel compositions important to the prediction of k in spent fuel.

The sensitivity coefficient was determined for a reference spent PWR fuel composition with 30 GWd/t and 3.5-wt % enrichment. This regime was deemed to be generally representative of the spent fuel assay data that has been applied to estimate isotopic biases and uncertainties attributed to code predictions for the important burnup-credit nuclides. The sensitivities for the 30-GWd/t and 3.5-wt % fuel were compared with those calculated for enrichment and burnup combinations that ranged from 3- to 5-wt % and 20 to 70 GWd/t for a reference cooling time of 5 years. Similarities between the different spent fuel regimes, expressed as the E-parameter with respect to 30 GWd/t and 3.5-wt % fuel, are listed in Table A.1 and are illustrated in Figure A.7 and Figure A.8.

Table A.1 E-parameter values for k_∞ relative to 30 GWd/t, 3.5-wt % fuel (5-year cooling)

Burnup	Initial enrichment (wt % ^{235}U)				
(GWd/t)	3.0	3.5	4.0	4.5	5.0
20	0.999	0.998	0.995	0.990	0.984
30	0.999	**1.000**	0.999	0.995	0.990
40	0.994	0.998	1.000	0.999	0.995
50	0.985	0.992	0.996	0.998	0.998
60	0.970	0.980	0.988	0.993	0.996
70	0.951	0.962	0.972	0.981	0.988

[‡] Neutron multiplication factor $k = \sum_i n_i \sigma_{i,f} \nu_i / \sum_i n_i \sigma_{a,i}$,

where n is the number density, σ is the cross section (f = fission, a = absorption), and ν is the number of neutrons per fission, summed over all nuclides i.

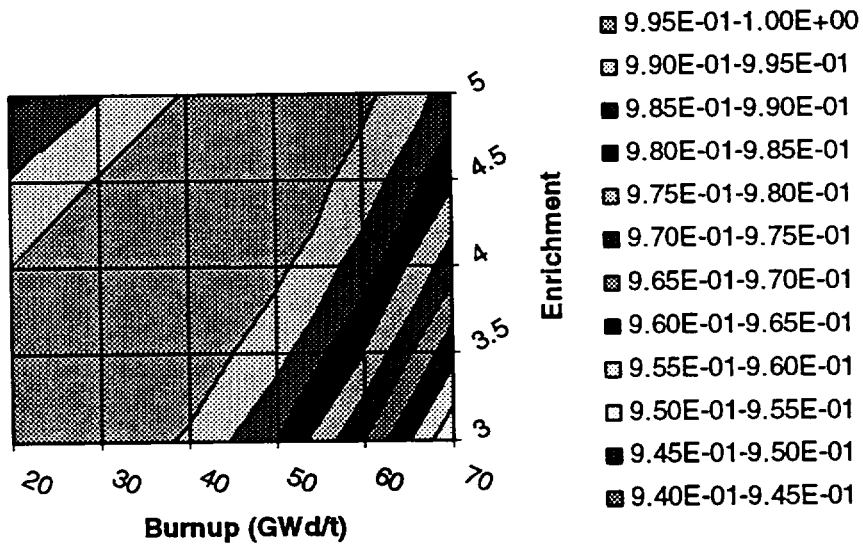

Figure A.7 E-parameter contours for k_∞ relative to 30-GWd/t, 3.5-wt % fuel (5-year cooling)

The E-parameter values indicate a remarkable similarity over a relatively wide enrichment and burnup domain, and most of the domain has an E value of greater than 0.97. As a comparison, the criterion currently used to establish similarity between different criticality systems (experimental benchmarks and applications) is an E-parameter value greater than 0.80. Although a suitable criterion for establishing similarity between the different enrichment and burnup regimes studied in this work has not been quantified to date, it is clear that such systems may be highly correlated over a relatively wide range. For example, if an arbitrary criterion of 0.97 were applied to this demonstration for 5-year-cooled fuel, it would suggest that validation data obtained for 30-GWd/t and 3.5-wt % fuel may be highly applicable to establishing code bias in calculated spent fuel compositions for most of the enrichment and burnup combinations studied. Only in a narrow regime that includes relatively low enrichment and high-burnup combinations (below 4 wt % and above 60 GWd/t) is the E parameter less than 0.97. Figure A.8 indicates that spent fuel with 3.5-wt % enrichment and 30 GWd/t is very similar to fuel with 4-wt % and 40-GWd/t, and 5-wt % and 50-GWd/t for the response (k) studied. The E parameters have also been found to exhibit very little variation with cooling time, with values that exceed 0.97 over most of the cooling times important to spent fuel transport and storage (<100 years).

These results provide a potential framework for establishing the range of applicability of validation data from one spent fuel regime to another. The basis for this applicability is that these systems exhibit highly similar sensitivities to the nuclear data parameters that drive the formation of the dominant nuclide compositions important to the k of the system. Perhaps the high degree of similarity between the different regimes is not so remarkable when one considers that many of the dominant burnup-credit nuclides are the major actinides which exhibit smooth and typically slow variations in concentration with increasing burnup.

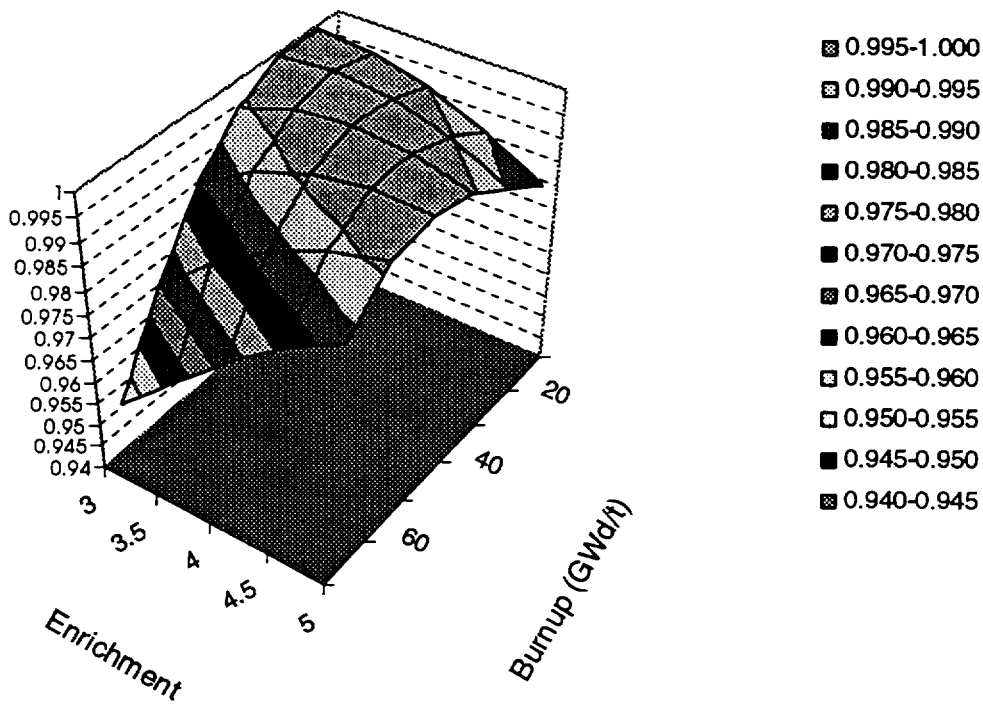

Figure A.8 *E*-parameter surface profile for k_∞ relative to 30-GWd/t, 3.5-wt % fuel (5-year cooling)

A ranking of the individual data parameter sensitivities that have the largest influence ($|S| > 10^{-4}$) on the nuclide composition in terms of their effect on *k* is listed in Table A.2 for the spent fuel with 30 GWd/t and 3.5 wt %, and a higher-burnup fuel of 50 GWd/t and 4.5 wt %. A review of the individual sensitivities can provide insight on which data parameters are being adequately exercised or "covered" in the reference regime, and which data parameters may be under-represented in the reference regime. A parameter that has a much greater sensitivity in the application regime compared with the experimental regime implies that the experimental regime does not provide a sufficient measure of that parameter. Consequently, any uncertainty or bias in that parameter could have a much larger adverse effect on the calculated *k* in the application regime. Conversely, a smaller sensitivity in the application area implies that the parameter is more important in the experimental regime and therefore any biases and uncertainties established for the parameter will be bounding in the application regime.

Table A.2 Comparison of sensitivity coefficients for two spent fuel regimes

Nuclide	Reaction type	30 GWd/t 3.5 wt %	50 GWd/t 4.5 wt %
^{238}U	(n,g)	2.710E–01	3.698E–01
^{239}Pu	(n,f)	–1.079E–01	–1.588E–01
^{239}Pu	(n,g)	–9.883E–02	–1.188E–01
^{235}U	(n,g)	–6.410E–02	–9.153E–02
^{235}U	(n,f)	–6.171E–02	–8.588E–02
^{240}Pu	(n,g)	5.005E–02	7.191E–02
^{241}Pu	(n,g)	–9.273E–03	–2.303E–02
^{241}Pu	(n,f)	–9.395E–03	–2.092E–02
^{155}Eu	(n,g)	5.022E–03	1.056E–02
^{103}Rh	(n,g)	5.312E–03	1.047E–02
^{238}U	(n,f)	7.730E–03	1.040E–02
^{149}Sm	(n,g)	9.495E–03	9.269E–03
^{150}Sm	(n,g)	–3.490E–03	–7.881E–03
^{236}U	(n,g)	–3.771E–03	–5.993E–03
^{143}Nd	(n,g)	2.665E–03	5.732E–03
^{237}Np	(n,g)	1.095E–03	4.608E–03
^{238}Pu	(n,g)	9.581E–04	4.183E–03
^{151}Sm	(n,g)	2.244E–03	3.524E–03
^{234}U	(n,g)	2.104E–03	2.591E–03
^{133}Cs	(n,g)	9.484E–04	2.305E–03
^{148}Pm	(n,g)	–1.502E–03	–2.047E–03
^{243}Am	(n,g)	1.913E–04	1.404E–03
^{154}Eu	(n,g)	–1.479E–03	–1.222E–03
^{241}Am	(n,g)	4.420E–04	1.117E–03
^{148m}Pm	(n,g)	–7.655E–04	–1.073E–03

As a further demonstration, *E*-parameters were calculated using total decay heat (Watts) as the response instead of *k*. The decay heat would be expected to exhibit a greater dependence on burnup than *k* due to the importance of ^{244}Cm to heat generation rates, which exhibit a significant burnup dependence. The results are illustrated in Figure A.9 for a reference cooling time of 5 years. The *E* parameter profiles for decay-heat generation show significantly greater variations over burnup and enrichment (Figure A.9) and suggest that the range of application for validation data obtained in one regime may be much more limited. Studies including cooling time have found that the *E*-parameters suggest dissimilar systems for even relatively small differences in cooling time. This comparison demonstrates that the degree of similarity over the spent fuel regime is highly dependent on the response type of interest. The degree of similarity in terms of the neutron multiplication factor appears to be much greater than for a decay-heat generation response, corroborating expectations.

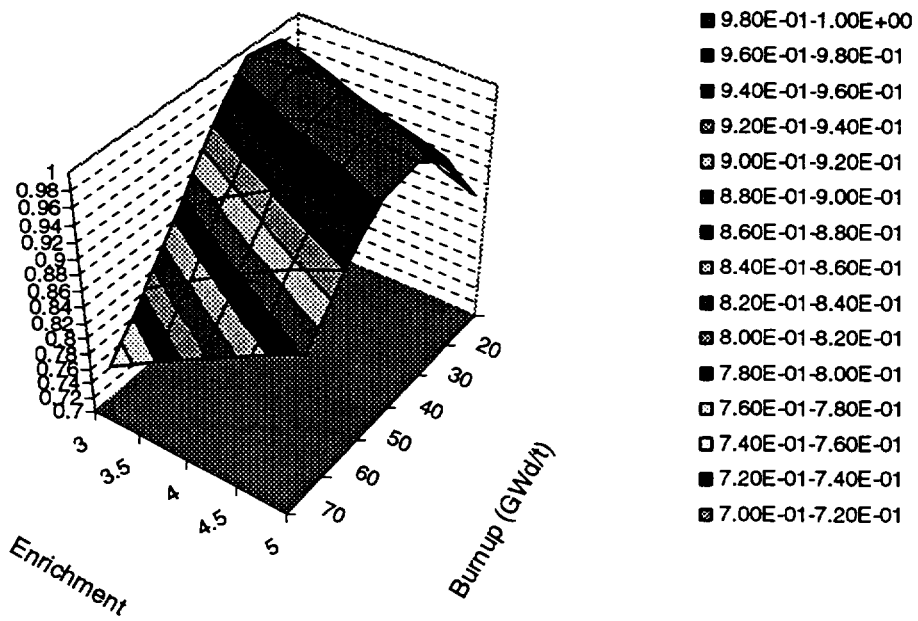

Figure A.9 *E*-parameter surface profile for decay heat relative to 30-GWd/t, 3.5-wt % fuel (5-year cooling).

A.8 Summary

The sensitivity-based techniques described appear to be a very promising tool for assessing the similarity of different spent fuel systems by evaluating their dependence on the underlying nuclear data that influence spent fuel compositions. Spent fuel regimes that exhibit a high degree of similarity suggest that validation data obtained in one regime may be highly applicable to another regime since the basic processes and nuclear data parameters that govern the spent fuel compositions as they affect the response of interest are largely the same. Efforts to further develop the methodology and analysis of the sensitivity coefficients are proceeding, with emphasis in the following areas: (1) apply sensitivity coefficients to assist in the development of a technical basis for establishing the range of application for experimental data, (2) identify spent fuel regimes that are currently deemed to be under-represented by experimental data, and (3) apply sensitivity techniques to obtain a better interpretation of code uncertainties and biases in regions with a limited amount of validation data.

The sensitivity studies described here only assess the influence of the underlying data parameters on the concentrations of the actinides and fission products as they effect the k of a spent fuel system. The studies do not address issues related to the potential direct effects of bias introduced by the cross sections in the away-from-reactor cask criticality safety evaluations (i.e., the reactivity worth of the individual actinides and fission products). Such studies are the subject of a parallel program that is evaluating the applicability of existing and planned criticality benchmark measurements to spent fuel cask transport and storage applications. A comprehensive sensitivity analysis will require a combination of these two sensitivity disciplines.

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APPENDIX B

EVALUATION OF APPLICABILITY OF ISOTOPIC ASSAYS OF HIGH-BURNUP H. B. ROBINSON AND LIMERICK-1 REACTOR FUEL TO GENERIC HIGH-BURNUP VALIDATION STUDIES

APPENDIX B

EVALUATION OF APPLICABILITY OF ISOTOPIC ASSAYS OF HIGH-BURNUP H.B. ROBINSON AND LIMERICK-1 REACTOR FUELS TO GENERIC HIGH-BURNUP VALIDATION STUDIES

This appendix describes the results of a sensitivity-based study to assess the applicability of several high-burnup commercial spent fuel assemblies currently being considered for isotopic analysis to support code validation of depletion analysis methods in the high-burnup regime. The study was performed to address concerns over the high burnup of the fuel with respect to the assembly enrichment and to provide recommendations on the potential usefulness of the samples for isotopic validation.

After the initial assessment of the H. B. Robinson fuel, new information was obtained regarding reconstitution of the fuel. This reconstitution involved removing candidate fuel rods from the original assembly and placing them with fresh fuel in a new configuration in order to achieve the artificially high burnup. The candidate H. B. Robinson fuel rods (2.90 wt-% ^{235}U) were irradiated in assembly G-38 and achieved a burnup of 47.5 GWd/t after 5 cycles. Twenty four of the rods were then reconstituted into subassembly S-15, along with 17 original rods (132 inches of 3.85% U-235 with 6 inches of 0.7% U-235 at each end), 4 gadolinia rods (10% Gd_2O_3 and 1.95% ^{235}U enrichment) irradiated for three cycles in Assembly S-31, and fresh rods. This reconstituted subassembly was then irradiated for two additional reactor cycles to achieve the final burnup of 72.9 GWd/t. The fact that the irradiated cycle 5 fuel rods were mixed with fresh fuel rods and partially irradiated gadolinia rods in a subassembly of only 45 rods introduces considerable assembly heterogeneity near the end of irradiation, which is significantly different than typical commercial reactor fuel which tends towards more uniform assembly compositions near the end of irradiation. It is not possible to accurately simulate this type of reconstitution using the 1-D methods widely used for spent fuel characterization. Ultimately, the complexity of the fuel reconstitution and the significant uncertainties it introduces would likely make it very difficult to confidently apply assay results from this reconstituted fuel to code validation for spent commercial fuel. Although the acquisition of isotopic data from the H. B. Robinson fuel is no longer highly recommended, this Appendix provides an illustrative example of sensitivity techniques that can be used to evaluate experimental data.

B.1 Background

The measured isotopic assay data currently available for spent nuclear fuel is limited to 46 GWd/t for PWR fuel and 34 GWd/t for BWR fuel. Measured isotopic assays for fuels having higher burnup are needed to assist in the validation of computational analysis methods. Fuel rods from two high-burnup assemblies currently are being considered for isotopic assay measurements: (1) a Siemens SPC 15 × 5 assembly design from the H. B. Robinson pressurized-water reactor (PWR) reactor having an initial enrichment of 2.90 wt-% ^{235}U and burnup of 73 GWd/t, and (2) GE-11 9 × 9 assembly from the Limerick-1 boiling-water reactor (BWR) reactor having initial enrichment pin splits of 3.40 to 3.95-wt % ^{235}U and a burnup range of 54 to 57 GWd/t. However, the applicability of these data to high-burnup spent fuel isotopic validation is of potential concern since the initial ^{235}U enrichments of these fuels are considerably lower than that normally associated with these burnup levels. In PWRs, enrichments below about 3 wt % are typically limited to a discharge burnup of about 35 GWd/t by the depletion of fissile material in the fuel. In typical reactor operations, an assembly burnup in excess of about 60 GWd/t will only be achieved with enrichments of 5 wt-% ^{235}U or more. Thus, the enrichment and burnup combinations of H. B. Robinson and Limerick spent fuel assemblies under consideration are fairly nontypical of normal discharged commercial reactor fuel (e.g., the assemblies are considerably "over-burned").

The relatively low enrichment of the H. B. Robinson fuel raises concerns relative to the applicability of the isotopic assay measurements to code validation in the high-burnup regime. For example: Would the calculated-to-experimental (C/E) isotopic inventory ratios determined from the H. B. Robinson fuel samples be representative of more typical high-burnup fuel having a higher initial enrichment? To what degree can the H. B. Robinson fuel characteristics be considered representative of typical high-burnup fuel, and in what areas is the fuel different? To help address these issues, sensitivity-based methods have been applied to depletion analysis simulations as a means of investigating the relative importances of the underlying nuclear data to the spent fuel concentrations of the dominant burnup-credit actinides important to the prediction of the neutron multiplication factor, *k*, for the H. B. Robinson fuel. Obviously, the basic physics that governs the depletion and decay processes do not change as the assembly burnup increases. However, there can be changes in the relative importance of the nuclear processes and data that can lead to increased uncertainties. To evaluate these potential changes, the sensitivity coefficients for H. B. Robinson fuel samples have been compared with those calculated for a hypothetical spent fuel sample having a more representative enrichment and burnup combination. These comparisons help to identify potential differences in the parameter sensitivities that could lead to different levels of uncertainty in the computational predictions for the two domains that could adversely affect predicted code bias based on the measured C/E ratios.

B.2 Actinide Concentration Sensitivities

Sensitivity coefficients were generated using a modified version of the ORIGEN-S isotope generation and depletion code application with derivative-taking capabilities added by the GRESS preprocessor (see Appendix A). The sensitivities were calculated for the key nuclear data parameters important to the prediction of the major actinide concentrations (atom number densities) for two cases: (1) fuel representative of the H. B. Robinson assembly having an enrichment of 2.90 wt % ^{235}U and burnup of 73 GWd/t, and (2) fuel having the same burnup as the H. B. Robinson fuel and an enrichment 5 wt %, a value more representative of high-burnup fuel. The original H. B. Robinson sensitivity study compared the two fuel regimes (different enrichments) using a so-called D-parameter as a measure of similarity in the earlier studies. These results are presented here. The D-parameters as defined here represents the difference of the sensitivities for the two cases and is defined as follows:

$$D = S_1 - S_2,$$

where S is the sensitivity coefficient for a particular parameter in the reference and application regime. The sign (positive or negative) of D is important because it indicates whether a given parameter is more or less sensitive in one system compared with the other. The main indicators on nonsimilarity in this evaluation are parameters with large positive sensitivities (i.e., parameters that have a higher sensitivity in the application regime than the experimental regime). An absolute criterion for establishing similarity, or lack thereof, has not been defined at this point. Based on initial trending analysis, it appears that an absolute value of S greater than about 0.1 can be considered a key parameter. A sensitivity of 0.1 means that a 10% change in the parameter will result in a 1% change in the response, in this case, the concentration of the particular actinide. Note that the T-parameters discussed in Appendix A are also used to evaluate differences in the individual parameter sensitivities.

The nuclear data and cross section parameters studied included (1) decay constants, (2) capture cross sections, (3) fission cross sections, (4) fast neutron (n,2n) reactions, and (5) neutron capture branching fractions. The parameter sensitivities were calculated for the isotopes of the elements U, Np, Pu, Am and Cm (e.g., ^{234}U – ^{244}Cm) for which nuclear data exist in the ORIGEN-S library. For each depletion analysis case sensitivity coefficients were calculated for 46 actinides with respect to each of the nuclear data parameter types listed above. Sensitivities were generated with respect to the atom number densities (concentrations in spent fuel) of 10 major actinides important to burnup credit, decay heat, and neutron source-term applications: ^{234}U, ^{235}U, ^{238}U, ^{239}Pu, ^{240}Pu, ^{241}Pu, ^{242}Pu, ^{241}Am, ^{238}Pu, and ^{244}Cm.

The D-parameter values for the two spent fuel cases involving 2.90- and 5-wt % enrichments, both with a burnup of 73 GWd/t, are tabulated for decay constants, capture cross sections, and fission cross sections, in Tables B.1 through B.3. Values greater than 0.1 are highlighted. Values for the (n,2n) cross sections and branching fractions are not listed since no values exceeded 0.1.

A review of the D parameters for these models indicates that the two calculations have a high degree of similarity, and that the importance of the major nuclear processes and nuclear data for these two systems are fundamentally the same. Many parameters exhibit significant relative variations in sensitivity between the two calculations, but have absolute sensitivities that are very low ($10^{-2} - 10^{-3}$). The initial criteria used for identifying potentially dissimilar parameter sensitivities were that the D value exceeded 0.1. However, it is also important to look at the relative sensitivity change since a D value of 0.1 represents a minor difference for a parameter with a relative sensitivity S that exceeds 1.0. Very few D values are greater than 0.1. The largest observed D value for the decay constants is ^{241}Pu decay to ^{241}Am. However, the decay constant of ^{241}Pu is known to high precision (about 0.7% in ENDF/B-VI), and no impact on the accuracy of the calculated of ^{241}Am between the two cases due to the decay constant is anticipated. The largest D value observed for the neutron cross sections is in the sensitivity of the ^{235}U concentration on the ^{238}U capture cross section and the ^{244}Cm concentration on the ^{235}U fission cross section. However, the sensitivities to these parameters are quite large (~3), and the relative change in the sensitivity is generally small. As an example, the relative sensitivity of the ^{235}U concentration on the ^{238}U capture cross section is about 2.8 for the 2.90-wt % fuel after a cooling time of 5 years. The sensitivity of the same parameter for 5-wt % fuel is about 1.9, which represents about a 30% change in the sensitivity coefficient. The magnitude of the relative sensitivity indicates that either case will be sensitive to the accuracy of the ^{238}U capture cross section. Furthermore, since the sensitivity to the ^{238}U capture cross section is actually higher in the case of H. B. Robinson fuel than typical high-burnup fuel, any calculational biases for ^{235}U attributed to the ^{238}U capture cross sections based on the analysis of 2.90-wt % fuel will be bounding if applied to analyses using 5-wt % fuel. Therefore, any trends in the D parameters must also be considered when evaluating different systems for applicability.

B.3 Neutron Multiplication Factor Sensitivities

Subsequent sensitivity studies were conducted to evaluate the similarity of the two spent fuels in terms of the neutron multiplication factor, k, using the E-parameter (see Appendix A) as the measure of global similarity. The study included sensitivity coefficients for all of the major burnup-credit actinides and fission products.

The E-parameter results are illustrated in Figure B.1 and show the similarity of the spent fuel regimes in terms of the importance of the data parameters to the prediction of the spent fuel compositions important to k. The E values are a global measure of system similarity between a moderate enrichment and high-burnup reference fuel (3 wt % and 70 GWd/t) and fuels having higher enrichments. The E values are seen to vary more gradually with changing enrichment than with burnup as the parameters are varied from the reference position. The E value for the H. B. Robinson fuel (2.9 wt % and 73 GWd/t) and more characteristic high burnup fuel is about 0.98, a value considered to represent a relatively high degree of similarity between the two regimes.

Table B.1 D values for actinide decay constants

Nuclide decay constant	D-values for the production of response actinides									
	U-234	U-235	U-238	Pu-239	Pu-240	Pu-241	Pu-242	Am-241	Pu-238	Cm-244
U234	2.25E–05	3.70E–07	8.37E–10	2.20E–10	–2.13E–09	–2.65E–09	–1.31E–08	–7.66E–10	–3.36E–09	–4.39E–08
U235	–6.76E–09	1.21E–08	3.67E–11	3.92E–11	–7.86E–11	–8.56E–11	–5.65E–10	–9.20E–11	4.28E–10	–1.90E–09
U236	–3.07E–08	–1.08E–09	–8.01E–12	1.59E–09	1.01E–09	7.64E–10	4.75E–10	7.19E–10	5.05E–08	5.21E–10
U237	1.01E–04	–3.42E–05	–6.02E–07	1.70E–05	4.40E–06	–6.02E–07	2.63E–06	–2.01E–06	4.17E–04	2.48E–05
U238	–8.18E–06	–7.74E–08	1.41E–09	7.66E–10	1.18E–09	1.30E–09	3.68E–09	5.85E–10	2.26E–09	1.12E–08
U239	–2.02E–05	–7.48E–05	–1.34E–06	7.93E–05	–8.16E–07	3.79E–06	1.67E–05	6.89E–07	2.67E–05	5.80E–05
Np237	–5.24E–07	–3.18E–08	–3.96E–10	1.12E–08	8.44E–09	7.11E–09	9.12E–09	5.62E–09	3.44E–07	2.18E–08
Np238	–1.92E–03	5.52E–04	9.98E–06	–3.32E–04	–2.43E–04	–2.07E–04	–2.48E–04	–1.67E–04	–5.95E–03	–5.58E–04
Np239	–2.87E–03	–1.11E–02	–1.99E–04	9.61E–03	6.02E–03	6.54E–03	8.10E–03	6.06E–03	4.45E–03	1.43E–02
Np240	–5.20E–08	–1.71E–07	–3.04E–09	4.55E–10	9.20E–07	–1.87E–07	–1.00E–07	–1.88E–07	4.57E–08	–9.42E–08
Pu236	–1.69E–07	–1.70E–08	–2.70E–10	1.30E–10	7.37E–10	1.06E–09	4.26E–09	–2.98E–10	5.36E–09	1.43E–08
Pu237	–1.22E–07	–9.68E–09	–1.68E–10	–1.20E–08	–7.42E–09	–5.44E–09	–5.18E–10	–6.10E–09	–3.68E–07	7.76E–09
Pu238	–3.18E–01	–7.93E–04	1.47E–06	2.42E–04	1.57E–04	1.19E–04	4.02E–05	1.23E–04	4.61E–02	–5.75E–05
Pu239	1.18E–06	–3.72E–04	1.56E–07	1.54E–04	1.35E–05	1.41E–05	1.31E–05	1.44E–05	–6.32E–06	6.91E–06
Pu240	–5.30E–06	2.48E–05	4.43E–07	–1.38E–06	5.90E–04	7.73E–05	7.09E–05	7.89E–05	–4.76E–05	4.63E–05
Pu241	–2.25E–02	3.97E–03	7.16E–05	–1.79E–03	–1.24E–03	2.57E–01	7.95E–03	–8.78E–01	–8.30E–02	1.12E–03
Pu242	2.13E–09	7.25E–09	–1.39E–08	–2.87E–09	4.02E–08	7.08E–10	1.10E–05	1.45E–09	–2.60E–09	1.40E–06
Pu243	1.16E–07	3.75E–07	6.68E–09	3.00E–08	4.39E–06	2.72E–07	2.17E–08	3.15E–07	–1.28E–07	1.44E–04
Am241	3.32E–06	1.02E–06	1.82E–08	–2.58E–07	1.34E–07	–1.67E–07	8.91E–06	4.57E–03	1.59E–05	1.07E–05
Am242m	–5.72E–06	5.14E–07	9.27E–09	–2.25E–07	–1.46E–07	–1.51E–07	–6.69E–06	–1.24E–07	–3.54E–05	5.96E–07
Am242	–8.13E–05	3.41E–05	6.12E–07	–7.93E–06	–5.51E–06	–5.85E–06	–5.14E–05	–3.57E–06	–3.17E–04	3.53E–06
Am243	–5.45E–07	–1.85E–06	–3.28E–08	–5.04E–05	–5.48E–07	–1.68E–06	–4.28E–07	–1.79E–06	6.04E–07	6.03E–05
Am244	2.20E–05	7.58E–05	1.35E–06	–6.75E–07	–6.52E–05	–1.10E–05	–2.39E–05	–3.82E–06	–2.70E–05	–2.02E–03
Cm242	–4.83E–05	–3.68E–04	–6.28E–06	–5.16E–04	–3.92E–04	–3.31E–04	–1.01E–04	–3.51E–04	1.15E–02	6.42E–04
Cm243	1.76E–08	9.01E–08	1.69E–09	–2.65E–05	–1.42E–06	–1.30E–06	–7.26E–07	–1.25E–06	–6.99E–08	5.21E–06
Cm244	–4.60E–05	–1.16E–04	–2.06E–06	7.65E–07	–2.69E–02	–2.02E–03	–9.19E–04	–1.96E–03	–4.37E–06	2.17E–01

Table B.2 D values for actinide capture cross sections

Nuclide capture cross section	D-values for the production of response actinides									
	U-234	U-235	U-238	Pu-239	Pu-240	Pu-241	Pu-242	Am-241	Pu-238	Cm-244
U232	3.17E–07	1.70E–08	4.15E–11	1.49E–10	1.95E–10	5.57E–10	1.62E–09	4.02E–10	2.12E–09	3.12E–09
U233	2.50E–07	3.88E–07	3.60E–10	–5.03E–10	–1.85E–09	–3.23E–09	–1.24E–08	–1.50E–09	–1.15E–08	–3.48E–08
U234	8.10E–02	–1.94E–02	–4.65E–05	4.25E–05	2.11E–04	3.27E–04	1.32E–03	2.16E–04	1.72E–03	3.81E–03
U235	1.64E–01	–5.48E–02	1.90E–03	–8.21E–03	–1.20E–02	–1.62E–02	–6.12E–02	–9.51E–03	–1.49E–01	–1.74E–01
U236	1.01E–01	6.62E–03	8.87E–05	–6.13E–03	–2.58E–03	2.40E–04	4.04E–03	4.00E–05	–9.68E–02	7.32E–03
U238	4.03E–02	8.86E–01	1.89E–03	1.94E–02	4.10E–03	6.68E–02	1.83E–01	6.29E–02	2.73E–01	1.89E–01
Np237	2.97E–02	3.93E–03	4.92E–05	–6.04E–03	–3.30E–03	–1.16E–03	2.28E–03	–1.38E–03	–1.27E–01	5.36E–03
Np239	2.19E–04	–4.92E–04	–9.10E–06	–2.63E–04	5.96E–04	6.70E–04	8.30E–04	6.94E–04	1.98E–04	1.09E–03
Pu238	–1.53E–01	1.57E–03	5.66E–05	–5.90E–03	–3.13E–03	–1.01E–03	2.03E–03	–1.14E–03	–1.06E–01	4.22E–03
Pu239	2.89E–02	–3.05E–02	–5.81E–04	–2.30E–03	–5.60E–03	–1.33E–02	–3.42E–02	–1.26E–02	4.50E–03	–5.52E–02
Pu240	–2.08E–03	2.08E–02	4.00E–04	1.24E–03	–7.00E–03	–5.06E–02	–8.23E–02	–5.54E–02	1.19E–02	–8.45E–02
Pu241	–1.04E–02	–3.70E–02	–6.82E–04	–1.14E–03	1.93E–02	–7.50E–03	–1.87E–02	–7.90E–03	–1.64E–02	–2.08E–02
Pu242	1.35E–03	7.48E–03	1.36E–04	8.09E–05	8.98E–03	1.29E–03	–1.38E–01	1.63E–03	–1.25E–04	–1.15E–01
Pu244	2.29E–15	9.74E–15	1.76E–16	–2.95E–17	–2.09E–16	–2.20E–16	–1.05E–15	9.25E–16	–1.33E–15	–5.43E–15
Pu245	–1.09E–18	–4.62E–18	–8.37E–20	1.51E–20	1.00E–19	1.07E–19	5.97E–19	–4.41E–19	6.44E–19	2.60E–18
Am241	1.87E–03	3.56E–04	5.60E–06	–8.01E–05	2.70E–06	6.53E–05	–6.50E–04	4.19E–03	–1.99E–03	–1.69E–03
Am242m	9.90E–07	–5.27E–05	–9.88E–07	–2.21E–06	3.48E–05	–2.13E–06	–1.41E–05	–2.32E–07	–2.29E–05	–9.53E–04
Am242	–2.69E–05	–4.21E–07	–4.70E–09	–5.97E–07	7.31E–07	–3.93E–07	–5.26E–07	–3.99E–07	–3.55E–05	–1.32E–05
Am243	1.13E–03	6.08E–03	1.11E–04	3.96E–05	7.05E–03	1.06E–03	4.71E–04	1.36E–03	–1.46E–04	–1.19E–01
Cm242	–1.87E–03	7.82E–05	1.61E–06	–3.69E–05	–2.66E–05	–2.72E–05	–8.85E–06	–2.67E–05	–2.69E–03	–1.03E–04
Cm243	–9.59E–07	–7.20E–06	–1.32E–07	–8.23E–07	8.20E–06	6.40E–07	2.65E–07	4.82E–07	–3.48E–07	–8.85E–05
Cm244	1.43E–03	7.55E–03	1.37E–04	5.97E–05	–1.11E–02	-8.10E–04	–7.47E–04	–3.50E–04	–4.48E–04	–8.80E–02

Table B.3 D values for actinide capture cross sections

Nuclide fission cross section	D-values for the production of response actinides									
	U-234	U-235	U-238	Pu-239	Pu-240	Pu-241	Pu-242	Am-241	Pu-238	Cm-244
U232	−9.17E−08	−1.02E−09	4.89E−11	4.73E−10	7.80E−10	2.68E−09	5.99E−09	1.17E−09	7.68E−09	1.32E−08
U233	−1.74E−06	−5.24E−07	−2.88E−09	2.11E−09	1.20E−08	1.78E−08	7.45E−08	1.46E−08	9.33E−08	2.19E−07
U234	1.09E−03	−6.10E−04	−1.34E−06	9.76E−07	5.61E−06	8.96E−06	3.58E−05	6.64E−06	3.82E−05	1.05E−04
U235	−2.18E−01	−9.73E−01	−5.74E−03	3.18E−03	2.16E−02	3.61E−02	1.38E−01	3.31E−02	1.37E−01	4.13E−01
U236	−2.70E−03	−1.23E−03	−2.05E−05	3.64E−05	1.33E−04	4.40E−04	1.08E−03	1.33E−04	−3.21E−04	2.88E−03
U237	−3.54E−05	−1.77E−06	−2.11E−08	−5.00E−09	−1.32E−07	2.87E−06	2.01E−06	−3.49E−07	−1.73E−05	6.10E−06
U238	−2.37E−03	6.62E−02	1.96E−04	1.42E−03	8.00E−04	1.24E−02	1.84E−02	5.84E−03	2.33E−02	2.47E−02
Np236	4.20E−11	1.45E−09	2.71E−11	4.95E−11	3.95E−11	2.60E−10	4.13E−10	1.58E−10	5.36E−10	5.30E−10
Np237	−1.01E−03	−5.29E−05	−6.79E−07	−6.00E−07	−1.01E−06	7.73E−05	6.93E−05	−5.23E−06	−5.43E−04	1.96E−04
Np238	−2.81E−03	−3.15E−05	2.20E−07	1.94E−05	−1.38E−05	2.28E−04	5.04E−05	−3.71E−05	−8.70E−04	1.98E−04
Pu236	−5.10E−08	1.82E−08	3.78E−10	1.21E−09	1.37E−09	6.19E−09	1.20E−08	2.88E−09	1.54E−08	2.21E−08
Pu238	−8.18E−03	4.70E−05	2.54E−06	−4.61E−05	−8.20E−05	4.82E−04	1.25E−04	−1.04E−04	−5.51E−03	4.47E−04
Pu239	−4.91E−02	5.26E−02	1.07E−03	2.40E−03	−1.70E−03	7.00E−01	1.35E−02	−4.70E−03	3.82E−02	5.08E−02
Pu240	−1.11E−04	1.90E−04	3.62E−06	8.45E−06	−8.70E−05	2.57E−03	−1.75E−04	−1.45E−04	1.60E−06	−1.41E−04
Pu241	−2.37E−02	4.84E−02	9.08E−04	1.36E−03	−1.66E−02	7.10E−01	−3.20E−02	−1.87E−02	−1.26E−02	−4.71E−02
Pu242	2.80E−06	2.44E−05	4.47E−07	4.77E−07	−8.65E−06	2.28E−06	−6.93E−05	7.23E−07	1.99E−06	−6.15E−05
Pu243	3.76E−08	2.79E−07	5.10E−09	4.45E−09	−1.03E−07	1.80E−08	−8.51E−09	−2.00E−11	−5.57E−09	−5.57E−07
Am241	−9.84E−05	5.34E−06	1.12E−07	−1.62E−06	−2.26E−06	3.43E−06	2.33E−06	1.21E−05	−1.20E−04	6.01E−06
Am242m	−1.22E−05	5.46E−05	1.04E−06	2.91E−06	−3.49E−05	2.06E−05	2.52E−05	2.37E−06	2.70E−05	8.41E−04
Am242	−2.40E−04	1.61E−05	3.27E−07	−4.74E−06	−6.38E−06	7.27E−06	−2.02E−06	−3.73E−06	−3.14E−04	−2.77E−06
Am243	7.00E−06	4.67E−05	8.53E−07	6.65E−07	−2.67E−05	3.93E−06	−5.70E−07	4.98E−07	7.90E−07	−1.74E−04
Am244	2.59E−05	1.64E−04	2.98E−06	2.12E−06	−1.16E−04	1.28E−05	−1.21E−05	−4.63E−06	−8.39E−06	−4.70E−04
Cm242	−1.21E−04	6.60E−06	1.33E−07	−2.61E−06	−2.27E−06	2.71E−06	−5.30E−08	−1.69E−06	−1.73E−04	1.80E−07
Cm243	6.90E−07	2.72E−05	5.06E−07	−5.40E−06	−8.16E−06	5.22E−06	4.94E−06	3.36E−07	7.21E−06	2.31E−05
Cm244	4.56E−05	2.50E−04	4.89E−11	2.30E−06	−2.47E−04	1.89E−05	−1.83E−05	−3.05E−06	−1.12E−05	−1.97E−03

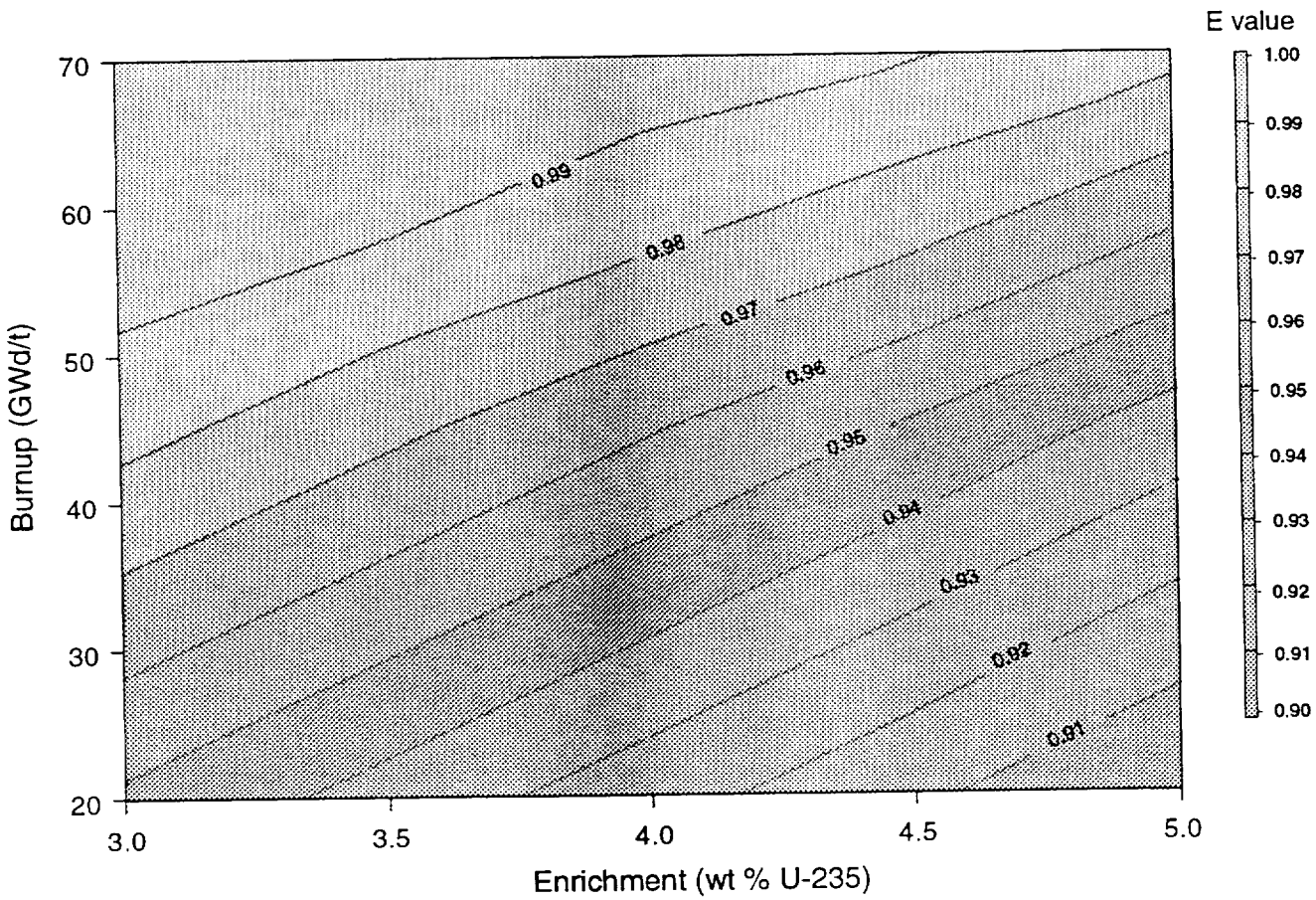

Figure B.1 *E*-parameter contour plot relative to 3.0-wt %, 70-GWd/t fuel for a *k* response

B.4 Recommendations

Sensitivity studies have been performed for the major uranium and plutonium isotopes, other burnup-credit actinides, and dominant actinides important to decay heat and neutron source terms. These studies demonstrate that the relative importance of the underlying processes and nuclear data fundamental to the calculation of the actinide number densities in the low-enrichment, high-burnup H. B. Robinson fuel are very similar to the more typical high-enrichment, high-burnup fuels. Similar conclusions are anticipated for the high-burnup Limerick-1 BWR fuel.

The applicability of the high-burnup spent fuel samples has been reviewed in terms of (1) the sensitivity of the calculated isotopic concentrations to the nuclear data parameters that are key in the production of the dominant actinides, and (2) the sensitivity of the data parameters in terms of their importance to the prediction of *k*. An assessment of the effects of the assembly lattice geometry on the accuracy of the cross sections was beyond the scope of this study. It is likely that the assembly designs for higher enriched fuels will be more complex and heterogeneous than the lower enrichment 15 × 15 or the 9 × 9 designs of the H. B. Robinson and Limerick assemblies being considered. Consequently, the application of measured isotopic inventory data from these assemblies to establishing the calculational bias for high-burnup fuel should also consider the differences in the

current and older assembly designs. As these designs become increasingly complex, the ability of the neutronics codes to generate accurate cross sections needs to be verified to ensure the reliability of the validation studies. A significant difference in the accuracy of the cross sections for different assembly designs would clearly impact the applicability of validation benchmarks. The ability of codes to accurately predict cross sections as assembly designs become increasingly complex is a code-specific issue and must be evaluated in combination with sensitivity studies, such as those described in this report.

Another area of concern with using the high-burnup H. B. Robinson fuel for validation is that the substantial depletion of fissile material will result in the fuel acting as a net absorber of neutrons (e.g., the number of fissions occurring in the fuel near discharge is less than the number of absorptions). This situation differs from normal operation where the number of fissions and absorptions is nearly balanced. This neutron production/absorption imbalance can affect the neutron spectrum in the fuel and, unless the imbalance is accurately considered in the computational analysis, may also have an impact on the accuracy of the cross sections used in the depletion analysis. This effect is currently being investigated.

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10. SUPPLEMENTARY NOTES

D.D. Ebert, NRC Project Manager

11. ABSTRACT *(200 words or less)*

This report has been prepared to review the technical issues important to the prediction of isotopic compositions and source terms for high-burnup light-water reactor (LWR) fuel as utilized in licensing of spent fuel transport and storage systems. The current trend towards higher spent fuel burnups than seen in the past has led to a situation where high-burnup assemblies from operating LWRs now extend beyond the area where available experimental data can be used to validate the computational methods employed to calculate spent fuel inventories and source terms. This report reviews currently available validation data and potential new sources of experimental data available in the near term. A review of the background issues important to isotopic predictions and some of the perceived technical challenges that high-burnup fuel presents to the current computational methods are discussed, and areas that need to be further investigated are presented. The methods and data development that may be required to address the possible shortcomings of physics and depletion methods in the high-burnup and high-enrichment regime are also discussed. Finally, a sensitivity analysis methodology being explored to help establish the range of applicability for experimental data in code validation is also discussed and demonstrated.

12. KEY WORDS/DESCRIPTORS *(List words or phrases that will assist researchers in locating the report.)*

high-burnup LWR fuel, spent fuel transport and storage systems, calculation of spent fuel inventories and source terms, burnup-credit

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