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Investigation of Nuclide Importance to Functional Requirements Related to Transport and Long-Term Storage of LWR Spent Fuel

> B. L. Broadhead M. D. DeHart J. C. Ryman J. S. Tang C. v. Parks

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Computational Physics and Engineering Division

INVESTIGATION OF NUCLIDE IMPORTANCE TO FUNCTIONAL REQUIREMENTS RELATED TO TRANSPORT AND LONG-TERM STORAGE OF LWR SPENT FUEL

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TABLE OF CONTENTS

<u>Page</u>

LIST OF FIGURES	iv . v vii
1. INTRODUCTION	1
2. APPROACH	3
2.1 AREAS OF INTEREST 2.2 ANALYSIS METHOD	3
3. RESULTS AND DISCUSSION	5
3.1 CRITICALITY SAFETY RANKINGS	5
3.2 SHIELDING RANKINGS	9
3.3 CURIE RANKINGS	14
3.4 DECAY HEAT RANKINGS	14
3.5 RADIOLOGICAL TOXICITY RANKINGS	17
3.6 SUMMARY	21
4. CONCLUSIONS	25
5. REFERENCES	26
APPENDIX A. CRITICALITY SAFETY PLOTS	29
APPENDIX B. DOSE RATE FRACTION PLOTS	49
APPENDIX C. CURIE LEVEL FRACTION PLOTS	81
APPENDIX D. DECAY HEAT FRACTION PLOTS	89
APPENDIX E. RADIOLOGICAL TOXICITY FRACTION PLOTS	97

LIST OF FIGURES

Figure		<u>P</u>	'age
1	Effect of long-term cooling on k_{∞} .		. 8

LIST OF TABLES

<u>Table</u>	<u>Pa</u>	ige
1	Burnup and enrichment combinations used in ranking studies and their	
	relationships to industry average burnups	4
2	Rankings of actinides with greater than 1% of total absorptions	
	at 5 and 30,000 years	6
3	Rankings of fission products with greater than 0.1% of total absorptions at	
	5 and 30,000 years	7
4	Actinide sensitivities and comparison of k _{eff} -vs-absorption rankings	10
5	Fission-product sensitivities and comparison of k _{eff} and absorption rankings	11
6	Shielding rankings of actinides with greater than 1% of total dose at	
	5 and 10,000 years	12
7	Shielding rankings of fission products and light elements with greater than 1%	
	of total dose at a 5-year cooling time	12
8	Curie rankings of actinides, fission products, and light elements with	
	greater than 0.1% of total curies at 5 and 10,000 years	15
9	Decay heat rankings of actinides, fission products, and light elements	
	with greater than 1% of total decay heat at 5 and 10,000 years	16
10	Ingestion toxicity rankings of light elements, actinides, and fission	
	products with greater than 0.1% of total potential committed effective	
	dose equivalent at 5 and 10,000 years	18
11	Inhalation toxicity rankings of light elements, actinides, and fission products	
	with greater than 0.1% of total potential committed effective dose equivalent	
	at 5 and 10,000 years	19
12	Summary rankings by analysis area (actinides)	22
13	Summary rankings by analysis areas (fission products and light elements)	23
14	Accuracy and applicability of current analytical methods	24

ABSTRACT

The radionuclide characteristics of light-water-reactor (LWR) spent fuel play key roles in the design and licensing activities for radioactive waste transportation systems, interim storage facilities, and the final repository site. Several areas of analysis require detailed information concerning the time-dependent behavior of radioactive nuclides including (1) neutron/gamma-ray sources for shielding studies, (2) fissile/absorber concentrations for criticality safety determinations, (3) residual decay heat predictions for thermal considerations, and (4) curie and/or radiological toxicity levels for materials assumed to be released into the ground/environment after long periods of time. The crucial nature of the radionuclide predictions over both short and long periods of time has resulted in an increased emphasis on thorough validation for radionuclide generation/depletion codes.

Current radionuclide generation/depletion codes have the capability to follow the evolution of some 1600 isotopes during both irradiation and decay time periods. Of these, typically only 10 to 20 nuclides dominate contributions to each analysis area. Thus a quantitative ranking of nuclides over various time periods is desired for each of the analysis areas of shielding, criticality, heat transfer, and environmental dose (radiological toxicity). These rankings should allow for validation and data improvement efforts to be focused only on the most important nuclides.

This study investigates the relative importances of the various actinide, fission-product, and lightelement isotopes associated with LWR spent fuel with respect to five analysis areas: criticality safety (absorption fractions), shielding (dose rate fractions), curies (fractional curies levels), decay heat (fraction of total watts), and radiological toxicity (fraction of potential committed effective dose equivalent). These rankings are presented for up to six different burnup/enrichment scenarios and at decay times from 2 to 100,000 years. Ranking plots for each of these analysis areas are given in an Appendix for completeness, as well as summary tables in the main body of the report. Summary rankings are presented in terms of high (greater than 10% contribution to the total), medium (between 1% and 10% contribution), and low (less than 1% contribution) for both short- and long-term cooling. When compared with the expected measurement accuracies, these rankings show that most of the important isotopes can be characterized sufficiently for the purpose of radionuclide generation/depletion code validation in each of the analysis areas. Because the main focus of this work is on the relative importances of isotopes associated with LWR spent fuel, some conclusions may not be applicable to similar areas such as high-level waste (HLW) and nonfuel-bearing components (NFBC).

1. INTRODUCTION

The Department of Energy's (DOE) Office of Civilian Radioactive Waste Management Program (OCRWM) was established, in accordance with the Nuclear Waste Policy Act of 1982, to serve as the lead office for (1) siting, constructing, and operating nuclear waste repositories, (2) transporting commercial spent nuclear fuel and high-level wastes, (3) developing a proposal to construct a monitored retrievable storage facility, and (4) aiding utilities in solving spent fuel storage problems. Information about the radionuclide characteristics of various spent fuel and high-level wastes during different time frames is necessary for each of these work areas.

To meet this need, the DOE-OCRWM has developed the Characteristic Data Base (CDB) to provide program participants with a consistent and qualified source of information about the quantities, dimensions, and compositions of spent fuel, high-level waste, and radioactive commercial nuclear reactor components. The CDB (currently revision 1) is comprised of a four-volume hard copy¹ report and six data bases that can be accessed via personal computer (PC). The CDB has undergone an extensive technical peer review, and has been "certified" by OCRWM for use in "quality affecting" work within the OCRWM program. One of the six data bases is the Radiological Data Base (RADDB) and provides users with compositions, curies, decay heat, photon source, and neutron source, based on the type of reactor [pressurized-water reactor (PWR) or boiling-water reactor (BWR)], initial enrichment, burnup, and cooling time of the fuel. The RADDB was created from multiple applications of the ORIGEN2 computer code² using recently developed cross-section libraries.³ One important outcome of the peer review was the recognition that future revisions of the CDB would require that the analysis methods used to generate the data comply with OCRWM quality assurance (QA) requirements for software.

OCRWM formed the CDB Users Group in April 1993 as a discussion forum for CDB Users nationwide and to provide technical input to help guide future CDB development. Many of the charter members of the CDB Users Group were assigned to subcommittees aligned technically with each of the six PC data bases. The radiological subcommittee was assigned to help guide further development of the RADDB and the verification and validation efforts related to the analysis methodology. The crucial nature of the radionuclide predictions over both short and long periods of time has resulted in a heightened interest in the accuracy of the radionuclide generation/depletion codes used in the design and safety evaluation of transportation system, interim storage, and high-level-waste repository facilities. However, the validation of analysis methods used to predict spent fuel isotopics is hampered by the paucity of measured data that are available relative to the more than 1600 nuclides typically tracked in a standard point-depletion calculation using an ORIGEN-type code. In addition, the available measured data cover only a limited range of possible spent fuel characteristics (burnup, initial enrichment, cooling time, and assembly type). Thus to help direct future validation efforts and assist OCRWM in identifying the nuclides of importance to the functional requirements, the radiological subcommittee of the CDB Users Group recommended that work be done to provide a quantitative measure of the nuclide ranking for important analysis areas.

The radionuclide characteristics play key roles in the transportation system, interim storage facility, and repository design and licensing processes for several analysis areas including (1) neutron/gamma-ray sources for shielding studies, (2) fissile/absorber concentrations for criticality safety determinations, (3) residual decay heat predictions for thermal considerations, and (4) curie and/or radiological toxicity levels for materials assumed to be released into the ground/environment after long periods of time. The transportation of radioactive waste from reactor sites to the interim retrievable storage facility and ultimately to the final repository typically involves 5 to 50 years out-of-reactor time frames. Once the waste is placed in a final repository, time frames of interest include 10 to 1,000 years after repository closure for heating considerations and 1,000 to 10,000 years after closure for curie levels and cumulative release limits to the environment. For

criticality concerns, the time frames of interest are somewhat open since the principal fissile isotopes have halflives on the order of 10^8 years.

Each radioactive isotope has a different half-life, which can shift the relative importance of each isotope over the long time periods of concern to the repository design. Thus a nuclide ranking study needs to consider the effect that radioactive decay has on the ranking for each analysis area—shielding, criticality, heat transfer, and environmental dose (radiological toxicity). These rankings should allow for detailed validation and data improvement efforts to be concentrated on the limited number of nuclides that may have a significant impact on the functional requirements of the OCRWM objectives.

The development of this nuclide importance ranking report is the culmination of the efforts by ORNL to provide OCRWM and the CDB Users Group radiological subcommittee with a quantitative means to assess (validate) the ORIGEN model (embodied in ORIGEN2 and ORIGEN-S,⁴ the SCALE system version of ORIGEN), as well as to judge the relative importance of obtaining additional isotopic data.

2. APPROACH

2.1 AREAS OF INTEREST

The rankings generated in this work cover the basic analysis areas of shielding, criticality safety, decay heat, curie levels, and radiological toxicity. In shielding analyses, the photon and neutron energy spectra from the spent fuel composition, coupled with the dimensions, compositions, and cross sections of the shielding material, contribute to the relative importance of each nuclide. Because of the importance of spent fuel transport and storage casks in the OCRWM program, dose rates at the cask surface for three cask types and two fuel burnup/enrichments were chosen as the basis for ranking nuclides on their importance to the shielding analysis area. For criticality safety, absorption fractions were generated for the major actinide and fissionproduct absorbers for six burnup/enrichment combinations. Even though the absorption fractions represent only an indirect measure of importance for criticality, absorption reactions contribute directly to keff; an absorption capture decreases keff, but an absorption fission increases keff. The fractional contributions to the total decay heat were obtained for two burnup/enrichment states. Similarly, fractional contributions to the total curie levels and radiological toxicity values as a function of decay time were obtained for the same two burnup/enrichment scenarios. These fractional contributions allow the ranking of radionuclide importances over time with respect to criticality safety, shielding, decay heat generation, curie levels, and toxicity for the LWR spent fuel burnup/enrichment cases considered. Not explicitly treated in this work are other types of HLW (i.e., ²³³U-fueled systems) and the associated irradiated components in a reactor (NFBC, pressure vessel, biological shield, etc.).

2.2 ANALYSIS METHOD

The basis for the ranking studies in each analysis area is a common set of radioisotope concentrations corresponding to Westinghouse 17×17 pressurized-water-reactor (PWR) fuel elements with enrichment and burnup characteristics, as given in Table 1. The spent fuel inventories were generated via the SAS2H/ORIGEN-S computer code⁵ assuming a reactor specific power of 40 MW/t and a power history with 80% uptime, 20% downtime in each cycle. Typical (0.46 wt%) loadings of Co impurity were assumed in the Inconel grid spacer material. The base cross-section library used in these burnup/depletion calculations was the recently developed SCALE 44-group library,⁶ based primarily on ENDF/B-V data, with ¹⁶O, ¹⁵⁴Eu, and ¹⁵⁵Eu data obtained from ENDF/B-VI. Major changes were made in the ENDF/B-VI data for ¹⁵⁴Eu and ¹⁵⁵Eu; hence, updated cross sections were used in this study. The end-of-irradiation concentrations were computed at decay times of 2, 5, 10, 20, 100, 200, 1,000, 3,000, 10,000, 30,000, and 100,000 years. Some analysis areas did not utilize all enrichment/burnup and cooling-time combinations. The shielding, decay heat, and curie rankings were desired at a low and a high burnup and used the 20-GWd/t and 50-GWd/t burnups with cooling time up to 10000 years. The absorption ranking studies investigated all six burnup/enrichment combinations and each of the eleven cooling times. Criticality rankings were desired at each burnup/enrichment combination to show the changes in importance for underburned vs overburned fuel. Overburned and underburned fuel correspond to the irradiation of a fuel assembly substantially longer or shorter than the industry average for a given enrichment (see Table 1). The toxicity rankings analyzed the 20-GWd/t and 50-GWd/t burnups and cooling times up to the full 100,000 years.

Enrichment, wt % ²³⁵ U	Burnup (GWd/t)	Estimated industry-average burnup (GWd/T)	Comments
3.0	20	27	Underburned
5.0	20	27	Underburned
3.0	35	27	Overburned
4.0	30	43	Underburned
4.0	40	43	Average
4.0	45	43	Overburned
4.5	50	53	Average

Table 1. Burnup and enrichment combinations used in ranking studies and their relationships to industry average burnups

Computed absorption fractions, curie levels, and decay heat values in watts were obtained directly from the ORIGEN-S outputs. The absorption fractions were plotted directly; the curie and decay heat results were converted separately to fractional values and plotted. The rankings were tabulated separately for actinide and fission product materials to show their relative importances.

For the shielding rankings, the neutron and gamma-ray sources were taken from the ORIGEN-S outputs and input into the one-dimensional (1-D) discrete-ordinates module SAS1/XSDRNPM-S⁷ to obtain the radial dose rate at the cask surface. Three cask models were analyzed to determine the variation of rankings with cask type. The first model consisted of a 27-cm carbon steel/13-cm resin shield, the second configuration contained a 12.7-cm lead/13-cm resin shield, and the final cask consisted of a 50-cm concrete shield. Calculations were performed using the SCALE coupled 27-neutron/18-gamma-group library with all radionuclides present to obtain total dose rate information, followed by repetitive calculations with individual isotopes to obtain partial dose rate information. Contributions to the neutron dose rates and primary/secondary gamma dose rates were separately tabulated to show their relative importances.

The radiological toxicity rankings were derived from the ORIGEN-S outputs by multiplying the activity of each nuclide by the committed effective dose equivalent per unit intake, taken from Federal Guidance Report No. 11.⁸ This report gives the potential committed effective dose equivalent (i.e., the effective dose equivalent that would be received over a 50-year period following intake, assuming all the nuclide present is ingested or inhaled). Thus the radiological toxicity rankings *do not* account for the mitigating effects of release fraction and dispersion (for airborne releases) or solubility, retardation, and exposure pathways (for underground waste package releases). The fraction of the total committed effective dose equivalent from each nuclide was derived both for ingestion and inhalation, and analyzed separately for the actinides, fission products, and light elements.

The rankings for each of the respective analysis areas are generated and presented as the *fractional* contribution to the *total* response. Thus for the criticality rankings, the fractional absorption for each nuclide is generated based on the total absorptions in the system. Similarly for decay heat, curies, and toxicity, the fractional contributions are based on the total watts, curies, and committed dose, respectively. For the shielding analyses, the fractional contributions are also based on the total dose rates. However, since actinides contribute to both neutron and gamma-ray doses, the fractional contributions from neutrons and gamma rays are listed separately for each actinide.

3. RESULTS AND DISCUSSION

3.1 CRITICALITY SAFETY RANKINGS

The nuclide importance rankings for criticality safety at decay times of 5 and 30,000 years are given in Tables 2 and 3 in this section. A complete set of fractional absorption plots for all decay times is provided in Appendix A. These rankings are based on the fractional absorption for each nuclide at each of the various decay times. The rankings are tabulated at 5 years to correspond to the minimum cooling time at which OCRWM will accept spent fuel. The 30,000-year ranking period was chosen because (1) the k_{∞} value of spent fuel has a local peak in that general time frame, as seen in Fig. 1 and (2) it represents significant decay time in terms of repository design issues.

The key features in these rankings are dominance of primary actinides ²³⁵U, ²³⁸U, and ²³⁹Pu, along with the large number of fission products which should be included for accurate results. Specific features seen are:

- 1. For low burnup and short cooling times, actinides are responsible for about 90% of all absorptions; after 100,000 years actinides still represent 87% of all absorptions. For high burnup, actinides absorb 85% of all neutrons after 5 years, but less than 79% of absorptions occur in actinides after 100,000 years.
- 2. ²³⁵U is the most important actinide absorber for underburned fuel only. It falls well below ²³⁸U and ²³⁹Pu for more highly burned fuel at 5-year decay times. The 5-year decay trends should follow closely the trends seen during irradiation where the concentration of ²³⁵U is constantly decreasing due to burnup, while the amount of ²³⁹Pu is generally increasing due to capture in ²³⁸U. At 30,000-year decay times, ²³⁵U is again the most important actinide absorber because ²³⁹Pu decays to ²³⁵U with a 24,000-year half-life.
- 3. ²³⁹Pu is the most important absorber for moderately burned fuel, but its fractional absorption decreases as fuel is overburned because the ²³⁹Pu present begins to serve as fuel when its concentrations become significant. However, the decrease is small and it remains the largest absorber until fuel is highly overburned.
- 4. ²³⁸U is always one of the top two absorbers, and its fractional contribution is insensitive to burnup because large initial inventories and relatively small cross sections allow a near-constant abundance during irradiation.
- 5. ¹⁴⁹Sm is the highest ranking fission-product absorber for low burnup; however, it is insensitive to burnup and becomes less important as burnup increases because its cross section is so large that depletion becomes significant for high burnups.
- 6. ¹⁴³Nd increases in importance with increasing exposure and becomes more important than ¹⁴⁹Sm for overburned fuel, since its much lower cross sections relative to ¹⁴⁹Sm allow it to continue to build up even with high burnups.
- 7. ¹⁵⁵Gd is sensitive to both burnup and cooling time. For underburned fuel it ranks ninth after 5 years of cooling and fifth after 30,000 years. For moderate- and high-burnup cases it increases in relative importance as an absorber. For the highly burned 4.5 wt %, 50-GWd/t case, ¹⁵⁵Gd ranks sixth after 5 years, but it is the second most important absorber after 30,000 years. Note that the use of

	Burnup/Enrichment							
Nuclide	20 GWd/t 3.0 wt %	35 GWd/t 3.0 wt %	30 GWd/t 4.0 wt %	40 GWd/t 4.0 wt %	45 GWd/t 4.0 wt %	50 GWd/t 4.5 wt %		
5-year rankings								
U-235	$1(29)^{a}$	3(15)	1(28)	3(20)	3(16)	3(17)		
U-238	2(26)	2(26)	2(24)	2(25)	2(25)	2(24)		
Pu-239	3(23)	1(27)	3(24)	1(25)	1(26)	1(26)		
Pu-240	4(5)	4(7)	4(6)	4(7)	4(7)	4(7)		
Pu-241	5(3)	5(6)	5(4)	5(6)	5(6)	5(6)		
Am-241	6(1)	6(1)	6(1)	6(1)	6(1)	6(1)		
<u>30,000-year rankings</u>								
U-235	1(44)	1(30)	1(43)	1(35)	1(31)	1(32)		
U-238	2(29)	2(30)	2(27)	2(27)	2(28)	2(27)		
Pu-239	3(12)	3(16)	3(13)	3(15)	3(16)	3(15)		
Np-237	4(2)	4(3)	4(2)	4(3)	4(3)	4(3)		
U-236	5(1)	5(1)	5(1)	5(1)	5(1)	5(1)		

Table 2. Rankings of actinides with greater than 1% of total absorptions at 5 and 30,000 years

^aIsotopes percentage contribution to the total number of absorptions.

Durren /Earlishmant								
	Burnup/Enrichment							
	20 GWd/t	35 GWd/t	30 GWd/t	40 GWd/t	45 GWd/t	50 GWd/t		
Nuclide	3.0 wt %	30 wt %	4 0 wt %	4.0 wt %	40 wt %	4 5 wt %		
Ituende	5.0 Wt /0	5.0 Wt /0	4.0 Wt /0	4.0 Wt /0	4.0 Wt /0	4.5 Wt /0		
5-year ran	kings/30,000 yea	rs						
Sm-149	$1/1^{a}(1/2)^{b}$	1/1(2/2)	1/1(1/2)	1/1(1/2)	2/2(2/2)	2/3(1/2)		
Nd-143	2/2(1/1)	2/2(1/2)	2/2(1/2)	2/2(1/2)	1/1(2/2)	1/1(2/2)		
Rh-103	3/3(0.8/1)	3/4(1/2)	3/3(1/1)	3/4(1/2)	3/4(1/2)	3/4(1/2)		
Sm-151	4/-(0.7/-)	4/-(0.9/-)	4/-(0.8/-)	4/-(0.9/-)	4/-(1/-)	4/-(1/-)		
Xe-131	5/6(0.5/0.5)	5/6(0.7/0.8)	5/6(0.6/0.6)	5/6(0.7/0.8)	6/6(0.7/0.8)	7/7(0.8/0.9)		
Cs-133	6/7(0.4/0.4)	7/7(0.6/0.7)	6/7(0.5/0.6)	6/7(0.7/0.7)	7/7(0.7/0.8)	5/6(0.8/0.9)		
Tc-99	7/9(0.3/0.3)	8/9(0.5/0.5)	7/8(0.4/0.4)	8/8(0.5/0.5)	8/8(0.6/0.6)	8/8(0.6/0.6)		
Sm-152	8/8(0.3/0.3)	9/8(0.5/0.5)	8/9(0.4/0.4)	9/9(0.5/0.5)	9/9(0.5/0.6)	9/9(0.5/0.6)		
Gd-155	9/5(0.3/0.6)	6/3(0.7/2)	9/4(0.4/0.9)	7/3(0.6/2)	5/3(0.7/2)	6/2(0.8/2)		
Nd-145	10/11(0.2/0.2)	11/11(0.3/0.4)	10/11(0.3/0.3)	11/11(0.3/0.4)	11/11(0.4/0.4)	11/11(0.4/0.5)		
Sm-147	11/10(0.2/0.2)	13/12(0.2/0.3))	12/10(0.2/0.3)	12/12(0.3/0.4)	13/12(0.3/0.4)	13/12(0.3/0.4)		
Eu-153	12/12(0.2/0.2)	10/10(0.4/0.4)	11/12(0.2/0.3)	10/10(0.4/0.4)	10/10(0.4/0.5)	10/10(0.5/0.5)		
Mo-95	13/14(0.1/0.2)	15/15(0.2/0.3)	13/14(0.2/0.2)	14/14(0.2/0.3)	14/14(0.3/0.3)	14/14(0.3/0.3)		
Sm-150	14/13(0.1/0.2)	12/13(0.3/0.3)	14/13(0.2/0.2)	13/13(0.3/0.3)	12/13(0.3/0.4)	12/13(0.3/0.4)		
Ag-109	15/15(0.1/0.1)	14/14(0.2/0.3)	15/15(0.2/0.2)	15/15(0.2/0.2)	15/15(0.3/0.3)	15/15(0.3/0.3)		
Ru-101	-	16/16(0.2/0.2)	16/16(0.1/0.1)	16/16(0.2/0.2)	16/16(0.2/0.2)	16/16(0.2/0.2)		
Pd-105	-	17/19(0.1/0.1)	-	17/19(0.1/0.1)	17/19(0.1/0.1)	17/18(0.1/0.2)		
Pr-141	-	-/18(-/0.1)	-	-/18(-/0.1)	18/18(0.1/0.1)	18/19(0.1/0.2)		
Gd-157	-	-/17(-/0.1)	-	-/17(-/0.1)	19/17(0.1/0.2)	19/17(0.1/0.2)		
Eu-151	-/4(-/0.7)	-/5(-/1)	-/5(-/0.8)	-/5(-/1)	-/5(-/1)	-/5(-/1)		

Table 3. Rankings of fission products with greater than 0.1% of
total absorptions at 5 and 30,000 years

^{*a*}Rankings correspond to 5/30,000 years. ^{*b*}Percentage contributions corresponding to 5/30,000 years.



K-infinity vs Decay Time

Fig. 1. Effect of long-term cooling on k_{∞} .

ENDF/B-V or earlier data for ¹⁵⁵Eu causes the importances for ¹⁵⁵Gd to be roughly double the values reported here. Major cross-section changes were made in the ENDF/B-VI ¹⁵⁵Eu data.⁹

- 8. ¹⁵¹Sm is important for early decay periods but decays (90-year half-life) to ¹⁵¹Eu, which then becomes important at later decay periods.
- 9. The rankings for extended decay times show interesting characteristics for various decay chains, such as ²⁴¹Pu→²⁴¹Am→²³⁷Np, where each nuclide is important over different periods of decay.

All criticality safety rankings presented thus far are based on the assumption that the relative rankings for fractional absorptions are the same as the relative rankings for the effective neutron multiplication factor, k_{eff} . To confirm this assumption, k_{eff} calculations were performed for an infinite array of fuel pins with the nuclide concentrations corresponding to the 3.0 wt %, 30-GWd/t case shown above at a cooling time of 5 years. The base case k_{eff} was computed with 11 actinides and 19 fission products included. Sensitivity calculations were then performed by changing concentrations for each nuclide individually and recomputing k_{eff} . The results shown in Tables 4 and 5 give the base k_{eff} as well as the percentage change in k_{eff} for actinide and fission-product perturbations, respectively. The actual rankings for the multiplication factor are then compared with those predicted from the absorption fractions. Identical rankings are seen, except for a few cases where two nuclides with very similar sensitivities have reversed importances.

3.2 SHIELDING RANKINGS

The rankings for the shielding portion of this work are given in Tables 6 and 7 for two burnups (20 GWd/t and 50 GWd/t) and their corresponding enrichments (3.0 wt % and 4.5 wt %) for three different cask types. These rankings are presented for decay times of 5 and 10,000 years after irradiation. Complete plots and rankings for the fractional contribution to the total dose rates for the various actinides, fission products, and light elements are shown in Appendix B for ten decay times ranging from 2 to 10,000 years.

Nuclide ranking studies for determining the important contributors to a shielding analysis are highly sensitive to the shield thickness and shield material. For thin shields, the gamma-ray energy is much less important than for thick shields, where typically particles with energies at or above 1 MeV dominate the dose contribution. The composition of the shielding material(s) affects the relative contributions of neutrons, primary gamma rays, and secondary gamma rays since hydrogenous materials are much more effective for attenuating neutrons, while high-Z materials are much more effective shields for gamma rays. The casks considered in this work represent three examples of thick shields: iron/resin, lead/resin, and concrete materials. The curie rankings for fission products and light elements given in a later section would be more appropriate dose rankings for a thin shield.

The key features seen in the rankings given in Tables 6 and 7 are the dominance for a 5-year decay time of the actinide ²⁴⁴Cm and the high-activity fission products and light elements with high- energy gamma rays. For a 10,000-year decay time, the fission products and light elements are unimportant, while the actinides ²⁴⁰Pu, ²⁴²Pu, ²³⁹Pu, and ²¹⁴Bi dominate the dose rate contributions. The specific features seen from an analysis of the plots shown in the Appendix include:

Case	Nuclide composition, % change	k _{eff}	Sensitivity ^b coefficient	Δk rank	Absorption fraction rank
Base case		0.982666			
Pu-239	1	0.984308	0.1642	1	1
U-238	1	0.981040	-0.1626	2	2
U-235	1	0.983751	0.1085	3	3
Pu-240	2	0.981465	-6.005E-2	4	4
Pu-241	2	0.983686	5.100E-2	5	5
Am-241	10	0.981345	-1.321E-2	6	6
Np-237	25	0.981191	-5.900E-3	7	8
U-236	25	0.981362	-5.216E-3	8	7
Pu-242	25	0.981525	-4.564E-3	9	9
Am-243	100	0.980841	-1.825E-3	10	10
U-234	100	0.981619	-1.047E-3	11	11

Table 4. Actinide sensitivities^{*a*} and comparison of k_{eff} and absorption rankings

 $^{a}\text{Corresponds}$ to 17 \times 17 PWR, 3% enrichment, 35-GWd/MTU with 5-year cooling time.

^bSensitivity coefficient, $S_N = \frac{\Delta k/k}{\Delta N/N}$, where k is the multiplication factor and N is the

nuclide concentration.

	Nuclide				
	composition,		Sensitivity ^b	Δk	Absorption
Case	% change	k _{eff}	coefficient	rank	fraction rank
Base case		0.982666			
Sm-149	10	0.980837	-1.829E-2	1	1
Nd-143	10	0.980970	-1.696E-2	2	2
Rh-103	10	0.981284	-1.382E-2	3	3
Sm-151	10	0.981621	-1.045E-2	4	4
Gd-155	25	0.980729	-7.748E-3	5	6
Xe-131	25	0.980950	-6.864E-3	6	5
Cs-133	25	0.981056	-6.440E-3	7	7
Tc-99	25	0.981391	-5.100E-3	8	8
Sm-152	25	0.981517	-4.596E-3	9	9
Eu-153	25	0.981617	-4.196E-3	10	10
Nd-145	25	0.981758	-3.632E-3	11	11
Sm-150	25	0.981941	-2.900E-3	12	12
Sm-147	25	0.982008	-2.632E-3	13	13
Ag-109	25	0.982035	-2.524E-3	14	14
Mo-95	25	0.982081	-2.340E-3	15	15
Ru-101	100	0.981042	-1.624E-3	16	16
Gd-157	100	0.981107	-1.559E-3	17	18
Pd-105	100	0.981506	-1.160E-3	18	17
Pr-141	100	0.981560	-1.106E-3	19	19

Table 5. Fission-product sensitivities^a and comparison of k_{eff} and absorption rankings

^{*a*}Corresponds to 17 × 17 PWR, 3% enrichment, 35 GWd/MTU with 5-year cooling time. ^{*b*}Sensitivity coefficient, $S_N = \frac{\Delta k/k}{\Delta N/N}$, where k is the multiplication factor and N is the

nuclide concentration.

$\frac{20 \text{ GWd/t}}{3.0 \text{ wt \%}} \frac{50 \text{ GWd/t}}{4.5 \text{ wt \%}} \frac{20 \text{ GWd/t}}{3.0 \text{ wt \%}} \frac{50 \text{ GWd/t}}{4.5 \text{ wt \%}} \frac{20 \text{ GWd/t}}{3.0 \text{ wt \%}} \frac{50 \text{ GWd/t}}{4.5 \text{ wt \%}} \frac{50 \text{ GWd/t}}{3.0 \text{ wt \%}} \frac{4.5 \text{ wt \%}}{4.5 \text{ wt \%}}$		Iron cask ^a		Lead	Lead cask ^a		Concrete cask ^a	
5-year rankings Cm-244 1/-/- ^b (1/-/-) ^c 1/1/-(12/7/-) 1/1/-(19/1/-) -/-/-(-/-) 1/1/-(2/2/-) 10,000-year rankings	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 %	
Cm-244 1/-/-b(1/-/-)c 1/1/-(12/7/-) 1/-/-(2/-/-) 1/1/-(19/1/-) -/-/-(-/-/-) 1/1/-(2/2/-) 10,000-year rankings 10	5-year rankings							
10,000-year rankings	Cm-244	$1/-/-^{b}(1/-/-)^{c}$	1/1/-(12/7/-)	1/-/-(2/-/-)	1/1/-(19/1/-)	-/-/-(-/-/-)	1/1/-(2/2/-)	
	10,000-year rankings							
Pu-240 1/1/1(34/3/17) 2/2/2(25/1/14) 1/2/2(50/1/2) 2/-/3(38/-/2) 1/1/1(23/6/15) 2/2/2(18/3/13)	Pu-240	1/1/1(34/3/17)	2/2/2(25/1/14)	1/2/2(50/1/2)	2/-/3(38/-/2)	1/1/1(23/6/15)	2/2/2(18/3/13)	
Pu-242 2/2/2(16/1/8) 1/1/1(30/1/16) 2/-/3(24/-/1) 1/-/2(44/-/2) 2/3/3(11/3/7) 1/1/1(21/4/16)	Pu-242	2/2/2(16/1/8)	1/1/1(30/1/16)	2/-/3(24/-/1)	1/-/2(44/-/2)	2/3/3(11/3/7)	1/1/1(21/4/16)	
Pu-239 3/-/3(10/-/5) 3/-/4(5/-/2) 3/-/-(16/-/-) 3/-/-(8/-/-) 3/4/4(7/2/4) 3/-/4(3/-/2)	Pu-239	3/-/3(10/-/5)	3/-/4(5/-/2)	3/-/-(16/-/-)	3/-/-(8/-/-)	3/4/4(7/2/4)	3/-/4(3/-/2)	
Bi-214 -/-/4(-/-/4) -/-/3(-/-/4) -/1/1(-/2/3) -/1/1(-/1/3) -/2/2(-/5/11) -/-/3(-/-/11)	Bi-214	-/-/4(-/-/4)	-/-/3(-/-/4)	-/1/1(-/2/3)	-/1/1(-/1/3)	-/2/2(-/5/11)	-/-/3(-/-/11)	

Table 6. Shielding rankings of actinides with greater than 1%of total dose at 5 and 10,000 years

"Gamma shields consist of 27-cm steel, 12.7-cm lead, and 50-cm concrete for the iron, lead, and concrete casks, respectively.

^bRankings with respect to neutron/primary gamma/secondary gamma dose rates. The /-/ symbol indicates all contributions less than 1%.

Percentage contribution from each isotope to the total dose, listing neutron/primary gamma/secondary gamma contributions separately.

	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)

Table 7. Shielding rankings of fission products and light elements with greater than1% of total dose at a 5-year cooling time

^{*a*}Gamma shields consist of 27-cm steel, 12.7-cm lead, and 50-cm concrete for the iron, lead, and concrete casks, respectively.

^bPercentage contribution to the total dose.

- 1. The three cask models studied exhibit similar trends with respect to the neutron-vs-gamma-ray contributions to the total dose. The primary gamma rays dominate the total dose for the first 50 to 100 years; the neutron/secondary gamma contribution dominates the remainder of the time up to 10,000 years. The neutron-vs-secondary-gamma contributions vary appreciably by cask type because of the differing attenuation and secondary particle generation properties of the shield materials.
- 2. For short cooling times (less than 100 years), ²⁴⁴Cm dominates the actinide contributions to the total dose. However, over the same time period the primary gamma dose dominates the total dose, except for the lead cask, where the neutron doses overtake those due to primary gammas at about 50 years. The fractional contribution of ²⁴⁴Cm to the total dose increases with increasing burnup.
- 3. For long cooling times (greater than 100 years), the actinides ²³⁹Pu, ²⁴⁰Pu, and ²⁴²Pu dominate the total dose. For decay times approaching 10,000 years, ²¹⁴Bi becomes increasingly important because it is a daughter of several long-lived actinides.
- 4. The dose contributions for actinides ²⁴⁰Pu, ²⁴²Pu, and ²¹⁴Bi are relatively insensitive to burnup. The dose contribution due to ²³⁹Pu decreases somewhat with increasing burnup because of its tendency to contribute significantly to the reactor power in the latter stages of burnup.
- 5. ⁶⁰Co dominates the contribution to the total dose rate from about 3 to 30 years; however, the initial levels of cobalt vary significantly. The initial amount of cobalt assumed in this work was 0.5% for the Inconel grid spacers, which is typically an upper limit for older assemblies. Most newer assemblies contain significantly lower initial concentrations, effectively lowering the large contributions seen for ⁶⁰Co.
- 6. ¹⁴⁴Pr is a very important contributor to the total dose for decay times of 5 years or less. Thereafter, the contribution decreases rapidly due to the 285-d half-life of its precursor, ¹⁴⁴Ce. The ¹⁴⁴Pr ranking is insensitive to burnup since it is a direct product of fission and its concentration should grow proportionally to the total burnup.
- 7. ¹³⁴Cs is an important contributor to the total dose during the 2- to 10-year time frame. Its contribution typically peaks at about 5 years. For higher burnups, the relative contribution increases because it is not a direct fission product but rather is produced from ¹³³Cs capture.
- 8. ¹⁵⁴Eu contributes substantially to the total dose between 5 and 50 years. The contribution peaks at about 20 years. The contribution to dose also increases with burnup due to its production from capture in ¹⁵³Eu.
- 9. The importances of the remaining fission products (other than those reasons mentioned in 5 through 7 above) decrease with increasing burnup because of the faster buildup of actinides relative to fission products during extended irradiations.
- 10. ¹⁰⁶Rh is an important contributor to the total dose but only for fairly short cooling times of 2 years or less.

11. ^{137m}Ba can be important for thin shields, but since its energy is somewhat low (0.66 Mev) it becomes less important for thick shields. The enhanced importance is also seen for the concrete cask, where low-energy gamma rays can be passed more readily than in the high-Z iron and lead casks.

3.3 CURIE RANKINGS

The curie rankings for this study are given in Table 8 for two burnups (20 GWd/t and 50 GWd/t) and their corresponding enrichments (3.0 wt % and 4.5 wt %). These rankings are presented for decay times of 5 and 10,000 years after irradiation. Plots of the fractional contribution to the total number of curies for the various actinides, fission products, and light elements are shown in Appendix C for ten decay times ranging from 2 to 10,000 years.

The curie rankings in Table 8, along with the total curies plots shown in the Appendix, show the domination of the fission products for decay times less than 200 years. After that time, the actinides are the dominant contributor to the total curie levels in the spent fuel. The primary actinide contributors at early decay times (²⁴¹Pu, ²³⁸Pu, and ²⁴¹Am) decay such that at the 10,000-year period, shown in Table 8, ²³⁹Pu and ²⁴⁰Pu are the primary contributors. For high burnups, ²⁴³Am and its daughter, ²³⁹Np, also contribute a few percent to the total curie levels.

The dominant fission-product contributors to the total curie levels at decay times less than 200 years exist primarily in parent-daughter pairs in secular equilibrium (i.e., they have identical activities). These parent-daughter pairs include the ¹³⁷Cs-^{137m}Ba, ⁹⁰Sr-⁹⁰Y, ¹⁴⁴Ce-¹⁴⁴Pr, and ¹⁰⁶Ru-¹⁰⁶Rh pairs, but ¹⁴⁷Pm, ¹³⁴Cs, and ⁸⁵Kr also contribute. All these fission products are essentially decayed out at 200 years. The only fission product that contributes appreciably beyond this time period is ⁹⁹Tc, which has a 213,000-year half-life.

3.4 DECAY HEAT RANKINGS

The decay heat rankings for this study are given in Table 9 for two burnups (20 GWd/t and 50 GWd/t) and their corresponding enrichments (3.0 wt % and 4.5 wt %). These rankings are presented for decay times of 5 and 10,000 years after irradiation. Plots of the fractional contribution to the total decay heat for the various actinides, fission products, and light elements are shown in Appendix D for ten decay times ranging from 2 to 10,000 years.

The decay heat rankings in Table 9, along with the total decay heat plots shown in Appendix D, show the domination of the fission products for decay times less than 70 years. After that time, the actinides are the dominant contributor to the total decay heat levels in the spent fuel. For the low-burnup case, the primary actinide contributors at early decay times, ²³⁸Pu and ²⁴¹Am, decay until at the 10,000-year period, shown in Table 9, ²³⁹Pu and ²⁴⁰Pu are the primary contributors. For high burnups, ²⁴⁴Cm and ²⁴³Am also contribute nonnegligible amounts to the total decay heat. The dominant fission-product contributors to the total decay heat levels at decay times less than 70 years exist largely in parent-daughter pairs in secular equilibrium. These pairs include the ¹³⁷Cs-^{137m}Ba, ⁹⁰Sr-⁹⁰Y, ¹⁴⁴Ce-¹⁴⁴Pr, and ¹⁰⁶Ru-¹⁰⁶Rh pairs, but ¹³⁴Cs, ¹⁵⁴Eu, and the light-element ⁶⁰Co also contribute. These parent-daughter pairs, while at secular equilibrium, have differing contributions to the total decay heat, since the Q-values or the heat generated per decay differ between the parent and daughter nuclides. All these fission products are essentially decayed out at 200 to 300 years. No fission products contribute appreciably to the decay heat beyond this time period.

	5 y	ears	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-241	$1(17)^{a}$	1(17)	-	-	
Pu-238	2(0.2)	2(0.7)	-	-	
Am-241	3(0.2)	3(0.2)	-	-	
Pu-240	-	-	2(32)	2(38)	
Pu-239	-	-	1(62)	1(49)	
Am-243	-	-	-	3(3)	
Np-239	-	-	-	4(3)	
Fission products an	d light elements				
Cs-137	1(15)	1(17)	-	-	
Ba-137m	2(14)	2(16)	-	-	
Sr-90	3(12)	3(12)	-	-	
Y-90	4(12)	4(12)	-	-	
Pm-147	5(10)	5(6)	-	-	
Ce-144	6(3)	9(2)	-	-	
Pr-144	7(3)	10(2)	-	-	
Ru-106	8(3)	7(3)	-	-	
Rh-106	9(3)	8(3)	-	-	
Cs-134	10(3)	6(6)	-	-	
Kr-85	11(1)	11(1)	-	-	
Tc-99	-	-	1(2)	1(3)	

Table 8. Curie rankings of actinides, fission products, and light elements with
greater than 0.1% of total curies at 5 and 10,000 years

^aPercentage contribution to the total curie levels.

	5 y	ears	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 an	d 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)	-	-	

Table 9. Decay heat rankings of actinides, fission products, and light elements with greater than 1% of total decay heat at 5 and 10,000 years

^{*a*}Percentage contribution to the total decay heat levels.

3.5 RADIOLOGICAL TOXICITY RANKINGS

The ranking of radiological toxicity, especially in regard to environmental dose, is a complex problem. A complete analysis must take into account leakage of nuclides from the fuel assemblies and environmental pathway analyses, both of which are beyond the scope of the present study. In this work, the measure of radiological toxicity has been taken to be the potential committed effective dose equivalent for exposure either by ingestion or inhalation. The adjective "potential" is used because the full amount of each isotope present in the assembly is considered when computing the committed effective dose equivalent. No credit has been taken for the retention of a nuclide in the assembly or for losses along an environmental pathway from the assembly location to the location of an exposed individual.

The activity of each nuclide was multiplied by the committed effective dose equivalent per unit intake for the exposure mode of interest. These conversion coefficients were taken from Federal Guidance Report No. 11.⁸ For the inhalation exposure mode, a nuclide generally has two or three conversion coefficients corresponding to compounds that have different clearance times from the lung. For ingestion exposure, some nuclides have more than one conversion coefficient because different compounds containing the nuclide will have a different fraction of the compound reaching body fluids after ingestion. For those nuclides that have more than one conversion coefficient used was the one that yields the largest effective dose equivalent.

Effective dose equivalent¹⁰ is a weighted sum of doses to individual organs and tissues of the body. The weighting factor for an individual organ or tissue corresponds to the fractional contribution of that organ or tissue to the total risk of stochastic effects when the entire body is uniformly irradiated. The committed dose is the sum of all doses projected to be received in the future from an intake at a given time. By convention, the sum is taken over 50 years, which represents the arbitrarily assumed remaining lifetime of an exposed adult worker. The conversion coefficients are derived from (1) metabolic models that represent chemical transport of compounds within, and excretion from the body; and (2) studies of radiation transport between pairs of source and target organs for radiations emitted by the nuclides of interest, including alphas, betas, discrete electrons, X rays, and gamma rays. Radioactive decay within the body is explicitly included. The details of these calculations can be found in Federal Guidance Report No. 11,⁸ ICRP Publication 26,¹⁰ and ICRP Publication 30 (ref. 11).

The rankings do not include elemental tritium, or any noble gas (argon, krypton, and xenon) nuclides. The primary exposure mode for these nuclides is submersion dose. To estimate submersion dose, the concentration of the nuclide in air is required. Since there is no way to relate a unit concentration of nuclide in air to the amount in the fuel assembly, no rankings can be computed here. In addition, the rankings do not include ²²⁰Rn, ²²²Rn, and their daughter products. Dose from these nuclides and their daughters is particularly difficult to calculate, and radiation protection for these nuclides is expressed in terms of exposure to the chain of parent and daughters in units of Working Level Months.⁸

The radiological toxicity rankings for exposure by ingestion and inhalation are given in Tables 10 and 11, respectively. Each table has rankings for two burnups (20 GWd/t and 50 GWd/t) and the corresponding enrichments (3.0 wt % and 4.5 wt %), presented for decay times of 5 and 10,000 years. Plots of the fractional contribution to the potential committed effective dose equivalent from ingestion and inhalation are shown in Appendix E for actinides and for fission products plus ⁶⁰Co at decay times from 2 through 100,000 years.

	5 ye	ears	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-241	$1(35)^{a}$	3(18)	-	-
Pu-238	2(20)	1(37)	-	-
Am-241	3(18)	4(11)	-	-
Pu-240	4(9)	5(4)	2(33)	2(41)
Pu-239	5(8)	6(3)	1(66)	1(54)
Cm-244	6(4)	2(24)	-	-
Am-243	7(0.1)	7(0.3)	3(0.5)	3(3)
Cm-243	-	8(0.2)	-	-
Am-242m	-	9(0.1)	-	-
Np-237	-	-	4(0.3)	5(0.4)
Pu-242	-	-	5(0.2)	4(0.6)
U-234	-	-	6(0.1)	6(0.2)
Fission Products				
Sr-90	1(4)	1(2)	-	-
Ru-106	2(0.4)	2(0.2)	-	-
Ce-144	3(0.3)	-	-	-
Cs-137	4(0.1)	-	-	-
Pm-147	5(0.1)	_	-	-

Table 10.	Ingestion toxi	city rankings	of actinides	and fission	products	with greater	than
0.1% c	of total potentia	al committed	effective dos	se equivalen	t at 5 and	10,000 year	S

^aPercentage contribution to the total potential committed effective dose equivalent.

	5 y	ears	10,000	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-241	$1(19)^{a}$	3(13)	-	-		
Pu-238	2(11)	1(25)	-	-		
Am-241	3(10)	4(7)	-	-		
Pu-240	4(5)	5(3)	2(33)	2(41)		
Pu-239	5(4)	6(2)	1(66)	1(54)		
Cm-244	6(2)	2(16)	-	-		
Am-243	-	7(0.2)	3(0.5)	3(3)		
Cm-243	-	8(0.1)	-	-		
Np-237	-	-	4(0.3)	5(0.4)		
Pu-242	-	-	5(0.2)	4(0.6)		
Fission Produ	cts and Light Elem	ents				
Sr-90	1(27)	1(18)	-	-		
Cs-137	2(12)	2(9)	-	-		
Cs-134	3(4)	3(4)	-	-		
Y-90	4(2)	4(1)	-	-		
Ru-106	5(1)	5(0.8)	-	-		
Ce-144	6(1)	6(0.4)	-	-		
Co-60	7(0.4)	7(0.2)	-	-		
Pm-147	8(0.2)	-	-	-		

Table 11. Inhalation toxicity rankings of actinides, fission products, and light elements with greater than 0.1% of total potential committed effective dose equivalent at 5 and 10,000 years

^{*a*}Percentage contribution to the total potential committed effective dose equivalent.

In addition, Appendix E contains plots of the relative contributions of the light-element, actinide, and fissionproduct groups to the total for the same range of decay times. It is clear from the latter plots that the actinides are the primary contributors to the potential committed effective dose equivalent. For ingestion exposure, the actinide group is the only major contributor for all decay times from 2 through 100,000 years. For inhalation exposure, the actinide group is the only major contributor from 100 through 100,000 years. For times less than 5 years, the fission-product group is a larger contributor than the actinide group for low-enrichment, lowburnup fuel. From 10 to 100 years, it remains a major contributor to inhalation exposure, but rapidly decreases in importance between 100 and 200 years. The light-element group is never a significant contributor to radiological toxicity.

In the light-element group, ⁶⁰Co is the only noticeable contributor for inhalation exposure, and only for decay times less than 10 years. It is negligible for greater decay times. No light-element nuclide makes a contribution greater than 0.1% for ingestion exposure at any decay time.

No fission product makes a significant contribution after 200 years decay time. Among the fission products, ⁹⁰Sr is the largest contributor for both exposure modes, except for the ingestion of ¹⁴⁴Ce at 2 years from low-enrichment, low-burnup fuel. For decay times less than 5 years, ¹⁴⁴Ce and ¹⁰⁶Ru are of interest for both exposure modes. For inhalation exposure, ¹³⁷Cs is of moderate importance until 100 years, but is barely of interest for ingestion exposure at decay times of 10 years or less and only for low-enrichment, low-burnup fuel. From 2 to 10 years decay time, ¹³⁴Cs is moderately important for inhalation exposure. At times of 5 years or less, ¹⁴⁷Pm is noticeable, but of small importance for both exposure modes.

As mentioned earlier, the actinide group contains the principal contributors to committed effective dose equivalent for all except the shortest decay times. For decay times from 2 through 100 years, ²³⁸Pu is important for both exposure modes. During this same time period, ²⁴¹Am steadily increases in importance, remains the principal contributor for both exposure modes until 1,000 years, ^{then} rapidly decreases, becoming of no interest after 3,000 years decay. For times less than 20 years, ²⁴¹Pu is important for both exposure modes, but loses importance rapidly, becoming of no interest after 100 years. The importance of ²⁴⁰Pu is moderate at decay times less than 100 years, but it becomes a major contributor at times between 200 and 30,000 years, with a peak at 3,000 years decay. The behavior of ²³⁹Pu is the same as that of ²⁴⁰Pu out to a decay time of 3,000 years. However, it keeps on increasing in importance, becoming the most important contributor for decay times greater than 10,000 years. For high-enrichment, high-burnup fuel, ²⁴³Am makes a modest contribution in the period from 1,000 to 30,000 years; for low-enrichment, low-burnup fuel, it makes only a small contributors: ²³⁷Np, ²⁴²Pu, and ²²⁹Th for both exposure modes; ²¹⁰Pb, ²¹⁰Po, and ²²⁶Ra only for inhalation exposure; and ²³⁴U and ²³⁰Th only for ingestion exposure. The other nuclides shown on the actinide plots make only small contributions at this time.

Note that conversion factors relating committed effective dose equivalent to ingestion or inhalation are not available for all radioactive isotopes in the ORIGEN-S decay library; therefore, some unknown fraction of the total committed effective dose equivalent is being neglected for both exposure modes. Given the energies and intensities of all radiations emitted by a nuclide, it is possible to derive conversion factors for that nuclide. When only the total alpha, beta, gamma, and neutron energy emitted by a nuclide is known, it should still be possible to derive a conservative conversion factor for that nuclide. However, the effort required to derive the additional conversion factors is far beyond the scope of the current work. In an effort to gain some basic understanding of the effect of neglecting those nuclides having no conversion factors, plots have been included in Appendix E that give the fraction of activity neglected in computing the committed effective dose equivalents. It can be seen from these plots that no light-element activity of consequence has been omitted. The neglected fraction of actinide activity is less than 1% for times less than 30,000 years, and reaches a maximum of about 9% at 100,000 years. The neglected fraction of fission product activity, is roughly 20% at 2 years decay time, slowly increases to a maximum of about 30% at 200 years decay time, and decreases rapidly (becoming negligible) at times of 1,000 years or greater. Considering the time dependence of the

actinide and fission-product group contributions to the committed effective dose equivalent (also shown in Appendix E), the effect of the neglected isotopes should be relatively small, since conversion factors have been tabulated for most nuclides of radiological significance. That is, the estimates of the total committed effective dose equivalent are low by some unknown (but probably fairly small) factor, and the relative rankings of nuclides that are significant contributors should not change.

3.6 SUMMARY

The previous sections have described the rankings of the various actinides, fission products, and light elements with respect to five different analysis areas. Tables 12 and 13 present these rankings in summary form. Table 12 gives the rankings for the actinides, and Table 13 gives rankings for the fission products and light elements. A scheme for easy selection of the most important nuclides for either short or long cooling times was chosen to represent these rankings in the two tables. The use is made of a high, medium, and low ranking, where high is defined to be a 10% or greater contribution to the total response; a medium ranking falls between 1 and 10% of the total response; and a low ranking corresponds to 0.1 to 1% contribution to the total response. To account for the variation in nuclide importance with decay, separate rankings are tabulated for short and long cooling times. For the purposes of this designation, short cooling times are assumed to be 100 years or less.

For the actinides, a number of nuclides, primarily the major plutonium, americium, and curium isotopes, have high-to-medium rankings in most or all of the analysis areas. The uranium isotopes are largely important only for criticality.

For the fission-product and light-element isotopes shown in Table 13, very few nuclides have importances in several areas. The shielding, curies, decay heat, and radiological toxicity categories are closely related and have several important isotopes in common. For the criticality rankings, there are no high importances for fission products, and only four isotopes have medium importances. However, there are some 16 nuclides with low importances, the sum of which would be a medium or high importance. This fact can complicate the validation process for these materials because of the large number of isotopes that must be characterized.

These importance rankings should facilitate validation efforts for radionuclide generation/depletion codes by indicating the nuclides that have the most significant effect on the responses of interest, and allowing experimental efforts to be emphasized for those nuclides. Furthermore, although good measurement accuracy is needed to minimize uncertainty in the validation process, the degree of accuracy needed may vary from nuclide to nuclide. These differing nuclide requirements are due to the differing accuracy requirements in the various analysis areas (e.g., acceptable criticality predictions are expected to be within a percent of k_{eff} measurements, whereas agreement within 10% could be acceptable for shielding applications). These rankings by analysis area should allow the varying accuracy requirements to be met in the most efficient manner. Indeed, certain groups of elements are more amenable to accurate measurements than others. For example, shown in Table 14 are applicable measurement techniques (see ref. 12) for a number of isotopes and their corresponding target accuracies. The highly accurate thermal-emission, isotopic dilution mass spectrometry (TEIDMS) method appears to be suitable for most of the important actinide absorbers and fission-product absorbers. For the important fission products where the TEIDMS method has not been applied, the radiochemical analysis (RCA) method appears to be of sufficient accuracy for the shielding and decay heat verification studies.

										Tox	cicity	
Actinides	Absorption		Shielding		Curies		Watts		Ingestion		Inhalation	
	Short ^b	Long ^c	Short	Long	Short	Long	Short	Long	Short	Long	Short	Long
Pb-210										L		М
Bi-214			Μ									
Po-210										L		Μ
Ra-225												L
Ra-226										L		Μ
Ac-227										Μ		L
Th-229										М		М
Th-230										М		L
Pa-231										L		L
U-233										L		L
U-234	L	L								М		L
U-235	Н	Н										
U-238	Н	Н								L		L
Np-237	L	М								Μ		М
Np-239						М						
Pu-238			Μ		Μ		Н		Н	Μ	Н	Н
Pu-239	Н	Н	L	М	L	Н	М	Н	М	Н	М	Н
Pu-240	М	М	Μ	Н	L	Н	М	Н	М	Н	М	Н
Pu-241	М				Н				Н		Н	
Pu-242	L	L	L	Н						Μ		М
Am-241	М		Μ		Μ	Н	Н	Н	Н	Н	Н	Н
Am-242m									L			
Am-243				L		М		Μ	L	М	L	М
Cm-242							L		L		L	
Cm-243									L		L	
Cm-244			Н				М		Н		Н	

Table 12. Summary rankings by analysis area $(actinides)^a$

 a H–High ranking (>10% of total); M–medium ranking (1 to 10% of total); L–low ranking (0.1 to 1% of total). The sum of all H, M, and L rankings should include virtually all (>95%) of the total response.

^bShort cooling times (<100 years).

^cLong cooling times (>100 years).

									Toxicity			
Isotope	Absor	ption	Shiel	lding	Cu	ries	Wa	atts	Inges	stion	Inhal	ation
	Short ^b	Long ^c	Short	Long	Short	Long	Short	Long	Short	Long	Short	Long
H-3									$\mathbf{S1}^{d}$		S 1	
C-14									$S2^d$	S2	S 2	S2
Co-60			Н				Μ		L		L	
Ni-59					$S3^d$	S 3						
Ni-63					S 3	S 3						
Kr-85					L							
Sr-90					Н		Μ		М		Н	L
Y-90			М		Н		Н		L		Μ	
Nb-94					L							
Mo-95		L										
Tc-99	L	L				Μ						
Ru-101	L	L										
Ru-106					М				М		М	
Rh-103	М	М										
Rh-106			Н		М		Н					
Pd-105	L	L										
Ag-109	L	L										
Ag-110m			L		М							
Xe-131	L	L										
Cs-133		L										
Cs-134			Н				Н				М	
Cs-137					Н		М		L		Н	L
Ba-137m			Н		Н		Н					
Ce-144					М		L		М			
Pr-141	L	L										
Pr-144			Н				Н				L	
Nd-143	М	М										
Nd-145	L	L										
Pm-147					М		М		L		L	
Sm-147	L	L										
Sm-149	М	М										
Sm-150	L	L										
Sm-151	L	L			L							
Sm-152	L	L										
Eu-151	L	L										
Eu-153	L	L										
Eu-154			М		М		М					
Gd-155	М	М										
Gd-157	L	L										

Table 13. Summary rankings by analysis areas (fission products and light elements)^a

^{*a*}H—high ranking (>10% of total); M—medium ranking (1% to 10% of total); L—low ranking (0.1% to 1% of total). The sum of all H, M, and L rankings should include virtually all (>95%) of the total response.

^bShort cooling times (<100 years).

^cLong cooling times (>100 years).

^{*d*}Special categories include the following:

S1 - Included due to special status in dosimetry studies.

S2 - Included because of importance to long-term waste package performance assessment.

S3 - These nuclides were not specifically identified in the analysis as important, but are included because of their special status in determining greater than Class C disposal limits of waste storage.

Element or				Element or			
radioisotope	SSMS ^a	$TEIDMS^b$	RCA ^c	radioisotope	SSMS ^a	TEIDMS ^b	RCA^{c}
т:	v			Sm 154	v	v	
LI Ti	Λ V			SIII-134 En 153	Λ V		
11 Cr	Λ V			Eu-155 Eu 154	Λ V	A V	
E	л V			Eu-154 Eu 155	л V	A V	
Ni	A V			Eu-133 Nd 143	A V	A X	
Mo	X			Nd-143 Nd-144	X	X	
Th	X			Nd-145	X	X	
R	X			Nd-145 Nd-146	X	X	
D V	X			Nd-148	X	X	
v Mn	X			Nd-150	X	X	
Co	X			Th-232	X	Λ	
Nh	X			11-232 11-232	11		X
	X			U-232	x	x	24
Pu	21	x		U-234	21	X	
Se-79	x	21		U-235		X	
Sr-90	X		x	U-236		X	
Zr-93	X			U-238		X	
Nb-94	X			Np-237	X	1	Х
Tc-99	X	Х		Pu-238		Х	
Ru-106	X		Х	Pu-239		X	
Pd-107	Х		Х	Pu-240		Х	
Sb-125	Х		Х	Pu-241		Х	
Sn-126	Х			Pu-242		Х	
I-129	Х		Х	Pu-244		Х	
Cs-133	Х			Am-241			Х
Cs-134	Х		Х	Am-242			Х
Cs-135	Х			Am-242m			Х
Cs-137	Х		Х	Am-243			Х
Ce-144	Х	Х	Х	Cm-242			Х
Pm-147	Х			Cm-243			Х
Sm-147	Х	Х		Cm-244			Х
Sm-148	Х	Х		Cm-245			Х
Sm-149	Х	Х		Cm-246			Х
Sm-150	Х	Х		Cm-247			Х
Sm-151	Х	Х		Cm-248			Х
Sm-152	Х	Х					

Table 14. Accuracy and applicability of current analytical methods

^{*a*}Beta-gamma spark-source mass spectrometry, $\pm 25\%$. Isotope dilution, in cases where an enriched spike is available, can be used to obtain $\pm 10\%$ precision. Chemical separations from elements with the same mass numbers may be necessary.

^bThermal-emission, isotopic dilution mass spectrometry, ±1 to 2%.

^{*c*}Radiochemical analyses, ±5 to 10%.

^{*d*}Uranium is often analyzed by extraction and a Davies-Gray potentiometric titration, $\pm 1\%$.

4. CONCLUSIONS

This study has investigated the relative importances of the various actinide, fission-product, and lightelement isotopes with respect to five analysis areas: criticality safety (absorption fractions), shielding (dose rate fractions), curies (fractional curie levels), decay heat (fraction of total watts), and radiological toxicity (fraction of committed effective dose equivalent). These rankings were presented for up to six different burnup/enrichment scenarios and at decay times from 2 to 100,000 years. For completeness, rankings in each of these analysis areas are plotted in the appendixes, as well as being summarized in Tables 2 through 11 in the main body of the report. In addition, Tables 12 and 13 give summary rankings in terms of high (greater than 10% contribution to the total), medium (between 1 and 10% contribution), and low (0.1 to 1% contribution) for both short- and long-term cooling. When compared with the target measurement accuracies given in Table 14, these rankings show that most of the important isotopes can be characterized sufficiently for the purpose of radionuclide generation/depletion code validation in each of the analysis areas.

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APPENDIX A

CRITICALITY SAFETY PLOTS

This appendix contains complete listings of all plots generated in this ranking study. For the criticality safety rankings, plots of the fractional absorption by nuclide over decay times from 2 to 100,000 years are included. For each of the six enrichment/burnup groups, there are three plots corresponding to:

- 1. the fractional absorption of both actinides and fission products, and the total number of absorptions, for cooling times from 2 to 100,000 years;
- 2. the fractional absorption in each of the top actinides for cooling times of 2 to 100,000 years; the legend gives the ranking (in terms of absorptions) of the actinides at 5 and 30,000 years;
- 3. the fractional absorption in each of the top fission products for cooling times of 2 to 100,000 years; the legend gives the ranking (in terms of absorptions) of the fission products at 5 and 30,000 years.









Absorptions vs Cooling Time 3.0 wt % U-235, 35 GWd/t









Absorptions vs Cooling Time 4.0 wt % U-235, 30 GWd/t Results with SCALE 44GROUPNDF5





Absorptions vs Cooling Time 4.0 wt % U-235, 40 GWd/t Results with SCALE 44GROUPNDF5

















Absorptions vs Cooling Time 4.5 wt % U-235, 50 GWd/t Results with SCALE 44GROUPNDF5







APPENDIX B

DOSE RATE FRACTION PLOTS

This section contains the complete listing of all plots generated for the shielding ranking portion of this work. Plots are given for three cask types: iron, lead, and concrete for two burnup/enrichment combinations—3.0 wt %, 20 GWd/t, and 4.5 wt %, 50 GWd/t. Five plots are given for each of these six cases corresponding to:

- 1. the variation in the total dose rate (mrem/h) over the entire cooling time of 2 to 10,000 years, followed by the fraction of the total doses due to neutrons, primary gammas, and secondary gammas;
- 2. the fraction of the total dose rates due to neutrons from selected actinides; the legend gives the relative rankings for these actinides at 5 and 10,000 years;
- 3. the fraction of the total dose rate (mrem/h) due to primary gammas from selected actinides; the legend gives the relative rankings for these actinides at 5 and 10,000 years;
- 4. the fraction of the total dose rate (mrem/h) due to primary gammas from selected fission products and light elements; the legend gives the relative rankings for these isotopes at 5 and 10,000 years;
- 5. the fraction of the total dose rate (mrem/h) due to secondary gammas from selected actinides; the legend gives the relative rankings for these isotopes at 5 and 10,000 years.
































Dose Rate vs Cooling Time TN-24 Cask; 4.5 wt % U-235, 50 gwd/t































APPENDIX C

CURIE LEVEL FRACTION PLOTS

This section contains the complete listing of all plots generated for the curie ranking portion of this work. Plots are given for two burnup/enrichment combinations: 3.0 wt %, 20 GWd/t, and 4.5 wt %, 50 GWd/t. Three plots are given for each of these two cases corresponding to:

- 1. the total curies for ten decay periods from 2 to 10,000 years, followed by the fractional contributions from the actinides, light elements, and fission products;
- 2. the fraction of the total curie levels due to individual light elements and fission products; the legend gives the relative rankings for these isotopes at 5 and 10,000 years;
- 3. the fraction of the total curie levels due to individual actinides; the legend gives the relative rankings for these isotopes at 5 and 10,000 years.

















APPENDIX D

DECAY HEAT FRACTION PLOTS

This section contains the complete listing of all plots generated for the decay heat ranking portion of this work. Plots are given for two burnup/enrichment combinations: 3.0 wt %, 20 GWd/t, and 4.5 wt %, 50 GWd/t. Three plots are given for each of these two cases corresponding to:

- 1. the total decay heat in watts for ten decay periods from 2 to 10,000 years, followed by the fractional contributions from the actinides, light elements, and fission products;
- 2. the fraction of the total decay heat levels due to individual light elements and fission products; the legend gives the relative rankings for these isotopes at 5 and 10,000 years;
- 3. the fraction of the total decay heat levels due to individual actinides; the legend gives the relative rankings for these isotopes at 5 and 10,000 years.















APPENDIX E

RADIOLOGICAL TOXICITY FRACTION PLOTS

This section contains the complete set of plots generated for the radiological toxicity ranking portion of this work. Plots are given for two burnup/enrichment combinations: 3.0 wt %, 20 GWd/t, and 4.5 wt %, 50 GWd/t. All plots include decay times from 2 through 100,000 years. Seven plots are given for each of these two cases, corresponding to:

- 1. the fraction of potential committed effective dose equivalent from ingestion of individual actinides;
- 2. the fraction of potential committed effective dose equivalent from ingestion of individual fission products;
- 3. the fraction of potential committed effective dose equivalent from inhalation of individual actinides;
- 4. the fraction of potential committed effective dose equivalent from inhalation of individual fission products and ⁶⁰Co;
- 5. the fractional contributions from the light-element, actinide, and fission-product groups to the potential committed effective dose equivalent from ingestion;
- 6. the fractional contributions from the light-element, actinide, and fission-product groups to the potential committed effective dose equivalent from inhalation; and
- 7. the fraction of activity neglected in computing the committed effective dose equivalent.



Fraction of Potential HE,50,1ng






Fraction of Potential He, 50, mh



Fraction of Potential HE, 50, ing



Fraction of Potential He, so, inh



Fraction of Activity







Fraction of Potential He, 50, inh



Fraction of Potential He, so, inh



Fraction of Potential HE,50,ing



Fraction of Potential HE, 50, inh



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