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ABSTRACT

This paper describes the relative importances of various actinide, fission product and light element isotopes with respect to storage and transportation shielding applications for spent fuel. Using simple computational models, the relative contributions of many isotopes can be individually analyzed and ranked. This ranking of importances is useful for both validation and understanding of the underlying importances of the many nuclides that can contribute to various waste handling issues such as dose rate analysis, criticality safety, decay, heat generation and residual activity levels.

I. INTRODUCTION

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 analyses, (3) residual decay heat predictions for thermal considerations, and (4) activity and/or radiological toxicity levels for materials assumed to be released into the ground/environment after long periods of time. 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 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 activation product isotopes associated with LWR spent fuel with respect to three analysis areas: shielding (dose rate fractions), activity (fractional Curie levels), and decay heat (fraction of total Watts). Rankings are presented for two different burnup/enrichment scenarios and at decay times of 5 and 10,000 years. Rankings for each of these analysis areas are presented as summary tables. 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 other areas such as non-UO_2 high-level waste (HLW) and nonfuel-bearing components (NFBC).

II. APPROACH

A. Areas of Interest

The rankings generated in Ref. 1 covered the basic analysis areas of shielding, criticality safety, decay heat, activity levels, and radiological toxicity. Only those associated with shielding, activity, and decay heat are included in this paper; for the other application areas the reader is referred to Ref. 1 for the complete set of results. 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 waste management issues, 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. The fractional contributions to the total decay heat were obtained for two burnup/enrichment states. Similarly, fractional contributions to the total activity levels 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 shielding, decay heat generation and activity levels for the LWR spent fuel burnup/enrichment cases considered. Not explicitly treated in this work are other types of HLW (e.g., ^{233}U-fueled systems) and the associated irradiated components in a reactor (NFBC, pressure vessel, biological shield, etc.).

B. 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 using the SAS2H/ORIGEN-S computer code[2] assuming a reactor specific power of 40 MW/t and a power history with 80% uptime, 20% downtime in each cycle. Conservative (0.46 wt %) loadings of Co impurity, typical of older fuel assembly designs, were assumed in the Inconel grid spacer material. The base cross-section library used in these burnup/depletion calculations was the SCALE 44-group library,[3] based primarily on ENDF/B-V data, with ^{16}O, ^{154}Eu, and ^{155}Eu data obtained from ENDF/B-VI. Major changes were made in the ENDF/B-VI data for ^{154}Eu and ^{155}Eu; hence, updated cross sections were used in this study. Some analysis areas did not utilize all enrichment/burnup and cooling-time combinations. The shielding, decay heat, and activity rankings were desired at a low and a high burnup and used the 20-GWd/t and 50-GWd/t burnups with cooling times of 5 and 10,000 years.

Computed activity levels and decay heat values in Watts were obtained directly from the ORIGEN-S outputs. The activity and decay heat results were converted separately to fractional values. The rankings were tabulated separately for actinide and fission product materials to show their relative importances. Ref. 1 includes plots of the results for cooling times up to 10,000 years.

For the shielding rankings, the neutron and gamma-ray sources were taken from the ORIGEN-S outputs and input to the one-dimensional (1-D) discrete-ordinates module SAS1/XSDRNPM-S[4] 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. Shielding 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.

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

Enrichment, wt % ^{235}U	Burnup (GWd/t)	Estimated industry-average burnup (GWd/t)	Relative burnup
3.0	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

The rankings for each of the respective analysis areas are generated and presented as the *fractional* contribution to the *total* response. Thus for decay heat and activity, the fractional contributions are based on the total Watts and Curies, 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.

III. RESULTS

A. Shielding Rankings

The rankings for the shielding portion of this work are given in Tables 2 and 3 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. Plots of the fractional contribution to the total dose rates for the various actinides, fission products, and light elements are shown in Fig. 1−4 for high burnup fuel and the iron shipping cask for decay times ranging from 2 to 10,000 years. The results for the other cask designs are presented in Appendix B of Ref. 1.

Table 2. Shielding rankings of actinides with greater than 1% of total dose at 5 and 10,000 years

	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 %
5-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-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)
Bi-214	-/-/4(-/-/4)	-/-/3(-/-/4)	-/1/1(-/2/3)	-/1/1(-/1/3)	-/2/2(-/5/11)	-/-/3(-/-/11)

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

Table 3. Shielding rankings of fission products and light elements with greater than 1% of total dose at a 5-year cooling time

	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)

[a]Gamma shields consist of 27-cm steel, 12.7-cm lead, and 50-cm concrete for the iron, lead, and concrete casks, respectively.
[b]Percentage contribution to the total dose.

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 spectrum 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 key features seen in the rankings given in Tables 2 and 3 are the dominance of the actinide ^{244}Cm for a 5-year decay time 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 ^{240}Pu, ^{242}Pu, ^{239}Pu, and ^{214}Bi dominate the dose rate contributions. The specific features seen from an analysis of the plots shown in Appendix B of Ref. 1 include:

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

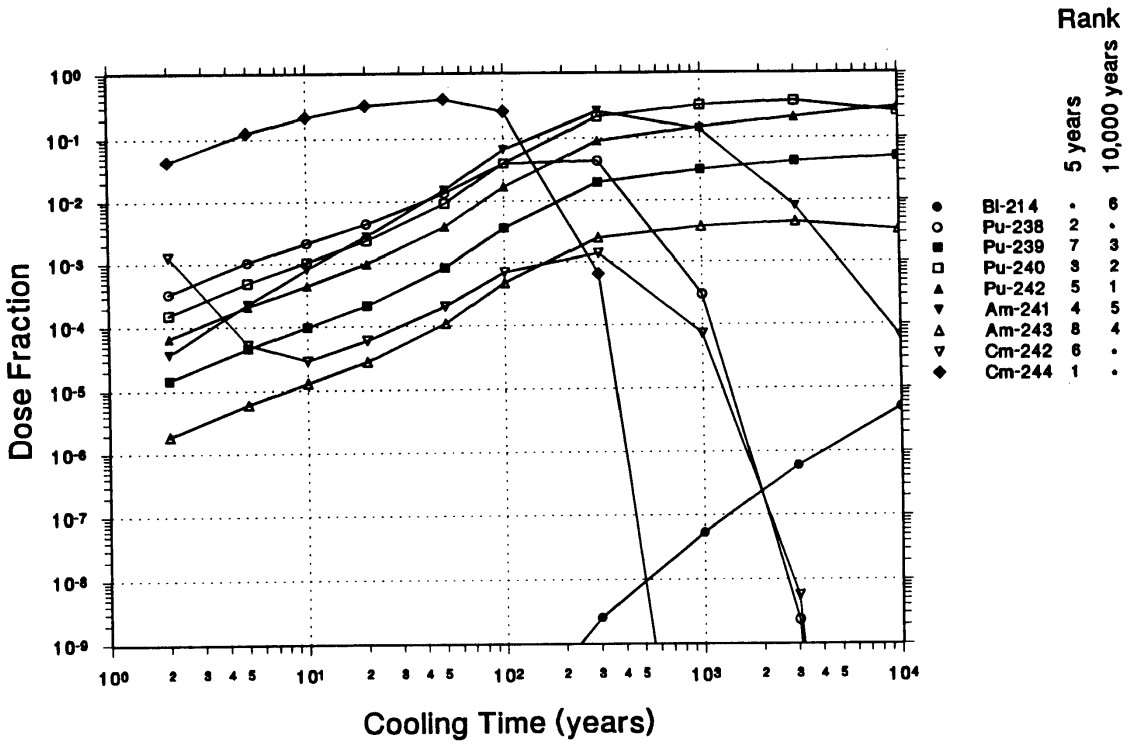

Figure 1. Neutron dose fraction for iron cask; 4.5 wt % ^{235}U, 50 GWd/t

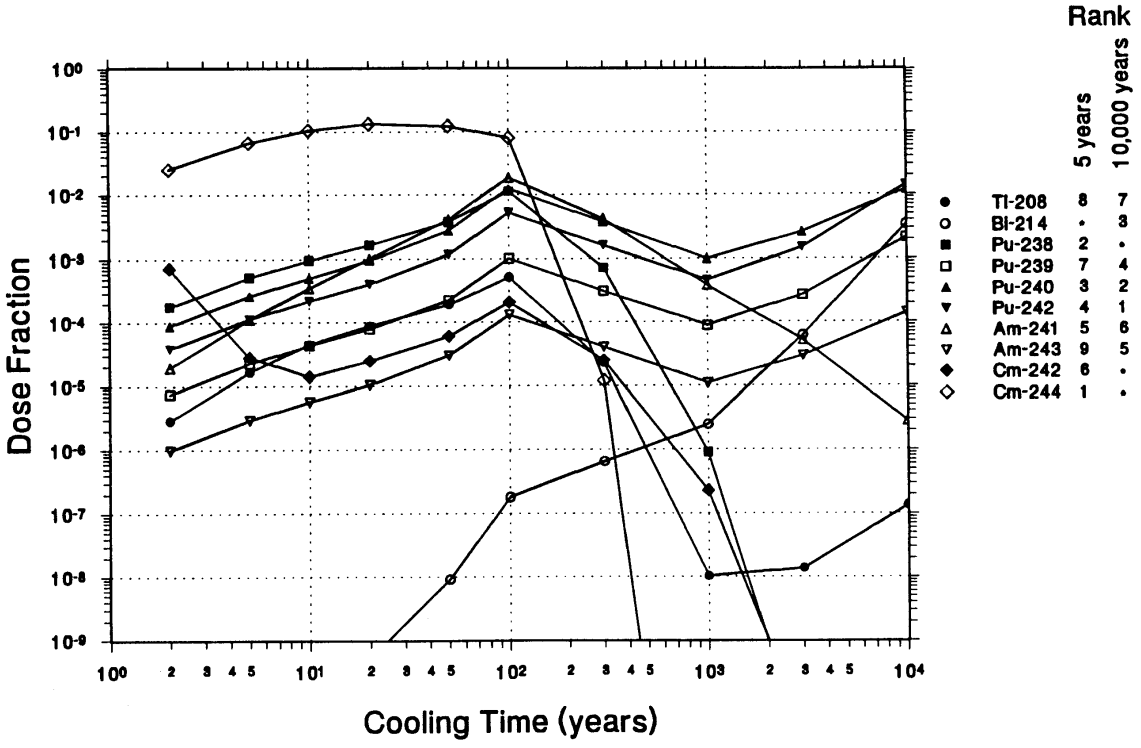

Figure 2. Primary gamma dose fraction (actinides) for iron cask; 4.5 wt % ^{235}U, 50 GWd/t

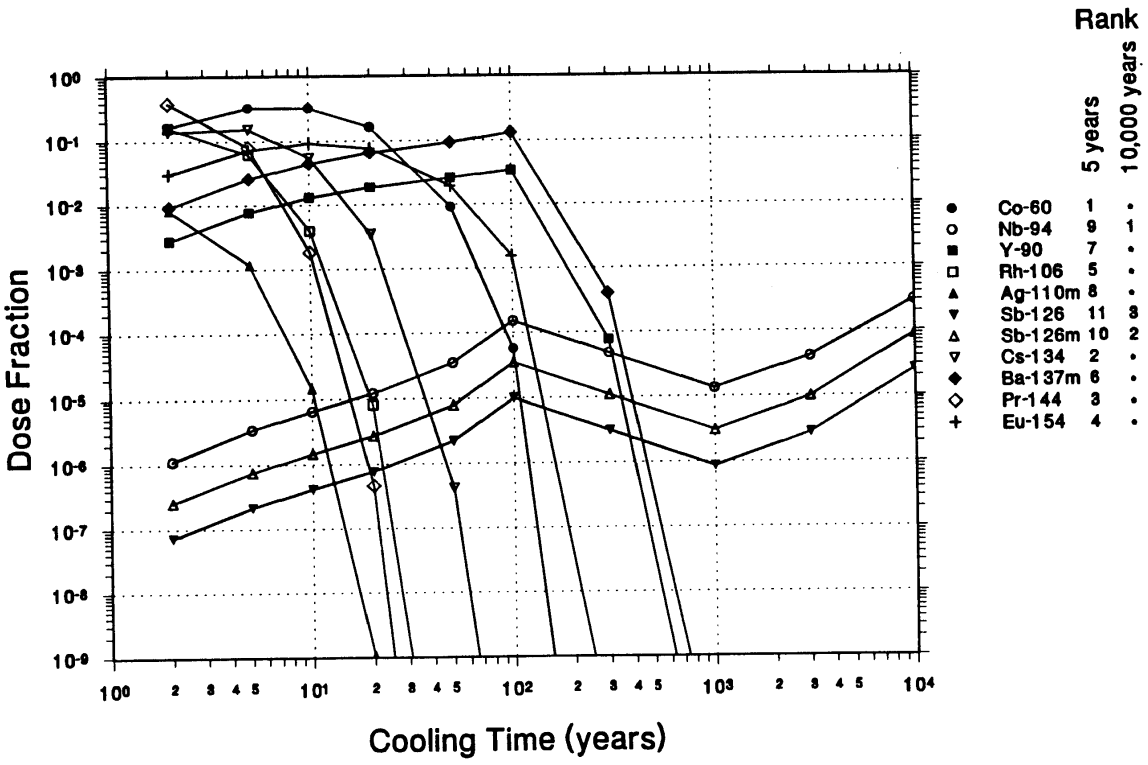

Figure 3. Primary gamma dose fraction (fission products and light elements) for iron cask; 4.5 wt % ^{235}U, 50 GWd/t

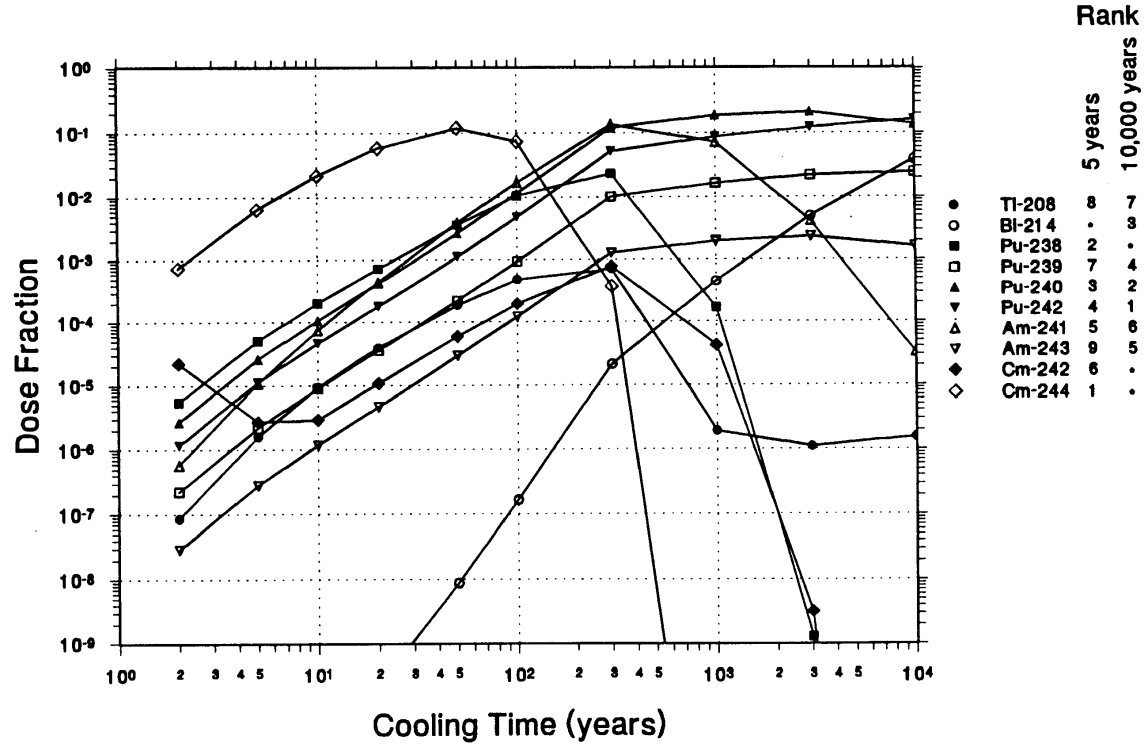

Figure 4. Secondary gamma dose fraction for iron cask; 4.5 wt % ^{235}U, 50 GWd/t

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), ^{244}Cm 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 dose exceeds that due to primary gammas at about 50 years. The fractional contribution of ^{244}Cm to the total dose increases with increasing burnup.

3. For long cooling times (greater than 100 years), the actinides ^{239}Pu, ^{240}Pu, and ^{242}Pu dominate the total dose. For decay times approaching 10,000 years, ^{214}Bi becomes increasingly important.

4. The dose contributions for actinides ^{240}Pu, ^{242}Pu, and ^{214}Bi are relatively insensitive to burnup. The dose contribution due to ^{239}Pu decreases somewhat with increasing burnup because of its tendency to contribute significantly to the reactor power in the latter stages of burnup.

5. ^{60}Co dominates the contribution to the total dose rate from about 3 to 30 years. However, 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 ^{60}Co in this study.

6. ^{144}Pr 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, ^{144}Ce. The ^{144}Pr ranking is relatively insensitive to burnup.

7. ^{134}Cs 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.

8. ^{154}Eu 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 ^{153}Eu.

9. The importances of most fission products decrease with increasing burnup because of the faster buildup of actinides relative to fission products during extended irradiations.

10. ^{106}Rh 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 penetrate more readily than in the high-Z iron and lead casks.

B. Activity Rankings

The activity rankings for this study are given in Table 4 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 of Ref. 1 for decay times ranging from 2 to 10,000 years.

The activity rankings in Table 4, along with the total activity plots shown in the Appendix of Ref. 1, 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 activity in the spent fuel. The primary actinide contributors at early decay times (^{241}Pu, ^{238}Pu, and ^{241}Am) decrease such that at the 10,000-year period, shown in Table 4, ^{239}Pu and ^{240}Pu are the primary contributors. For high burnups, ^{243}Am and its daughter, ^{239}Np, also contribute a few percent to the total activity.

The dominant fission-product contributors to the total activity 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 ^{137}Cs-^{137m}Ba, ^{90}Sr-^{90}Y, ^{144}Ce-^{144}Pr, and ^{106}Ru-^{106}Rh pairs, but ^{147}Pm, ^{134}Cs, and ^{85}Kr also contribute. All these fission products are essentially gone after 200 years. The only fission product that contributes appreciably beyond this time period is ^{99}Tc, which has a 213,000-year half-life.

C. Decay Heat Rankings

The decay heat rankings for this study are given in Table 5 for two burnups (20 GWd/t and 50 Gwd/t) and their corresponding enrichments (3.0 wt % and 4.5 wt %).

Table 4. Activity rankings of actinides, fission products, and light elements with greater than 0.1% of total Curies at 5 and 10,000 years

	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-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 and 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)

[a]Percentage contribution to the total Curie levels.

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 of Ref. 1 for decay times ranging from 2 to 10,000 years.

The decay heat rankings in Table 5 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, ^{238}Pu and ^{241}Am, decay until at the 10,000-year period, shown in Table 5, ^{239}Pu and ^{240}Pu are the primary contributors. For high burnups, ^{244}Cm and ^{243}Am also contribute significant 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 ^{137}Cs-^{137m}Ba, ^{90}Sr-^{90}Y, ^{144}Ce-^{144}Pr, and ^{106}Ru-^{106}Rh pairs, but ^{134}Cs, ^{154}Eu, and ^{60}Co also contribute. These parent-daughter pairs, while at secular equilibrium, have differing contributions to the total decay heat, since the energy released per decay differs 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.

IV. CONCLUSIONS

This study has investigated the relative importances of the various actinide, fission-product, and light-element isotopes with respect to three analysis areas: shielding (dose rate fractions), activity (fractional Curie levels), and decay heat (fraction of total Watts). These rankings were presented for two different burnup/enrichment scenarios and at decay times from 2 to 10,000 years. For completeness, rankings in each of these analysis areas are plotted in the appendices of Ref. 1, as well as being summarized in Tables 2 through 5 in this paper. In addition, Ref. 1 gives 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 to expected measurement

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

	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.

accuracies, summarized in Table 14 of Ref. 1, these rankings show that most of the important isotopes can be characterized sufficently for the purpose of radionuclide generation/depletion code validation in each of the analysis areas.

V. REFERENCES

1. B. L. Broadhead, M. D. DeHart, J. C. Ryman, J. S. Tang and C. V. Parks, "Investigation of Nuclide Importance to Functional Reguirements Related to Transport and Long-TermStorage of LWR Spent Fuel," ORNL/TM-12742, Oak Ridge National Laboratory (1995).

2. O. W. Hermann, "SAS2(H): A Coupled One-Dimensional Depletion and Shielding Analysis Code," Sect. S2 of *SCALE: A Modular Code System for Performing Standardized Computer Analyses for Licensing Evaluation*, NUREG/CR-0200, Revision 4 (ORNL/NUREG/CSD-2/R4), Vols. I, II, and III (draft February 1990). Available from Radiation Shielding Information Center as CCC-545.

3. M. D. DeHart, *Validation of the SCALE Broad Structure 44-Group ENDF/B-V Cross-Section Library for Use in Criticality Safety Analyses*, NUREG/CR-6102 (ORNL/TM-12460), U.S. Nuclear Regulatory Commission, September 1994.

4. J. R. Knight, C. V. Parks, S. M. Bowman, L. M. Petrie, and J. A. Bucholz, *SAS1: A One-Dimensional Shielding Analysis Module*, Sect. S1 of *SCALE: A Modular Code System for Performing Standardized Computer Analyses for Licensing Evaluation*, NUREG/CR-0200, Revision 4 (ORNL/NUREG/CSD-2/R4), Vols. I, II, and III (draft February 1990). Available from Radiation Shielding Information Center as CCC-545.