REVIEW OF RESULTS FOR THE OECD/NEA PHASE VII BENCHMARK: STUDY OF SPENT FUEL COMPOSITIONS FOR LONG−TERM DISPOSAL

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Abstract - This paper summarizes the problem specification and results for the Phase VII Benchmark—Study of Spent Fuel Compositions for Long-Term Disposal (Ref. 1) organized by the Organisation for Co-operation and Development Nuclear Energy Agency Expert Group on Burn-up Credit Criticality Safety. The Phase VII benchmark was developed to study the ability of computer codes and associated nuclear data to predict spent fuel isotopic compositions and corresponding k_{eff} values in a cask configuration over the time duration relevant to spent nuclear fuel disposal. The benchmark was divided into two sets of calculations: (1) decay calculations out to 1,000,000 years for provided pressurized-water-reactor UO_2 discharged fuel compositions and (2) burnup credit criticality calculations for a representative cask model at selected time steps. Contributions from 15 organizations in 10 countries were submitted to the Phase VII benchmark exercise. This paper provides a description of the Phase VII benchmark and detailed comparisons of the participants' isotopic compositions and k_{eff} values that were calculated with a diverse set of computer codes and nuclear data libraries. Differences observed in the calculated time-dependent nuclide densities are attributed to different decay data or code-specific numerical approximations. The variability of the k_{eff} results is consistent with the evaluated uncertainty associated with cross-section data.

I. INTRODUCTION

This paper describes the results for Phase VII benchmark entitled "Study of Spent Fuel Compositions for Long-Term Disposal" organized by the Organisation for Co-operation and Development (OECD) Nuclear Energy Agency (NEA) Expert Group on Burn-up Credit Criticality Safety. The purpose of the Phase VII benchmark was to study the ability of relevant computer codes and associated nuclear data to predict spent nuclear fuel (SNF) isotopic compositions and corresponding k_{eff} values in a spent fuel cask configuration over the time duration relevant to SNF disposal. The expected outcome of the benchmark exercise includes an improved understanding relative to potential differences in international nuclear data sets and better understanding and/or confidence in the ability to predict k_{eff} and source terms for time frames relevant to long-term disposal of SNF, as well as for extended long-term storage of SNF.

I.A. Benchmark Description

The Phase VII benchmark specified: (1) the discharged fuel composition of a representative pressurized-water-reactor (PWR) UO_2 fuel assembly of 4.5-wt% initial enrichment and 50 GWd/MTU burnup including actinide and fission product nuclides relevant to burnup credit and radiological dose assessment as well as their precursors (113 total nuclides) for use as the initial fuel composition in decay calculations; (2) the geometry characteristics and structural material compositions of a representative spent fuel cask loaded with 21 PWR UO_2 17×17 fuel assemblies for use in criticality calculations. The cask model assumed intact fuel assemblies. The benchmark requested calculations of decayed fuel compositions and k_{eff} values for 30 post-irradiation time steps, out to 1,000,000 years.

Forty-four light element, fission product, and actinide nuclides were selected for comparison based on their relevant contributions to radiation dose from nuclear waste repositories. The set of nuclides relevant to radiological dose assessment included: ^{14}C, ^{36}Cl, ^{41}Ca, ^{59}Ni, ^{79}Se, ^{93}Zr, ^{90}Sr, ^{93m}Nb, ^{94}Nb, ^{93}Mo, ^{99}Tc, ^{107}Pd, ^{126}Sn, ^{126}Sb, ^{126m}Sb, ^{129}I, ^{135}Cs, ^{137}Cs, ^{151}Sm, ^{210}Pb, ^{226}Ra, ^{228}Ra, ^{227}Ac, ^{229}Th, ^{230}Th, ^{232}Th, ^{231}Pa, ^{232}U, ^{233}U, ^{234}U, ^{235}U, ^{236}U, ^{238}U, ^{237}Np, ^{238}Pu, ^{239}Pu, ^{240}Pu, ^{241}Pu, ^{242}Pu, ^{241}Am, ^{242m}Am, ^{243}Am, ^{245}Cm, and ^{246}Cm. Two different sets of nuclides relevant to burnup credit criticality safety analyses consisting of either actinides or actinides and fission product nuclides, which are identified in Table I as

Set 1 and Set 2, respectively, were specified for use in criticality calculations.

Table I. Nuclide Sets for Criticality Calculations

Set 1: Actinide-only burnup-credit nuclides (11 total)
^{233}U, ^{234}U, ^{235}U, ^{236}U, ^{238}U, ^{238}Pu, ^{239}Pu, ^{240}Pu, ^{241}Pu, ^{242}Pu, and ^{241}Am
Set 2: Actinide + fission product burnup-credit nuclides (30 total)
^{233}U, ^{234}U, ^{235}U, ^{236}U, ^{238}U, ^{237}Np, ^{238}Pu, ^{239}Pu, ^{240}Pu, ^{241}Pu, ^{242}Pu, ^{241}Am, ^{242m}Am, ^{243}Am, ^{95}Mo, ^{99}Tc, ^{101}Ru, ^{103}Rh, ^{109}Ag, ^{133}Cs, ^{143}Nd, ^{145}Nd, ^{147}Sm, ^{149}Sm, ^{150}Sm, ^{151}Sm, ^{152}Sm, ^{151}Eu, ^{153}Eu, and ^{155}Gd

I.B. Participating Codes and Nuclear Data Libraries

Contributions from 15 organizations/companies in 10 countries were submitted to the Phase VII benchmark exercise. The participating decay codes included ACAB (Ref. 2), CINDER 90 (Ref. 3), DARWIN 2.0 and 2.1 (Ref. 4), ORIGEN-S in SCALE 6.1 (beta), SCALE 5.1, and SCALE 6.0 (Ref. 5), ORIGEN 2.2-UPJ (Ref. 6), ORIGEN-X-2008 (Ref. 7), PHOENIX 1.0.0a (beta) (Ref. 8) and TIBSO (Ref. 9). The primary sources of the associated decay data were the United States Evaluated Nuclear Data File (ENDF)/B-VII.0, ENDF/B-VI.8, NEA Joint Evaluated Fission and Fusion File (JEFF)-3.1, European Fusion File (JEF)-2, Evaluated Nuclear Structure Data File (ENSDF), Japan Evaluated Nuclear Data Library (JENDL)-3.3, and the ORIGEN 2.2 decay library. In addition to the variety of decay data libraries, the participating decay codes used different methods for solving decay chain equations including the matrix exponential method (e.g., ORIGEN-based decay codes such as ORIGEN-S, ORIGEN 2.2-UPJ, ORIGEN-X-2008, and ACAB), fourth-order Runge-Kutta numerical method (e.g., PHOENIX), and analytical method (e.g., CINDER 90).

The criticality codes used by the participants included the Monte Carlo codes KENO-V.a in SCALE 6.1 (beta) and SCALE 6.0, KENO-VI in SCALE 5.1, MCNP5 (Ref. 10), MCNPX 2-4 and 2-5 (Ref. 10), MORET 4.B.2 in CRISTAL V1.0 (Ref. 11), and MORET 5 (Ref. 12) with either multi-group or continuous-energy cross-section data based on ENDF/B-V, -VI, and -VII, JEF-2.2, and JEFF-3.1.1.

II. RESULTS AND DISCUSSION

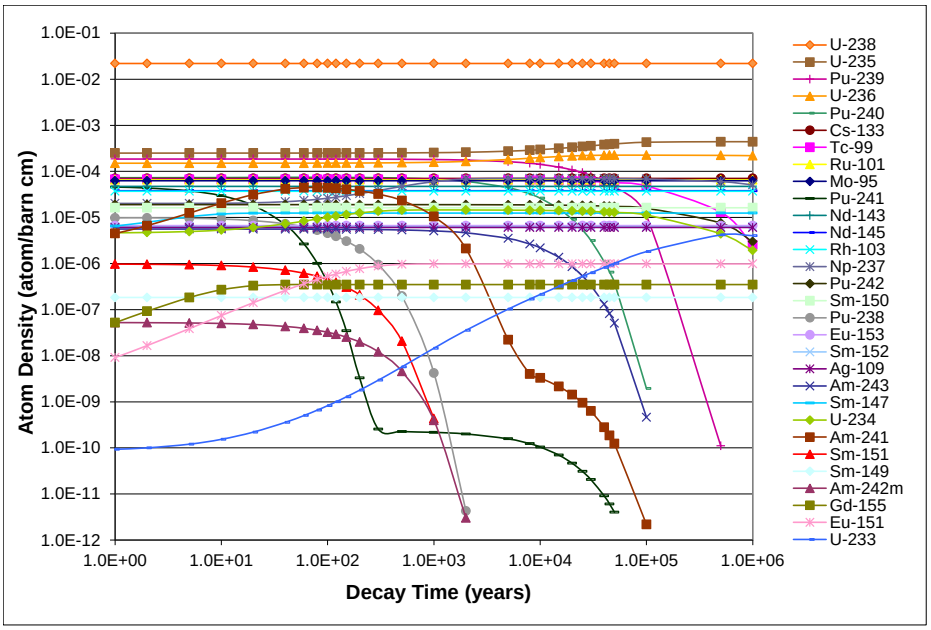
This section describes the contributed results of the decay and criticality calculations and identifies the impact of different nuclear data libraries and numerical solvers on the results. The standard deviation of the calculated values and the plots of the relative difference between the calculated and the mean values as a function of decay time were useful in identifying differences in nuclear data and code-specific numerical approximations.

II.A. Benchmark Decay Calculations

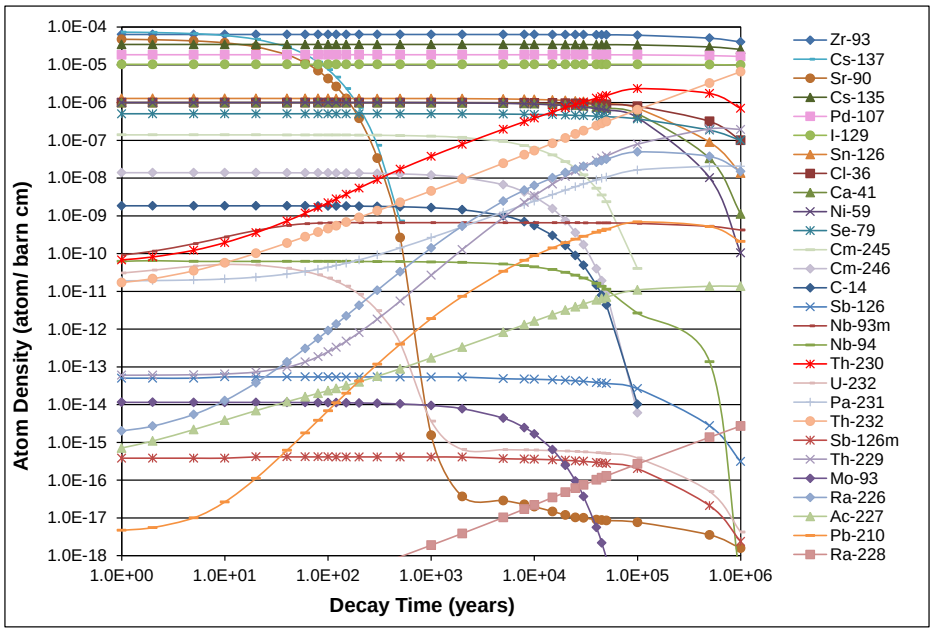
The mean values of the calculated atom densities are represented as a function of time in Fig. 1. The burnup credit nuclides are illustrated in Fig. 1 (a). Additional nuclides relevant to radiation dose are illustrated in Fig. 1(b). As seen in the figure, the fuel concentrations for nuclides ^{90}Sr, ^{137}Cs, ^{151}Sm, ^{238}Pu, ^{241}Pu, and ^{242m}Am decrease to insignificant levels after approximately 1,000 years; the fuel concentrations for nuclides ^{151}Eu, ^{210}Pb, ^{226}Ra, ^{228}Ra, ^{227}Ac, ^{229}Th, ^{230}Th, ^{232}Th, ^{231}Pa, ^{233}U, ^{235}U, and ^{236}U increase with decay time.

The spread of the decay calculation results varied considerably depending on the nuclide. For nuclides such as ^{235}U, ^{236}U, ^{238}U, and ^{239}Pu, the decay calculation results were practically identical. The greatest variability of the decay calculation results was obtained for ^{126}Sn, ^{126}Sb, and ^{126m}Sb. Figure 2 (a)-(f) shows the atom density relative difference from the mean value as a function of decay time for ^{79}Se, ^{90}Sr, ^{93}Mo, ^{126}Sn, ^{151}Sm, and ^{242m}Am, respectively. A review of the ENDF/B-VII.0, JEFF-3.1, ENDF/B-VI.8, and JEF-2 decay data (Refs. 13 and 14) identified a series of nuclides for which significant differences exist among the decay data evaluations associated with these nuclides. The participating decay codes, with the exception of PHOENIX and ORIGEN 2.2-UPJ, used one of those libraries. PHOENIX used the ORIGEN 2.2 decay library, the origins of which are not readily traceable. However, most of the decay data in ORIGEN 2.2-UPJ have been taken from ENSDF, which has been used as the basis for the ENDF/B evaluations, and from the JENDL-3.3 Fission Product (FP) Decay Data File.

For many nuclides, correlations may be established between the half-life and/or branching ratio values associated with the ENDF/B-VII.0, JEFF-3.1, ENDF/B-VI.8, and JEF-2 decay libraries and the observed trends of nuclide concentration values relative to the mean values. There is a significant difference between the recently evaluated ^{126}Sn half-life value of 2.30×10^5 years (ENDF/B-VII.0 and JEFF-3.1) and the previously evaluated ^{126}Sn half-life value of 9.99×10^4 years (ENDF/B-VI.8, JEF-2, and JENDL-3.3). The uncertainty associated with the ENDF/B-VII.0 and JEFF-3.1 half-life values for ^{126}Sn is ~ 6%; the older evaluations did not provide an uncertainty value for the ^{126}Sn half-life. This significant difference in the ^{126}Sn half-life values affected the calculated decay concentrations for ^{126}Sn and its short-lived daughter nuclides ^{126m}Sb and ^{126}Sb. Another nuclide significantly impacted by different half-life values was ^{79}Se.

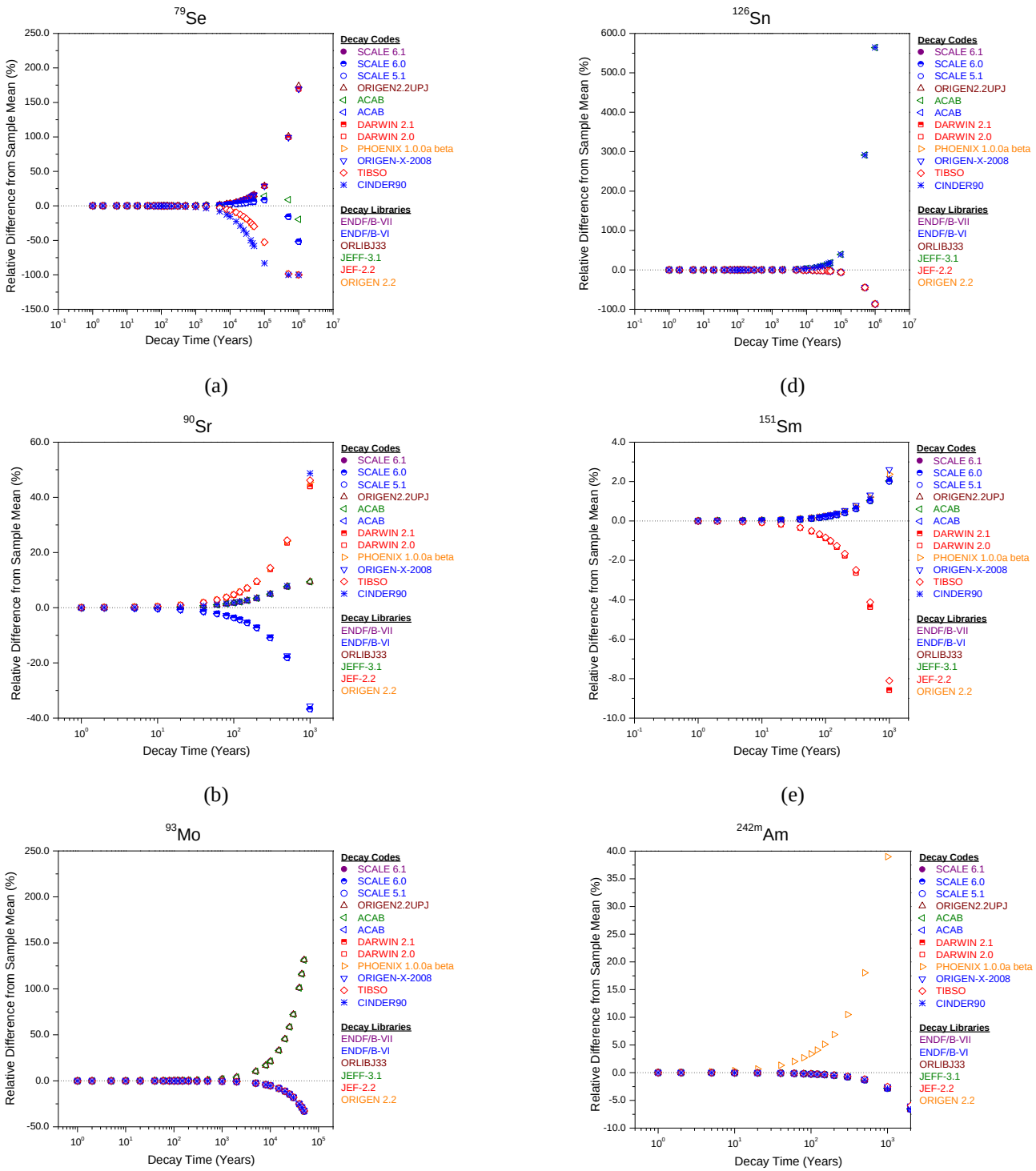


(a)



(b)

Fig. 1. Atom density as a function of time for nuclides relevant to burnup credit criticality safety analyses and radiological dose assessment.



(a) (d)

(b) (e)

(c) (f)

Note: Primary decay data source is indicated in the figure for a decay code using multiple sources of decay data.

Fig. 2. Relative difference from sample mean as a function of decay time for (a) ^{79}Se, (b) ^{90}Sr, (c) ^{93}Mo, (d) ^{126}Sn, (e) ^{151}Sm, and (f) ^{242m}Am atom densities.

The half-life values for ^{79}Se in ENDF/B-VII.0, JEFF-3.1, ENDF/B-VI.8, and JEF-2.2 are 2.95×10^5, 3.77×10^5, 3.30×10^4, and 6.50×10^4 years, respectively. The branching ratio values for the beta decay transition of ^{93}Zr to ^{93m}Nb in ENDF/B-VII.0, ENDF/B-VI.8, JEFF-3.1, and JENDL-3.3 libraries, which are 0.925, 1.0, 0.925, and 0.95, respectively, affected the calculated ^{93m}Nb densities. Nuclides mostly impacted by different half-life values associated with the ENDF/B-VII.0, JEFF-3.1, ENDF/B-VI.8, and JEF-2 libraries are summarized in Table II.

TABLE II. Half-life Values in ENDF/B-VII.0, JEFF-3.1, ENDF/B-VI.8, and JEF-2 for nuclides with largest differences

Nuclide	ENDF/B-VII.0 Half-life (s)	JEFF-3.1 Half-life (s)	ENDF/B-VI.8 Half-life (s)	JEF-2.2 Half-life (s)
^{14}C	1.7988E+11	1.7987E+11	1.8082E+11	1.8082E+11
^{41}Ca	3.2189E+12	3.2504E+12	3.2504E+12	3.2504E+12
^{59}Ni	2.3984E+12	2.3984E+12	2.3668E+12	2.3636E+12
^{79}Se	9.3095E+12	1.1897E+13	1.0414E+12	2.0512E+12
^{90}Sr	9.0854E+08	9.0852E+08	8.8833E+08	9.1894E+08
^{94}Nb	6.4062E+11	6.3072E+11	6.4061E+11	6.4061E+11
^{93}Mo	1.2623E+11	1.2623E+11	1.1045E+11	1.1045E+11
^{99}Tc	6.6618E+12	6.7532E+12	6.6617E+12	6.7216E+12
^{126}Sn	7.2582E+12	7.2581E+12	3.1557E+12	3.1557E+12
^{151}Sm	2.8402E+09	2.8402E+09	2.8401E+09	2.8000E+09
^{155}Eu	1.4999E+08	1.4999E+08	1.4769E+08	1.5652E+08
^{229}Th	2.3163E+11	2.3163E+11	2.4867E+11	2.3163E+11
^{232}U	2.1743E+09	2.2027E+09	2.2027+09	2.2027E+09
^{236}Np	4.8599E+12	4.7968E+12	3.6291E+12	4.7967E+12
^{236}Pu	9.0192E+07	9.0192E+07	9.1515E+07	9.1515E+07
^{246}Cm	1.5021E+11	1.4927E+11	1.4926E+11	1.4926E+11

Differences observed between the atom densities calculated with PHOENIX and the atom densities calculated with the other decay codes for ^{59}Ni, ^{41}Ca, ^{93m}Nb, ^{228}Ra, ^{238}Pu, ^{239}Pu, ^{240}Pu, ^{242}Pu, and ^{242m}Am may be attributed to the differences between the decay data in the ORIGEN 2.2 library and more recent decay data evaluations.

ORIGEN-X-2008 atom densities for nuclides such as ^{226}Ra, ^{229}Th, ^{237}Np, ^{232}U, and ^{233}U show small deviations from the atom density values calculated with the other decay codes due to a limited number of decay steps used by the code that impact the accuracy with which contributions from short-lived parents are calculated. ORIGEN-X-2008 has been dedicated to activation calculations and only recently tested for long time decay. The version of ORIGEN-X-2008 used supported 10 decay steps, so several runs had to be performed to get the results for all time steps in the benchmark.

Differences between the TIBSO atom densities and the other calculated atom densities have been observed for a series of nuclides including ^{94}Nb, ^{210}Pb, ^{226}Ra, ^{228}Ra, ^{230}Th, ^{234}U, ^{241}Pu, and ^{241}Am.

The ^{210}Pb atom densities calculated with CINDER 90 were significantly lower than the ^{210}Pb atom densities calculated using the other decay codes.

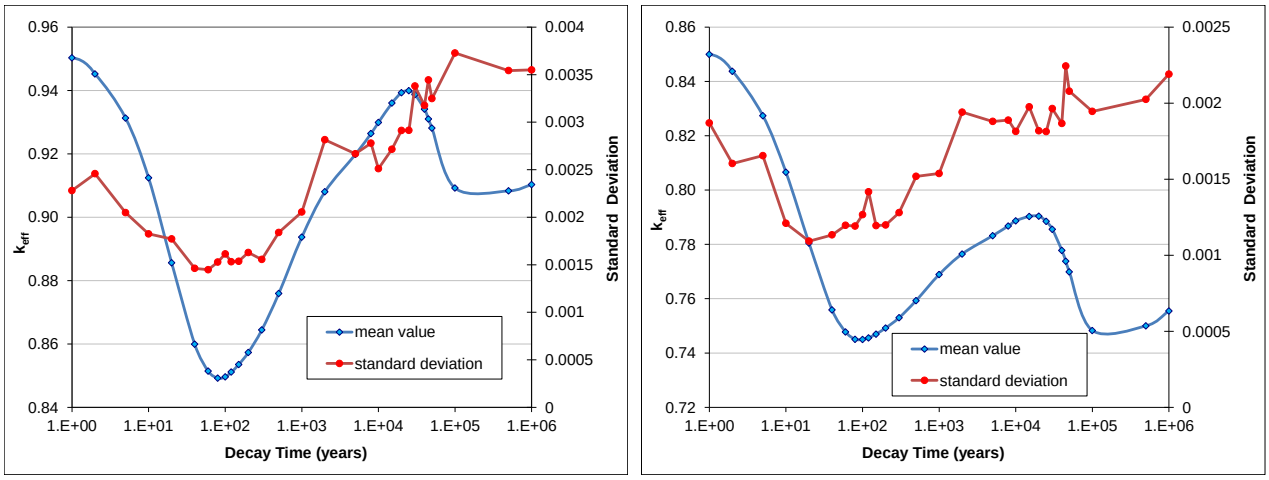
Code-specific approximations affecting rigorous representations of relevant precursor decay chains and numerical approximations made by participants would also contribute to differences in the calculated time-dependent nuclide concentrations.

II.B. Benchmark Criticality Calculations

The mean and standard deviation values of the participants' k_{eff} results for a representative spent fuel cask are represented as a function of decay time in Fig. 3 (a) and (b) for actinide-only fuel compositions and for actinide and fission product fuel compositions, respectively. The average value and the standard deviation of the k_{eff} results for fresh fuel were 1.1486 and 0.0026, respectively. As seen in Fig. 3, k_{eff} values continually decrease until approximately 100 years after fuel discharge, then increase with increasing time reaching a peak value at approximately 25,000 years after fuel discharge. The decrease in k_{eff} for the first 100 years after fuel discharge is caused primarily by the decay of the fissile nuclide ^{241}Pu ($T_{1/2}$ = 14.29 years) to ^{241}Am (neutron absorber), whereas the increase in fuel reactivity after this time is caused by the decay of neutron absorber nuclides ^{238}Pu ($T_{1/2}$ = 87.7 years), ^{240}Pu ($T_{1/2}$ = 6,561 years), ^{241}Am ($T_{1/2}$ = 432.6 years), and ^{243}Am ($T_{1/2}$ = 7,370 years) [see Fig. 1 (a)].

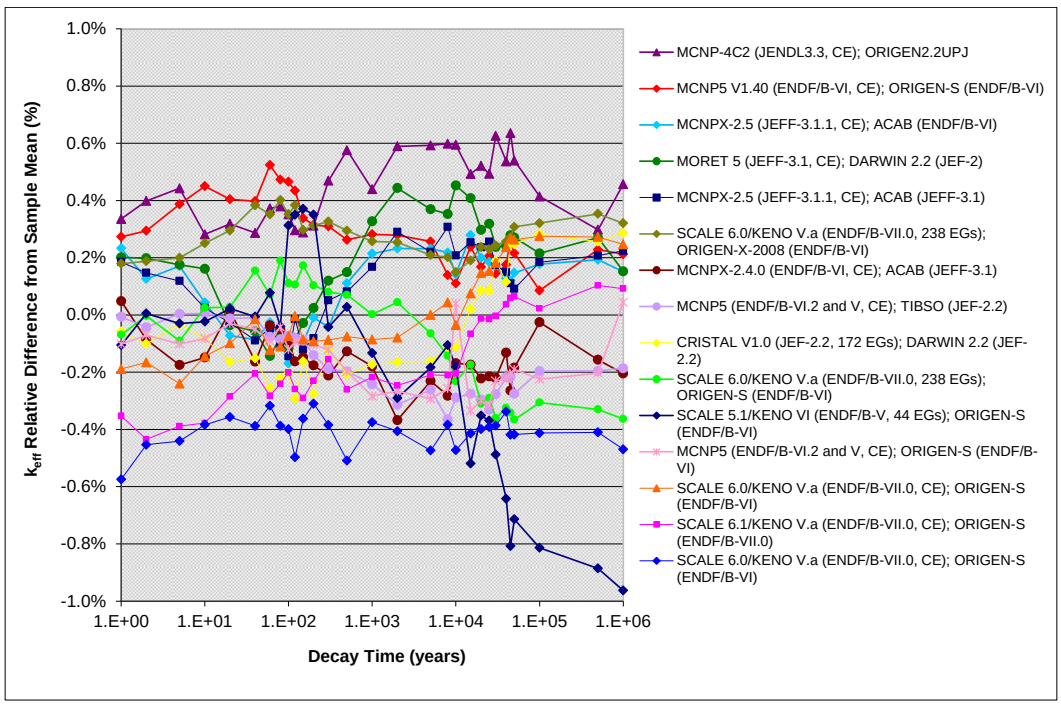
The dispersion of the participants' k_{eff} results varies depending on the decay time, as can be seen in Fig. 4 for the actinide and fission product fuel compositions. Given that insignificant differences in the calculated concentrations for the burnup credit nuclides were obtained, the observed variability in the k_{eff} results may be attributed to the criticality codes and cross-section data as well as to possible modeling approximations made by the participants.

To evaluate the impact of cross-section data uncertainties on k_{eff}, a cross-section data sensitivity and uncertainty (S/U) analysis was performed using the TSUNAMI tools in SCALE (Ref. 5). The S/U calculations used the SCALE 238 energy-group cross section library based on ENDF/B-VII.0 and SCALE 44-group covariance library based on a variety of sources including evaluations from ENDF/B-VII, ENDF/B-VI, and JENDL. Energy and region integrated k_{eff} sensitivities to nuclide total and reaction-specific cross sections and the relative uncertainty in k_{eff} due to cross-section covariance data have been calculated using TSUNAMI. The sensitivity of k_{eff} to a particular nuclide-reaction pair macroscopic cross section, referred to as the sensitivity coefficient, provides a measure of the first-order effect of perturbations in the nuclear reaction for the nuclide on k_{eff}.



(a) (b)

Fig. 3. k_{eff} mean and standard deviation values as a function of decay time for fuel compositions consisting of burnup-credit (a) actinide-only nuclides and (b) actinide and fission product nuclides.



Note: The legend indicates: criticality code (nuclear data) and decay code (decay library) used to calculate time-dependent fuel compositions for criticality calculations. EGs = energy groups. CE = continuous energy.

Fig. 4. Comparison of the k_{eff} results relative to the mean value for fuel compositions consisting of burnup-credit actinide and fission product nuclides.

The sensitivity coefficients for the actinide and fission product fuel compositions are illustrated in Fig. 5 as a function of decay time. As seen in the figure, k_{eff} has large sensitivities to ^{239}Pu, ^{235}U, and ^{238}U total cross sections. Therefore, uncertainties associated with the cross-section data for those nuclides will have a dominating effect on the uncertainty in k_{eff}. The values of the uncertainty in k_{eff} due to cross-section covariance data obtained directly from TSUNAMI calculations are represented as a function of decay time in Fig. 6 for the actinide and fission product fuel compositions. The uncertainty in k_{eff} due to cross-section covariance data varies from 0.0057 (1,000 years) to 0.0036 (100,000 years). Based on these values, it can be concluded that the variability of the k_{eff} results (see Figs. 3 and 4) is consistent with the uncertainty in k_{eff} due to cross-section covariance data.

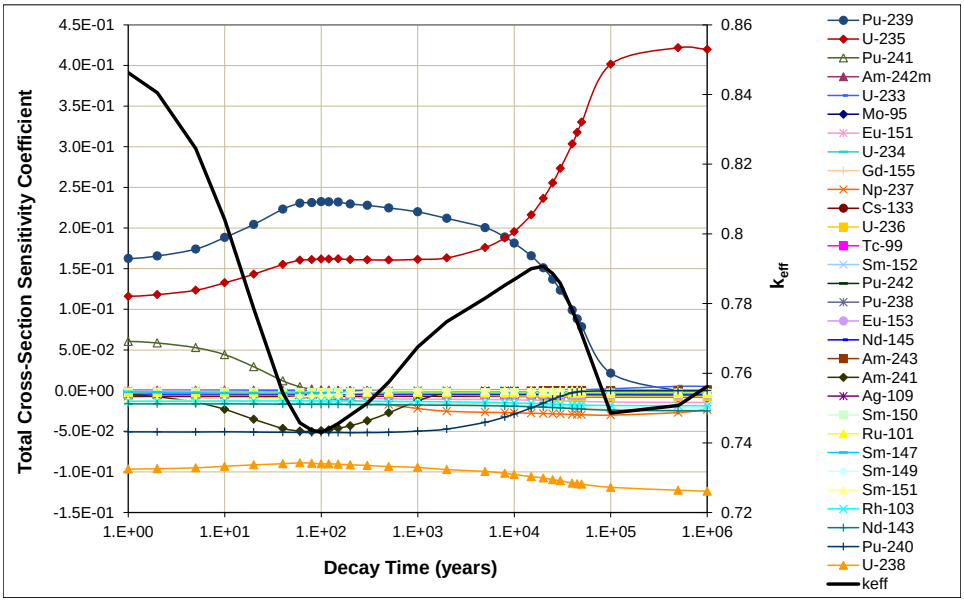
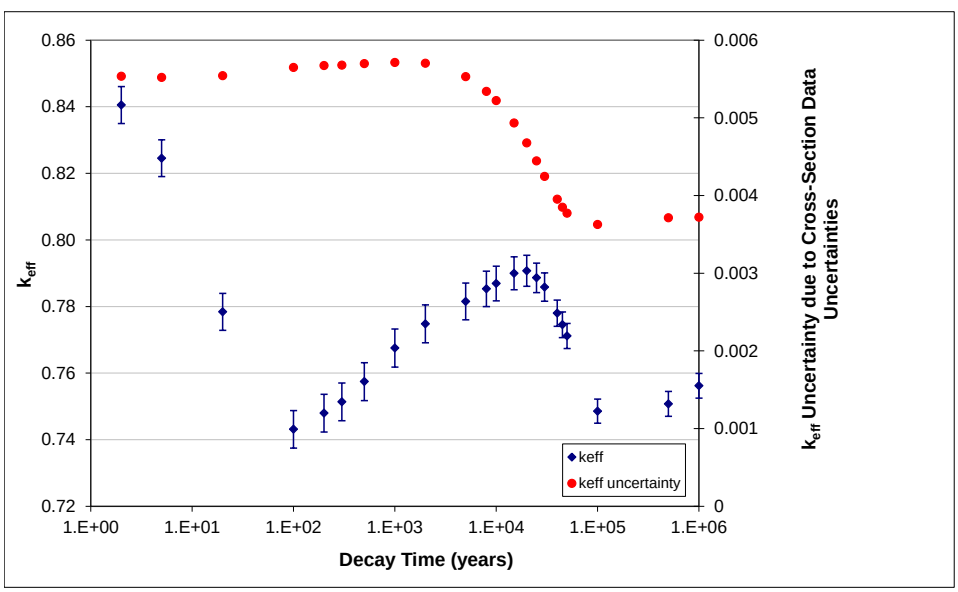


Fig. 5. k_{eff} sensitivity to nuclide total cross section as a function of decay time for fuel compositions consisting of burnup credit actinide and fission product nuclides.



Note: Error bars show the uncertainty in k_{eff} due to uncertainties associated with cross-section data.

Fig. 6. k_{eff} uncertainty due to cross-section covariance data as a function of decay time for fuel compositions consisting of burnup credit actinide and fission product nuclides.

II. CONCLUSIONS

Contributions from 15 organizations in 10 countries were submitted to the Phase VII benchmark exercise, the purpose of which was to evaluate the ability of decay and criticality codes to predict SNF nuclide concentrations and corresponding k_{eff} values in a representative cask over the time duration relevant to SNF disposal. The results obtained by the participants using a variety of codes and nuclear data have been evaluated. Differences observed in the calculated atom densities for nuclides relevant to burnup credit criticality analyses and radiological dose assessments may be attributed to (1) different decay data, (2) the ability of the decay codes to consider relevant precursor decay chains contributing to the formation of a nuclide of interest, and (3) approximations related to the number of time steps allowed by the code. A review of the ENDF/B-VII.0, JEFF-3.1, ENDF/B-VI.8, and JEF-2 decay data identified significant differences among these data evaluations for a series of nuclides (e.g., half-life values of ^{79}Se, ^{90}Sr, and ^{126}Sn and branching ratio values for ^{93}Zr). The observed variability of the k_{eff} results is consistent with an evaluated uncertainty in k_{eff} due to cross-section covariance data and may be predominantly attributed to the criticality calculation methods and cross-section data.

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