EFFECTS OF SPENT FUEL TYPES ON OFFSITE CONSEQUENCES OF HYPOTHETICAL ACCIDENTS[1]

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ABSTRACT

Argonne National Laboratory (ANL) conducts experimental work on the development of waste forms suitable for several types of spent fuel at its facility on the Idaho National Engineering and Environmental Laboratory (INEEL) located 48 km West of Idaho Falls, ID. The objective of this paper is to compare the offsite radiological consequences of hypothetical accidents involving the various types of spent nuclear fuel handled in nonreactor nuclear facilities. The highest offsite total effective dose equivalents (TEDEs) are estimated at a receptor located about 5 km SSE of ANL facilities. Criticality safety considerations limit the amount of enriched uranium and plutonium that could be at risk in any given scenario. Heat generated by decay of fission products and actinides does not limit the masses of spent fuel within any given operation because the minimum time elapsed since fissions occurred in any form is at least five years. At cooling times of this magnitude, fewer than ten radionuclides account for 99% of the projected TEDE at offsite receptors for any credible accident. Elimination of all but the most important nuclides allows rapid assessments of offsite doses with little loss of accuracy. Since the ARF (airborne release fraction), RF (respirable fraction), LPF (leak path fraction) and atmospheric dilution factor (χ/Q) can vary by orders of magnitude, it is not productive to consider nuclides that contribute less than a few percent of the total dose. Therefore, only ^{134}Cs, ^{137}Cs-^{137m}Ba, and the actinides significantly influence the offsite radiological consequences of severe accidents. Even using highly conservative assumptions in estimating radiological consequences, they remain well below current Department of Energy guidelines for highly unlikely accidents.

DESCRIPTION OF FUEL TYPES

There are three generic types of fuel handled within ANL facilities. Two are metallic fuels designed for the Experimental Breeder Reactor II (EBR-II); the other is slightly enriched uranium dioxide (UO_2) for light water cooled and moderated power reactors (LWRs). One metallic fuel is called binary because it contained 90% uranium and 10% zirconium before irradiation. The other metallic fuel, ternary, typically contains about 23% plutonium and 10% zirconium; the remaining 67% is uranium. Uranium enrichments up to 67% by weight are found in both binary and ternary fuels. Unirradiated binary fuel assemblies contain up to 4.45 kg of uranium; fresh ternary assemblies contain up to 4.48 kg of actinides. Enrichments up to 4.5% are found in fresh UO_2 fuel. Both unirradiated and spent fuel are routinely handled in ANL facilities at the INEEL; this paper considers only irradiated fuels.

Reference 1 describes the version of the ORIGEN program used to find the radionuclide content of the spent fuel as a function of cooling times between 5 and 30 years. No metallic fuel with

shorter cooling times are available because the EBR-II was shutdown in October 1994; spent fuel assemblies from LWRs are expected to remain at power plants for at least five years before they would be shipped offsite to ANL. Calculations for binary and ternary fuels used actinide cross sections developed for metallic fuels irradiated in EBR-II; cross sections for LWR fuels were unmodified from those in the ORIGEN program data package described in Reference 1.

Since the actual uranium enrichment in any type and the amount of plutonium present in ternary fuels can vary, ORIGEN calculations are performed for maximum enrichment expected in any fuel form. Enrichments up to 67% are found in the metallic fuels; an enrichment of 4.25% is selected for the oxide fuel for LWRs. In the ternary fuel, plutonium content of 23.4% is chosen as a reasonable upper bound of the fuel weight. While fission product yields and transuranic element contents in spent fuels vary with the actinide isotope content of fresh fuel, we believe that our selected parameters are adequate for radiological consequence assessment considering the degree of accuracy of the calculations. Table I shows the actinide masses that were input to the ORIGEN calculations for each of the unirradiated fuel assemblies.

Table I
Masses of Actinides in Unirradiated Fuel Assemblies

Nuclide	Binary Fuel (g)	Ternary Fuel (g)	LWR Fuel (g)
U-234	30.4	0	292.9
U-235	2,980	2,300	42,500
U-236	18.2	0	0
U-238	1,420	1,130	957,500
Pu-239	0	836	0
Pu-240	0	188	0
Pu-241	0	15.7	0
Pu-242	0	5.32	0
Total Masses (g)	4,449	4,475	1,000,293

To assure conservative results, very high burnups are assumed. For the LWR fuel, a value of 50,000 Megawatt-days per metric ton (MW-d/T) is selected. This level is being reached in the late 1990s for LWRs operating in the United States. Most of the spent fuel that will be available to ANL in the next few years has burnups of less than 40,000 MW-d/T. Both binary and ternary fuel assemblies are assumed to produce 592 MW-d in somewhat less than 4.5 kg of heavy metal for a burnup of about 133,000 MW-d/T; only a few assemblies have exceeded this value in EBR-II. All of the fission energy is assumed to be produced in a single irradiation period at constant power rather than over a longer time that includes several cycles of shutdown and power production at varying levels. Activities found by ORIGEN for each fuel assembly were scaled up

or down to those expected in 20 kg of unirradiated heavy metal to make the comparisons shown in this paper.

HYPOTHETICAL ACCIDENTS

Any series of events that produce offsite consequences must be severe enough to compromise all of the passive barriers to release and to impart enough kinetic energy to the radioactive materials to cause them to remain airborne for an extended period of time within the facility. Because of the cooling times, the only radioactive gases are ^{3}H and ^{85}Kr; all other radionuclides are solids. Four volatile fission product solids might contribute to the TEDE at offsite receptors; they are ^{113m}Cd, ^{129}I, ^{134}Cs, and ^{137}Cs (with its short-lived progeny, ^{137m}Ba). The other nuclides, mainly actinide isotopes, are more refractory solids. To be a candidate for release, a given radionuclide must escape one or more barriers to enter a volume served by the building ventilation system. Airborne materials are moved through a filtration system, that might be damaged, into the atmosphere by the building exhaust fans. If the exhaust system does not operate, materials could escape through expansion of the building atmosphere if there is a significant release of energy. Actions by emergency response personnel could cause small amounts of radioactive materials to be released from the facility into the atmosphere unless contamination control procedures are implemented.

The generic event most likely to cause radionuclides to escape confinement is a fire in metallic fuel. Usually, such materials are maintained in an inert atmosphere, such as argon gas, but limited amounts of fuel may be transferred in approved containers between units of equipment through normally occupied areas. Since the LWR fuel is already oxidized, it can not burn. However, a nearby fire could cause the air surrounding the UO_2 to expand and entrain fine particles for enough time to enter the building exhaust system. Irradiated fuels are maintained within shielded volumes such as hot cells and massive shipping casks. To have offsite consequences, events that involve mechanical energy (rather than thermal energy) must be severe enough to extensively damage all of the confinement barriers and impart enough kinetic energy to the radioactive materials to cause them to become airborne.

RADIOLOGICAL CONSEQUENCE ANALYSIS

Radiological consequences of accidents involving the types of materials handled by ANL personnel are dominated by the committed effective dose equivalent (CEDE) from the inhalation of radionuclides, particularly cesium and the actinide elements. While the CEDE is defined as the dose received over a 50 year period following an acute intake, convention assumes that the dose is imparted immediately. In accident analysis, CEDEs from ingestion are ignored because they would be expected to be much less than those from inhalation of effluent without respiratory protection. The TEDE is defined as the CEDE plus the deep dose equivalent (DDE) from radionuclides that remain outside the body. The DDE is imparted by photons that can deposit their energy at a one centimeter depth into the human body. Another parameter of interest is the shallow dose equivalent (SDE); it does not contribute to the TEDE. The SDE was formerly

called the skin dose; it is dominated by beta particles and low energy photons. For incidents involving the materials handled at ANL, both DDE and SDE values are dwarfed by the CEDE from inhalation.

For any credible event, the amount of material at risk (MAR) is defined, usually in units of activity, Curies (Ci) or Becquerels (Bq). Considering the physical form of the radionuclide and the hypothetical sequence of events, an appropriate airborne release fraction (ARF) is selected. While the ARF is function of time, an integrated value of this unitless parameter is selected using guidance found in References 2 and 3. To be transported through the atmosphere to offsite receptors, solids must be in the form of fine particles. To contribute to the CEDE from inhalation, the particles must be small enough to enter the pulmonary region of the human lung. By convention, a nominal size of 10μm is used as an upper limit to the size of respirable particles. A distribution of particle sizes would be formed in actual events; the parameter, respirable fraction (RF), is used to specify that fraction of the airborne particles below 10μm. Appropriate values for the RF for a variety of events are suggested in References 2, 3, and 4.

Not all of the radioactive particles that become airborne within a facility can escape into the environment. Passive effects, such as gravity induced fallout and chemical reactions with surfaces and active systems, such as filter trains, can remove a significant fraction of airborne particles before they reach the atmosphere. To assure conservative estimates of TEDE values, some degree of degradation of the efficiencies of active systems is assumed. The ratio of activity of a given nuclide escaping a facility to the activity of the same nuclide is called the leak path fraction (LPF). For ^{85}Kr, tritium gas ($^{3}H_2$ or T_2), and tritium oxide vapor (HTO or T_2O), the LPF is usually assumed to be at its maximum value, unity. Guidance for the LPF for fine particles for exhaust systems containing high efficiency particulate - air (HEPA) filters under accident conditions is found in Reference 5. As with the ARF and RF, a review of the detailed scenario of the abnormal event helps the person responsible for predicting radiological consequences to select an appropriate value for the LPF. Multiplication of the MAR by the product of the ARF, RF, and LPF gives the activity of each radionuclide that is released from the facility into the atmosphere. A recently issued standard, Reference 4, recognizes the validity of engineering judgement in the selection of specific numerical values for all of the factors needed to estimate the kinds and quantities of radioactive materials released from the facility in hypothetical accidents.

Typically, offsite receptors are positioned on the nearest site boundary and at locations where individuals have established temporary or permanent residences. Usually radiological consequence analyses of accidents consider the TEDEs and SDEs imparted to individuals, rather than collective values that might be received by the surrounding population. For releases at or near the ground level, the maximum downwind concentrations of airborne effluents are found at the locations nearest the point of release. For elevated releases, maximum concentrations can occur at more distant receptors. Most accident scenarios for ANL facilities postulate near ground level releases, so the highest offsite concentrations are found at the nearest site boundary, 5 km SSE of the release point. Within the accuracy of the calculations, it is not worthwhile to consider the small variation in distances between the ANL facilities that handle significant amounts of radioactive materials and any offsite receptors.

Dilution of released effluent depends on the distance to the receptor of interest, the wind speed, and the stability of the atmosphere. The atmospheric dilution factor, χ/Q, in seconds per cubic meter is developed for low wind speeds, and very stable atmospheric conditions. These conditions are assumed to prevail for the duration of the release and transport of the effluent from the facility to the receptor. More realistic assumptions on meteorological conditions lead to concentrations of airborne materials one to three orders of magnitude less than those used to project offsite doses. For the nearest receptor located 5 km from the release point, the χ/Q is estimated to equal 4×10^{-5} s/m^3. Multiplication of the activity released into the atmosphere and the χ/Q yields the time integrated concentration in Ci-s/m^3 for each nuclide of interest. Conversion factors found in Reference 6 convert the integrated airborne concentrations for each nuclide to the DDE and SDE in units of rem. Unless only the inert gas, ^{85}Kr, is released, both the DDE and SDE are negligible compared to the inhalation CEDE for unirradiated fuel and in spent fuel cooled for five or more years.

Multiplication of the integrated concentration in Ci/s-m^3 by a breathing rate in m^3/s yields the inhaled activity. There are several sources of conversion factors that convert the inhaled activity in Ci to CEDE in rem for each nuclide. The highest values of rem per unit activity inhaled are found in Reference 7; Reference 8 updates the conversions for the actinides. References 9 and 10 reflect some 20 years of advances in dosimetry and radiobiology; they contain conversions that are factors of two to three lower than those in References 7 and 8 for nuclides handled at ANL. Terminology has been changed in the latter two references. The CEDE of References 7 and 8 has been replaced by the committed effective dose (CDE) in References 9 and 10. A total effective dose (TED) is found by adding the DDE to the CDE; the TED is equivalent to the TEDE. References 9 and 10 express the CDE, DDE, and TED in Sieverts (Sv); these parameters are expressed in rem in References 6, 7, and 8. Because of their conservative predictions and acceptance by regulatory bodies, our studies use data from References 6 and 7 for the fission products and data from Reference 8 for the actinides. The older terminology and units are adopted in this paper to be consistent with most of the safety documentation generated in support of ANL operations.

SELECTION OF RADIONUCLIDES

Estimations of the inhalation CEDE and DDE values were performed using several spreadsheet programs. Only nuclides with activities greater than 0.02 Ci after five years of cooling are selected from the ORIGEN output to be entered into the spreadsheets. There are only 30 fission products and 15 actinides that meet this criterion in the three types of spent fuel considered in this study. Preliminary estimations of the TEDE indicated that 24 of the fission products and 8 actinides could be neglected; these 32 nuclides account for less than 0.01% of the total TEDE for any fuel type for cooling times between 5 and 30 years. Therefore, 16 nuclides, including the short-lived progeny of ^{90}Sr, ^{137}Cs, and ^{144}Ce, determine the TEDE associated with hypothetical accidents in any ANL nonreactor facility. Fires, rather than spills, drops, or other abnormal events that involve only mechanical energy, are treated in this paper because they have higher release fractions and lead to higher offsite CEDE and TEDE estimates.

Inhalation conversion factors depend on the chemical form of the inhaled nuclide; all materials should be oxides by the time they reach offsite receptors. Only tritium gas and ^{85}Kr would not be oxidized and neither contributes to the CEDE. All materials, other than tritium and ^{85}Kr, are particles that are assigned airborne release and respiratory fractions based on their volatility. Three volatile fission products, ^{113m}Cd, ^{134}Cs, and ^{137}Cs, are assigned an ARF of 0.35 based on experimental data from metallic fuels involved in fires documented in Reference 11. All other materials are assigned an ARF of 0.0005 based on recommendations in Reference 2. The RF for all particulate materials involved in fires is 0.5, the maximum value suggested by Reference 2. Data listed in Table II are used to predict the contribution of each nuclide to the CEDE at the site boundary for fires in ANL facilities.

Table II
Parameters Used To Predict The Inhalation CEDE From Fires

Nuclide	ARF * RF	Conversion (rem/μCi)	Lung Class
Sr-90	$2.50x10^{-4}$	$8.88x10^{-2}$	Day
Y-90	$2.50x10^{-4}$	$5.55x10^{-3}$	Year
Cd-113m	$1.75x10^{-1}$	$1.15x10^{-1}$	Year
Cs-134	$1.75x10^{-1}$	$2.44x10^{-2}$	Day
Cs-137 + Ba-137m	$1.75x10^{-1}$	$1.70x10^{-2}$	Day
Ce-144	$2.50x10^{-4}$	$1.96x10^{-1}$	Year
Pr-144	$2.50x10^{-4}$	$6.66x10^{-2}$	Year
Pu-238	$2.50x10^{-4}$	$2.78x10^{2}$	Year
Pu-239	$2.50x10^{-4}$	$3.00x10^{2}$	Year
Pu-240	$2.50x10^{-4}$	$3.00x10^{2}$	Year
Pu-241	$2.50x10^{-4}$	$4.80x10^{0}$	Year
Am-241	$2.50x10^{-4}$	$4.44x10^{2}$	Week
Am-243	$2.50x10^{-4}$	$4.44x10^{2}$	Week
Cm-244	$2.50x10^{-4}$	$2.37x10^{2}$	Week

An LPF of 0.5, the highest value recommended by Reference 4, is selected for each barrier; at least two barriers, local confinement (a glove box or a container) and the building surround the MAR. In more realistic scenarios, only ^{85}Kr is likely to escape into the atmosphere and reach a receptor several km away. This inert gas imparts a small DDE and somewhat higher SDE to receptors submerged in the cloud of effluent. Fourteen other radionuclides, including the short-

lived progeny of the relatively long-lived fission products ^{137}Cs and ^{144}Ce contribute to the DDE or SDE. Because of the low energy of their radiations, ^{3}H, ^{90}Sr, ^{90}Y, ^{113m}Cd, ^{137}Cs, and ^{241}Pu do not impart a DDE; except for ^{3}H and ^{241}Pu, these nuclides emit beta particles that contribute to the SDE. Conversion factors taken from Reference 6 in units of mrem / year per $\mu Ci / m^3$ are listed in Table III for the 16 nuclides considered in our safety studies.

Table III
Conversion Factors for the Deep Dose Equivalent (DDE) and Shallow Dose Equivalent (SDE)

Nuclide	DDE (mr/yr per $\mu Ci/m^3$)	SDE (mr/yr per $\mu Ci/m^3$)
Kr-85	11.2	1,580
Sr-90	0	1,110
Y-90	0	7,380
Cd-113m	0	989
Cs-134	8,010	11,700
Cs-137	0	875
Ba-137m	3,060	4,600
Ce-144	90.9	345
Pr-144	179	9,870
Pu-238	0.441	3.06
Pu-239	0.411	1.45
Pu-240	0.432	2.93
Pu-241	0	0
Am-241	95	141
Am-243	256	336
Cm-244	0.418	2.95

ESTIMATIONS OF CEDE, DDE, TEDE, AND SDE VALUES

To compare the radiological consequences of accidents involving the three types of spent fuel, a mass of 20 kg of each is selected to be the MAR at risk in a fire. This mass of MAR is based on criticality prevention limits on the metallic fuels. While up to 200 kg of UO_2 could be at risk in any given process, we have elected to compare the fuels on an equal mass basis. Data shown in Tables I, II, and III are used to obtain the CEDE, DDE, TEDE, and SDE at the nearest site boundary. In our judgement, the hypothetical fire involving 20 kg of any fuel would be a highly

unusual event based on experience with the processes, equipment, procedures, and safety systems in use at ANL in 1999. The inhalation CEDE at the nearest site boundary for the three fuels in the range of cooling times of interest are given in Table IV.

Table IV
Committed Effective Dose Equivalent For A Fire Involving 20 Kilograms Of Fuel

Cooling Time (Years)	Binary Fuel (Rem)	Ternary Fuel (Rem)	LWR Fuel (Rem)
5	0.521	1.39	0.995
10	0.448	1.37	0.788
15	0.396	1.37	0.716
20	0.355	1.36	0.670
25	0.318	1.35	0.633
30	0.278	1.34	0.602

Because of the relatively high content of long-lived transuranic nuclides, the ternary fuel gives the highest values of the CEDE for all cooling times. Decay of the cesium fission products is counteracted by ingrowth of ^{241}Am from the decay of the short-lived ^{241}Pu; as a result, the CEDE is almost constant for cooling times between five and thirty years. The CEDE drops by a factor of about two for the binary fuel over the same period because of the low content of the transuranic nuclides. The rate of decrease in the LWR fuel is between those of the two metallic fuels because the relatively high ^{238}U content produces more transuranic nuclides than the highly enriched binary fuel, but the total percentages of isotopes of plutonium and americium are less than in the ternary fuels.

For the materials of interest, the TEDE is essentially equal to the CEDE because the DDE is about three orders of magnitude less than the inhalation CEDE. Charged particles and low energy photons emitted by the actinides are not effective in delivering DDEs. Of the actinides considered in this paper, only ^{241}Am emits gamma rays that can penetrate one centimeter into the human body. For all types of fuel, the DDE decreases with increasing cooling time. Site boundary DDE values are shown in Table V; note that the DDE are given in mrem, rather than in rem.

Table V
Deep Dose Equivalent For A Fire Involving 20 Kilograms Of Fuel

Cooling Time (Years)	Binary Fuel (mRem)	Ternary Fuel (mRem)	LWR Fuel (mRem)
5	1.220	1.220	1.370
10	0.994	1.000	0.553
15	0.869	0.880	0.369
20	0.766	0.778	0.305
25	0.680	0.693	0.268
30	0.609	0.619	0.238

Only ^{85}Kr, ^{134}Cs, and the ^{137m}Ba produced by the decay of ^{137}Cs contribute to the DDE. Of these, the ^{137m}Ba usually dominates with ^{85}Kr accounting for less than 1% the total DDE. The ^{134}Cs is only significant at cooling times less than about 12 years. Contributions from individual nuclides are given in Table VI.

Table VI
Contributions Of The Nuclides To The Total DDE For Each Fuel Type

Cooling (Years)	Nuclide	Binary Fuel	Ternary Fuel	LWR Fuel
5	Kr-85	0.82 %	0.69 %	0.21 %
5	Cs-134	10.72 %	9.52 %	68.93 %
5	Ba-137m	88.46 %	89.79 %	30.86 %
10	Kr-85	0.73 %	0.61 %	0.38 %
10	Cs-134	2.44 %	2.16 %	31.67 %
10	Ba-137m	96.83 %	97.23 %	67.95 %
15	Kr-85	0.60 %	0.50 %	0.41 %
15	Cs-134	0.52 %	0.46 %	8.83 %
15	Ba-137m	98.88 %	99.04 %	90.76 %
20	Kr-85	0.50 %	0.41 %	0.35 %
20	Cs-134	0.11 %	0.10 %	2.00 %
20	Ba-137m	99.39 %	99.49 %	97.65 %
25	Kr-85	0.40 %	0.34 %	0.29 %
25	Cs-134	0.02 %	0.02 %	0.42 %
25	Ba-137m	99.58 %	99.64 %	99.29 %
30	Kr-85	0.33 %	0.27 %	0.24 %
30	Cs-134	0.00 %	0.00 %	0.09 %
30	Ba-137m	99.76 %	99.73 %	99.67 %

In a more realistic treatment of the hypothetical scenario, it is likely that only the ^{85}Kr would escape the facility because passive processes within the facility would be effective in eliminating particulate matter from the effluent. Thus, the already low DDE values would be reduced by a factor of two orders of magnitude, and any increase in the DDE rate at the site boundary would be within the statistical variation of naturally occurring radiation sources at the ANL site.

Shallow dose equivalent values at the site boundary are greater than the DDE, but they are still in the range of mrem. In contrast to the DDE, ^{85}Kr accounts for 15% to 40% of the total SDE. The isobaric pair, ^{137}Cs-^{137m}Ba, accounts for 30% to 80% of the total. For the LWR fuels, ^{134}Cs is a significant contributor at cooling times less than about 25 years; even for decay periods of less

than 10 years, its contributions are less than 11% for the metallic fuels. Other fission products are minor contributors, and the SDE from the actinides are negligible. Total SDE values are given in Table VII.

Table VII
Shallow Dose Equivalent For A Fire Involving 20 Kilograms Of Fuel

Cooling Time (Years)	Binary Fuel (mRem)	Ternary Fuel (mRem)	LWR Fuel (mRem)
5	3.59	3.39	2.560
10	2.83	2.69	1.240
15	2.33	2.24	0.878
20	1.94	1.88	0.711
25	1.64	1.60	0.602
30	1.40	1.38	0.517

As with the DDE, realistic scenarios that consider the release of only ^{85}Kr would impart SDEs that would not exceed two mrem to the receptor at the nearest site boundary. The higher SDEs for the binary fuel are caused by the greater yield of ^{85}Kr by fissions in ^{235}U than in other fissionable actinides.

Since the TEDE is of more interest to regulators than the SDE and the projections of the SDE at the site boundary are far less than the appropriate guidelines for abnormal conditions, contributions to the inhalation CEDE are the parameter of most interest in radiological consequence assessment. As shown in Tables IV and V, the DDE contributes less than one percent to the TEDE for all fuels in the cooling time range of interest. Therefore, it is the comparisons of the contributors to the inhalation CEDE that are important to safety analysts. Metallic fuels are irradiated in "fast" reactors, so the fission product yields from the fissionable nuclides exposed to higher energy neutrons influence the isotopic content of the binary and metallic fuels. All of the actinides have some fission cross section for fast neutrons. In the LWR, most of the fissions occur in ^{235}U until a significant amount of the transuranic nuclides are produced by neutron captures in ^{238}U. Subsequent decays and nonfission captures produce a mixture of nuclides that can be fissioned by fast and thermal neutrons. All of these complex reactions are treated rigorously by the ORIGEN program. Both the neutron spectra and time-dependent mixture of actinides influence the fission product distributions. Unlike the DDE and SDE, actinides strongly influence the CEDE as a function of fuel type, burnup, and cooling time. By selecting high values of burnup, the fission product and actinide contents of a given mass of spent fuel are maximized.

By combining the contributions of three isobaric fission product pairs (^{90}Sr- ^{90}Y, ^{137}Cs- ^{137m}Ba, and ^{144}Ce-^{144}Pr), only five components need to be considered in estimating inhalation CEDE

values. Seven actinides must be considered to characterize the three fuel types. Tables VIII through XIII present the relative contributions of the nuclides for each fuel for the six cooling times studied.

Table VIII
Percentage Contributions To The CEDE For Five Years Cooling Time

Nuclides	Binary Fuel	Ternary Fuel	LWR Fuel
Sr-90 + Y-90	0.65 %	0.19 %	0.10 %
Cd-113m	0.17 %	0.13 %	0.04 %
Cs-134	5.01 %	1.67 %	18.95 %
Cs-137 + Ba-137m	88.65 %	33.77 %	18.19 %
Ce-144 + Pr-144	0.29 %	0.09 %	0.04 %
Pu-238	2.91 %	1.52 %	12.37 %
Pu-239	2.29 %	20.27 %	0.096 %
Pu-240	0.01 %	19.82 %	1.46 %
Pu-241	0.01 %	9.44 %	7.34 %
Am-241	0.01 %	12.96 %	6.83 %
Am-243	0.00 %	0.02 %	0.48 %
Cm-244	0.00 %	0.11 %	33.24 %

Table IX
Percentage Contributions To The CEDE For Ten Years Cooling Time

Nuclides	Binary Fuel	Ternary Fuel	LWR Fuel
Sr-90 + Y-90	0.66 %	0.18 %	0.11 %
Cd-113m	0.16 %	0.10 %	0.04 %
Cs-134	1.08 %	0.32 %	4.44 %
Cs-137 + Ba-137m	92.13 %	30.53 %	20.43 %
Ce-144 + Pr-144	0.00 %	0.00 %	0.00 %
Pu-238	3.26 %	1.49 %	14.91 %
Pu-239	2.66 %	20.64 %	1.22 %
Pu-240	0.01 %	20.18 %	1.86 %
Pu-241	0.01 %	7.57 %	7.29 %
Am-241	0.01 %	18.90 %	14.66 %
Am-243	0.00 %	0.02 %	0.61 %
Cm-244	0.00 %	0.09 %	34.43 %

Table X
Percentage Contributions To The CEDE For FifteenYears Cooling Time

Nuclides	Binary Fuel	Ternary Fuel	LWR Fuel
Sr-90 + Y-90	0.06 %	0.16 %	0.11 %
Cd-113m	0.14 %	0.08 %	0.04 %
Cs-134	0.23 %	0.06 %	0.91 %
Cs-137 + Ba-137m	92.96 %	27.26 %	20.07 %
Ce-144 + Pr-144	0.00 %	0.00 %	0.00 %
Pu-238	3.55 %	1.44 %	15.88 %
Pu-239	3.01 %	20.63 %	1.34 %
Pu-240	0.01 %	20.18 %	2.07 %
Pu-241	0.01 %	5.98 %	6.32 %
Am-241	0.02 %	24.12 %	21.24 %
Am-243	0.00 %	0.02 %	0.67 %
Cm-244	0.00 %	0.07 %	31.36 %

Table XI
Percentage Contributions To The CEDE For Twenty Years Cooling Time

Nuclides	Binary Fuel	Ternary Fuel	LWR Fuel
Sr-90 + Y-90	0.66 %	0.14 %	0.11 %
Cd-113m	0.13 %	0.06 %	0.03 %
Cs-134	0.05 %	0.01 %	0.18 %
Cs-137 + Ba-137m	91.94 %	24.42 %	19.05 %
Ce-144 + Pr-144	0.00 %	0.00 %	0.00 %
Pu-238	3.83 %	1.39 %	16.33 %
Pu-239	3.36 %	20.79 %	1.43 %
Pu-240	0.02 %	20.33 %	2.23 %
Pu-241	0.01 %	4.74 %	5.33 %
Am-241	0.03 %	28.04 %	26.98 %
Am-243	0.00 %	0.02 %	0.71 %
Cm-244	0.00 %	0.06 %	27.62 %

Table XII
Percentage Contributions To The CEDE For Twenty Five Years Cooling Time

Nuclides	Binary Fuel	Ternary Fuel	LWR Fuel
Sr-90 + Y-90	0.65 5	0.13 %	0.10 %
Cd-113m	0.11 %	0.05 %	0.02 %
Cs-134	0.01 %	0.00 %	0.04 %
Cs-137 + Ba-137m	91.32 %	21.96 %	17.99 %
Ce-144 + Pr-144	0.00 %	0.00 %	0.00 %
Pu-238	4.11 %	1.35 %	16.63 %
Pu-239	3.75 %	20.96 %	1.51 %
Pu-240	0.02 %	20.50 %	2.38 %
Pu-241	0.00 %	3.78 %	4.46 %
Am-241	0.03 %	31.20 %	31.96 %
Am-243	0.00 %	0.02 %	0.75 %
Cm-244	0.00 %	0.05 %	24.15 %

Table XIII
Percentage Contributions To The CEDE For Thirty Years Cooling Time

Nuclides	Binary Fuel	Ternary Fuel	LWR Fuel
Sr-90 + Y-90	0.64 %	0.11 %	0.10 %
Cd-113m	0.10 %	0.04 %	0.02 %
Cs-134	0.00 %	0.00 %	0.01 %
Cs-137 + Ba-137m	90.67 %	19.78 %	16.88 %
Ce-144 + Pr-144	0.00 %	0.00 %	0.00 %
Pu-238	4.37 %	1.32 %	16.83 %
Pu-239	4.16 %	21.14 %	1.59 %
Pu-240	0.02 %	20.67 %	2.51 %
Pu-241	0.00 %	3.01 %	3.69 %
Am-241	0.04 %	33.87 %	36.48 %
Am-243	0.00 %	0.02 %	0.79 %
Cm-244	0.00 %	0.04 %	21.11 %

Tables VIII to XIII show that the fission products, especially ^{137}Cs-^{137m}Ba, dominate the CEDE for the binary fuel at all cooling times. Percentage contributions from ^{134}Cs drop rapidly after 10 years, and are slowly replaced by ^{238}Pu and ^{239}Pu. The higher actinides are not important because of the fast neutron spectra in metallic fuels and the low concentration of ^{238}U in the fresh fuel. Actinides dominate the CEDE from the ternary fuels because of the relatively high concentrations of Pu in the unirradiated fuel. While the fresh LWR fuel contain no transuranic elements, the abundance of ^{238}U and the low energy neutron spectra stimulate capture reactions to form isotopes of Pu, Am, and Cm. Contributions from ^{241}Am and ^{244}Cm that are important for LWR spent fuel are not significant in the metallic fuels. Elimination of ^{90}Sr-^{90}Y, ^{113m}Cd, and ^{144}Ce-^{144}Pr would simplify Tables VII to XII with no great loss of accuracy.

CONCLUSIONS

Relatively few nuclides influence the offsite radiological consequences of abnormal events involving spent fuel cooled for five or more years. Only cesium and the actinides contribute significantly to the CEDE from inhalation. Since the CEDE dominates the TEDE, submersion in airborne particles or gases can be neglected for most analyses if conservative (high) values are selected for the release fractions. While the assumptions in this paper have been developed for operations at ANL, the relative importance of the specific nuclides can be applied to any facility handling spent fuel with cooling times between five and thirty years. Reference 12, a review of

practices developed for several facilities that handle transuranic waste, found that there exists diverse approaches to performing radiological consequence analyses that have been approved by regulatory bodies. Elimination of all but the most important nuclides allows rapid assessments of offsite doses with little loss of accuracy. Since the ARF, RF, LPF and χ/Q can vary by orders of magnitude, it is not productive to consider nuclides that contribute less than a few percent of the total dose. Therefore, only ^{134}Cs, ^{137}Cs-^{137m}Ba, and the actinides significantly influence the offsite radiological consequences of severe accidents.

FOOTNOTES

[1]The submitted manuscript has been created by the University of Chicago as Operator of Argonne National Laboratory ("Argonne") under Contract No. W-31-109-ENG-38 with the U.S. Department of Energy. The U.S. Government retains for itself, and others acting on its behalf, a paid-up, nonexclusive, irrevocable worldwide license in said article to reproduce, prepare derivative works, distribute copies to the public, and perform publically, by or on behalf, of the Government.

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