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Source Terms for Radioactive Gaseous Effluents from a Model High-Level Waste Solidification Facility

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Prepared for the
U.S. Nuclear Regulatory Commission
Office of Nuclear Reactor Regulation
Under Interagency Agreement ERDA 40-549-75

Contract No. W-7405-eng-26

CHEMICAL TECHNOLOGY DIVISION

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Manuscript Completed - June 15, 1976

Date Published - November 1976

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Prepared by the
OAK RIDGE NATIONAL LABORATORY
Oak Ridge, Tennessee 37830
operated by
UNION CARBIDE CORPORATION
for the
ENERGY RESEARCH AND DEVELOPMENT ADMINISTRATION

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SOURCE TERMS FOR RADIOACTIVE GASEOUS EFFLUENTS FROM A
MODEL HIGH-LEVEL WASTE SOLIDIFICATION FACILITY

H. W. Godbee and A. H. Kibbey

ABSTRACT

The source terms (rates at which radionuclides are released) are estimated for the gaseous effluents from a model high-level waste solidification facility (WSF). The WSF is envisaged as being similar to the New Waste Calcining Facility (NWCF) being constructed at the Idaho National Engineering Laboratory (INEL) but with provisions for incorporating the calcine into a glass. The decontamination factor (DF) is estimated to be one for tritium, 100 for iodine, and 5.0×10^8 for ruthenium. The DFs for other nuclides are in the range of mid to high 10^9 . The volatile radionuclide of primary concern in waste solidification is ruthenium (in particular, ^{106}Ru). With an estimated DF of 5.0×10^8 , the ^{106}Ru expected to be released from the WSF amounts to 3.4, 2.9, and 0.091 mCi/day for immediate solidification, a freshly filled waste tank (189 days), and five years of tank storage, respectively. By way of comparison, it may be noted that the Final Safety Analysis Report of the Barnwell Nuclear Fuel Plant (BNFP) Separations Facility implies that 4.6 mCi/day of ^{106}Ru might be released from the stack of the separations facility and states that such a release meets all standards and specifications promulgated by both State and Federal regulatory agencies.

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BACKGROUND

Federal regulations (Appendix F of 10 CFR 50) require that the inventory of high-level liquid waste at a fuel reprocessing plant must be limited to that generated in the prior five years. In addition, these regulations require that the liquid waste must be converted to dry solids and transferred to a Federal repository no later than ten years following separation of fission products from irradiated fuels. Based on the success^{1,2} at the Idaho National Engineering Laboratory (INEL) in calcining a variety of Energy Research and Development Administration (ERDA) wastes in the Waste Calcining Facility (WCF), the application of fluidized-bed calcination to commercial high-level radioactive liquid waste is being considered.^{2,3} The calcined product is generally suitable for interim storage or for post-calcination treatment such as incorporation in a glass, ceramic, metal, or other thermally stable matrix.

Among the important considerations in evaluating the environmental impact of a waste solidification facility are the radiological doses due to nuclides released in gaseous effluents. Dose calculations require a "source term" for each nuclide released. In this memo, we have estimated the source terms (rates at which radionuclides are released) for the gaseous effluents from a model high-level waste solidification facility (WSF). The WSF is envisaged as being similar to the New Waste Calcining Facility (NWCF) being constructed at Idaho⁴ but with provisions for incorporating the calcine into a glass. The facility would probably have a stack which is about 100-m (330-ft) high. The total process off-gas (not ventilating air) flow through the stack might range from about 4 to 45 standard m³ (150 to 1600 ft³) per minute depending upon the concentration of the waste and the method of heating as well as fluidizing the calciner bed.⁵ The effect of isotopic exchange, radioactive decay, and recycle on the decontamination factor (DF) for an element is not taken into consideration in this study; i.e., the DF is assumed to be the same for each isotope of an element.

HIGH-LEVEL LIQUID WASTE

The high-level liquid waste used in this study comes from the reprocessing of spent light-water reactor (LWR) fuel with an initial enrichment of 2.8% which is irradiated to 29,000 MWd/MTU at a specific power of 26 MW/MTU and cooled 213 days before reprocessing.⁶ The plant operates 300 days per year and reprocesses fuel at the rate of 5 MTU/day. The waste is assumed to contain the following percentages of the fission-product and actinide radionuclides originally in the spent fuel: 5% of tritium;⁷ 5% of iodine;⁸ all of other semi-volatile and nonvolatile fission products;^{9,10} 1% of uranium and plutonium;^{11,12} and all of other actinides.^{9,10} Waste stored under three sets of conditions is considered (a) no tank storage, i.e., immediate solidification; (b) storage until a tank is full, i.e., 189 days for the 300,000-gal tank with a cooling capacity of 3.7×10^7 Btu/hr used in this study; and (c) five years of tank storage. The curies of fission products and actinides in the waste under each of these sets of conditions were calculated¹³ using the ORIGEN code¹⁴ and are given in Tables 1a and 1b, respectively.

GASEOUS SOURCE TERMS

In the WSF, liquid radioactive waste will be converted to granular solids in a fluidized-bed calciner. The free-flowing solids from the calciner will be mixed with glass frit. The calcine-frit mixture may be heated in the storage pot or in a separate glass-forming vessel from which the glassy product is drained into the storage pot. In this study, the daily production of glass is contained in two 0.9-ft-diam x 10-ft-long pots, each 80% full. The off-gas from the glass-forming equipment passes into the off-gas cleanup system of the calciner. This system consists of a series of cleanup devices, viz., a dry-collection device (cyclone) for removal of bulk solids, a wet-scrubbing device (venturi) for removal of fine particulates, silica-gel beds for removal of volatile ruthenium, a silver-zeolite bed for removal of iodine, a high-efficiency particulate air (HEPA) filter, a deep-bed filter, and a HEPA filter. The decontamination factors expected with this system

Table 1a. Curies of fission product radionuclides calculated to be present in the high-level liquid waste generated by the reprocessing of spent light water reactor (LWR) fuel^{a,b}

Fission product isotope	Curies/MTU processed			Fission product isotope	Curies/MTU processed		
	Time in waste storage tank				Time in waste storage tank		
	None ^c	189 days ^d	5 years		None ^c	189 days ^d	5 years
H-3	2.23E-01	2.55E-01	1.70E-01	Te-123m	1.56E-01	9.51E-02	1.93E-06
Zn-72	1.78E-32	2.67E-34	0	-125m	2.82E-03	2.67E-03	7.46E-02
Ge-72	2.56E-32	3.87E-34	0	-127m	4.16E-03	2.42E-03	2.19E-02
As-77	4.86E-37	5.77E-39	0	-127	4.12E-03	2.40E-03	2.17E-02
Se-77m	1.46E-39	1.79E-41	0	-129m	8.10E-02	2.05E-02	1.41E-14
-79	3.46E-01	3.47E-01	3.47E-01	-129	5.20E-02	1.32E-02	9.01E-15
Br-82	6.10E-41	6.86E-43	0	-130	2.40E-14	5.90E-16	0
Rb-86	1.81E-01	2.58E-02	1.05E-31	I-131	1.66E-07	1.67E-03	1.67E-03
-87	1.69E-07	1.69E-05	1.69E-05	-131	5.18E-04	3.19E-05	0
Sr-89	4.38E-04	1.60E-04	4.31E-07	-132	1.24E-15	6.01E-16	0
-90	6.55E-04	6.54E-04	5.78E-04	Cs-134	1.73E-05	1.59E-05	2.93E-04
Y-90	6.58E-04	6.55E-04	5.78E-04	-135	2.20E-01	2.20E-01	2.20E-01
-91	7.98E-04	3.20E-34	1.44E-05	-136	6.92E-01	6.87E-02	0
Zr-93	1.64E-00	1.64E-00	1.64E-00	-137	9.34E-04	9.29E-04	8.28E-04
-93	1.47E-05	6.34E-04	2.22E-04	Ba-137m	8.74E-04	8.69E-04	7.74E-04
Nb-93m	1.72E-01	1.92E-01	5.16E-01	-140	1.51E-01	1.40E-00	0
-93m	3.14E-03	1.35E-03	4.72E-06	La-140	1.74E-01	1.70E-00	0
-95	2.94E-05	1.31E-05	4.32E-04	Ce-141	1.57E-04	3.81E-03	4.15E-14
Mo-92	1.76E-17	3.75E-19	0	-143	3.06E-41	3.22E-43	0
Tc-99m	1.68E-17	3.62E-19	0	-144	5.88E-05	4.72E-05	5.47E-03
-99	1.26E-01	1.26E-01	1.26E-01	Pr-143	3.00E-01	3.14E-00	0
Ru-103	3.16E-04	9.21E-03	1.23E-10	-144	5.88E-05	4.72E-05	5.47E-03
-106	3.40E-05	2.86E-05	9.09E-03	Nd-147	1.06E-00	8.94E-02	0
Nb-103m	3.16E-04	9.22E-03	1.23E-10	Pm-147	8.58E-04	8.02E-04	2.14E-04
-105	1.15E-37	1.31E-37	0	-148m	1.06E-03	3.26E-02	2.71E-11
-106	3.40E-05	2.86E-05	9.09E-03	-148	8.54E-01	2.61E-01	2.17E-12
Pd-107	1.02E-01	1.02E-01	1.02E-01	-149	7.08E-24	1.20E-25	0
Ag-103m	3.02E-06	2.61E-06	1.60E-07	Sa-151	1.13E-03	1.13E-03	1.09E-03
-110m	1.93E-03	1.51E-03	1.01E-01	-153	9.22E-26	1.38E-29	0
-110	2.92E-02	1.96E-02	1.32E-00	Eu-152	1.03E-01	1.01E-01	7.59E-00
-111	1.23E-04	7.04E-25	0	-154	5.94E-03	5.87E-03	4.73E-03
Cd-105	3.02E-06	2.61E-06	1.60E-07	-155	5.32E-03	4.83E-03	7.11E-02
-113m	9.08E-00	8.99E-00	6.98E-00	-156	1.29E-01	1.47E-00	0
-115m	1.92E-01	6.00E-00	9.94E-13	Gd-153	1.89E-01	1.46E-01	7.83E-02
-115	1.47E-25	2.48E-27	0	Tb-160	1.69E-02	7.78E-01	1.85E-06
In-113m	7.96E-02	2.81E-02	2.87E-13	-161	3.58E-07	1.89E-06	0
-114	7.68E-02	2.72E-02	2.76E-13	Dy-166	1.56E-18	4.04E-20	0
-115m	1.59E-25	2.71E-27	0	Ho-166m	6.00E-04	6.01E-04	5.99E-04
Sa-117m	1.66E-03	1.78E-04	0	-166	2.32E-18	6.03E-20	0
-119m	8.50E-00	6.61E-00	4.18E-02	Total			
-121m	4.76E-04	4.73E-04	4.34E-04	F.P.s	3.12E-06	2.36E-06	3.65E-05
-123	2.70E-03	1.68E-03	6.78E-02				
-125	2.38E-03	1.70E-04	0				
-126	4.96E-01	4.95E-01	4.95E-01				
Sb-122	1.41E-20	3.01E-22	0				
-124	3.20E-01	1.30E-01	8.96E-09				
-125	6.94E-03	6.50E-03	1.80E-03				
-126m	4.94E-01	4.95E-01	4.95E-01				
-126	4.90E-01	4.91E-01	4.90E-01				
-127	2.28E-12	6.77E-14	0				

^aFuel with an initial enrichment of 2.8% which is irradiated to 29,000 MWD/MTU at a specific power of 26 MW/MTU and cooled 213 days before reprocessing.

^bWaste contains the following percentages of the fission-product and actinide radionuclides originally in the spent fuel: 7% of tritium; 5% of iodine; all of other semi-volatile and nonvolatile fission products; 1% of uranium and plutonium; and all of other actinides.

^cWaste as discharged from separations facility.

^dRepresents time required to fill a 300,000-gal tank (having a cooling capacity of 3.7×10^7 Btu/hr) with the waste from reprocessing spent fuel at the rate of 5 MTU/day.

Table 1b. Series of actinide nuclides calculated to be present in the high-level liquid waste generated by the reprocessing of spent light water reactor (LWR) fuel^{a,b}

Curies/MTU processed				Curies/MTU processed			
Actinide isotope	Time in waste storage tank			Actinide isotope	Time in waste storage tank		
	None ^c	189 days ^d	5 years		None ^c	189 days ^d	5 years
Tl-207	1.08E-06	1.24E-06	4.21E-06	U-232	6.32E-05	6.89E-05	1.29E-04
-208	7.32E-04	6.76E-04	1.44E-04	-233	3.94E-07	7.10E-07	7.21E-06
-209	6.56E-10	6.63E-10	7.07E-10	-234	6.48E-03	6.52E-03	7.71E-03
Pb-209	2.98E-08	1.02E-06	3.22E-08	-235	1.51E-04	1.51E-04	1.51E-04
-210	8.34E-10	9.98E-10	7.19E-09	-236	2.44E-03	2.44E-03	2.44E-03
-211	1.08E-07	1.25E-06	4.23E-06	-237	2.32E-02	2.29E-02	1.80E-02
-212	2.04E-07	1.88E-07	1.99E-04	-238	3.04E-03	3.04E-03	3.04E-03
-214	1.87E-08	2.08E-08	6.17E-08	-240	3.82E-17	1.24E-15	2.46E-14
Bi-210	8.34E-10	9.86E-10	7.10E-09	Wp-237	2.86E-01	2.85E-01	2.86E-01
-211	1.08E-06	1.25E-06	4.22E-06	-238	1.14E-25	1.84E-27	0
-212	2.04E-03	1.88E-03	3.99E-04	-239	1.61E+01	1.62E+01	1.61E+01
-213	2.98E-08	3.02E-08	3.22E-08	-240m	3.82E-15	1.25E-15	2.46E-14
-214	1.87E-08	2.08E-08	6.17E-08	Pu-236	2.50E-03	2.35E-03	6.96E-04
Po-210	5.70E-10	7.02E-10	6.74E-09	-238	2.36E+01	4.54E+01	9.02E+01
-211	3.24E-09	3.74E-09	1.27E-08	-239	3.12E+00	3.11E+00	3.11E+00
-212	1.30E-03	1.21E-03	2.55E-04	-240	4.54E+00	4.59E+00	5.54E+00
-213	2.32E-08	2.95E-08	3.14E-08	-241	9.62E-02	9.50E-02	7.49E-02
-214	1.87E-08	2.08E-08	6.17E-08	-242	1.26E-02	1.26E-02	1.26E-02
-215	1.08E-06	1.25E-06	4.22E-06	-243	2.00E-09	2.01E-07	2.01E-07
-216	2.04E-03	1.87E-03	1.99E-04	-244	3.84E-17	1.25E-15	2.47E-14
-218	1.87E-08	2.08E-08	6.17E-08	Am-241	1.67E+02	1.67E+02	1.72E+02
At-217	2.98E-08	3.02E-08	3.22E-08	-242m	8.40E+00	8.39E+00	8.20E+00
Fr-221	2.98E-08	3.02E-08	3.22E-08	-242	8.40E+00	8.43E+00	8.20E+00
-223	1.62E-08	1.85E-08	5.90E-08	-243	1.61E+01	1.62E+01	1.61E+01
Ra-223	1.08E-06	1.25E-06	4.22E-06	-244	4.98E-18	1.62E-18	3.21E-17
-224	2.04E-03	1.87E-03	1.99E-04	-245	2.58E-08	2.12E-08	3.76E-10
-225	3.00E-08	3.02E-08	3.22E-08	Om-242	1.39E-04	9.56E+03	1.08E+01
-226	1.87E-08	2.08E-08	6.17E-08	-243	3.74E+00	3.71E+00	3.33E+00
-228	3.86E-12	4.53E-12	1.46E-11	-244	2.06E+03	2.04E+03	1.68E+03
Ac-225	2.98E-08	3.02E-08	3.22E-08	-245	2.98E-01	2.99E-01	2.89E-01
-227	1.16E-06	1.32E-06	4.22E-06	-246	5.58E-02	5.58E-02	5.58E-02
-228	3.86E-12	4.53E-12	1.46E-11	-247	2.00E-07	2.01E-07	2.01E-07
Th-227	1.10E-06	1.26E-06	4.16E-06	-248	6.10E-07	6.10E-07	6.10E-07
-228	2.02E-03	1.85E-03	3.97E-04	-249	2.54E-12	3.47E-13	4.77E-14
-229	3.02E-08	3.02E-08	3.22E-08	-250	7.98E-14	7.99E-14	7.99E-14
-230	1.88E-05	1.88E-05	1.90E-05	Bk-249	1.72E-03	1.41E-03	3.57E-05
-231	1.51E-02	2.73E-04	1.51E-04	-250	7.98E-14	8.00E-14	7.99E-14
-232	2.92E-11	2.92E-11	2.98E-11	Cf-249	3.26E-06	4.02E-06	7.33E-06
-234	3.04E-01	5.80E-02	3.03E-03	-250	2.76E-05	2.73E-05	2.10E-05
Pa-231	2.08E-05	2.08E-05	2.06E-05	-251	2.04E-07	2.04E-07	2.03E-07
-232	9.18E-50	9.26E-52	0	-252	3.14E-05	2.93E-05	8.19E-04
-233	2.96E-01	2.85E-01	2.86E-01	-253	8.20E-10	1.11E-10	1.53E-11
-234m	3.04E-01	5.80E-02	3.03E-03	-254	7.86E-11	3.22E-11	2.65E-20
-234	3.04E-04	5.86E-05	3.03E-06	Es-253	1.28E-07	2.14E-09	8.88E-36
Total				Actinides	1.72E+04	1.28E+04	2.77E+03

^aFuel with an initial enrichment of 2.8% which is irradiated to 29,000 MMd/MTU at a specific power of 26 MW/MTU and cooled 213 days before reprocessing.

^bWaste contains the following percentages of the fission-product and actinide radionuclides originally in the spent fuel: 5% of tritium; 5% of iodine; all of other semi-volatile and nonvolatile fission products; 1% of uranium and plutonium; and all of other actinides.

^cWaste as discharged from separations facility.

^dRepresents time required to fill a 300,000-gal tank (having a cooling capacity of 3.7×10^7 Btu/hr) with the waste from reprocessing spent fuel at the rate of 5 MTU/day.

during calcination for all elements except iodine are taken from ref. 15 and are listed in Table 2. A conservatively low, average, removal efficiency of 99% (DF = 100) for iodine on a silver-zeolite bed is used. The DF for calcination is defined as the ratio of the amount of an isotope in the liquid feed to the amount of that isotope in the off-gas from the facility during calcination. The DFs expected during glass formation were estimated as follows:

- (1) utilizing the data given in ref. 16 to calculate, as illustrated in the Appendix, the fraction of semi-volatile rubidium, molybdenum, ruthenium, tellurium, and cesium lost from a 0.9-ft-diam pot of glass at 1000°C for 12 hr to be 3.6×10^{-4} , 5.1×10^{-5} , 2.6×10^{-5} , 1.0×10^{-3} , and 1.3×10^{-3} , respectively;
- (2) assuming that the fraction of each nonvolatile element lost or entrained from the pot under these conditions is 1.0×10^{-5} ; and
- (3) taking the DF of the calciner off-gas cleanup system to be 1.0×10^7 when it treats off-gas from the glass-forming vessel.

The DFs for glass formation estimated (see Appendix) with these assumptions are listed in Table 2. This DF is defined as the ratio of the amount of an isotope in the calcined solid to the amount of that isotope in the off-gas from the facility during glass formation. The overall DFs (see Appendix) for calcination plus glass formation, defined as the ratio of amount in liquid feed to amount in total off-gas, are also listed in Table 2. The gaseous source terms for this calcination-glass forming facility were calculated using the following information: (a) the facility solidifies the high-level liquid waste from a plant which reprocesses spent fuel at the rate of 5 MTU/day for 300 days per year; (b) the curies of radionuclides in the waste are those in Tables 1a and 1b, and (c) the overall DFs are those in Table 2. These source terms are listed in Tables 3a and 3b for fission products and actinides, respectively.

Table 2. Decontamination factors expected during the calcination of and glass formation with high-level liquid waste

Element	Decontamination factor ^a		
	Calcination	Glass formation	Overall ^b
Tritium	1.0E+00	c	1.0E+00
Iodine	1.0E+02	c	1.0E+02
Particulates	1.0E+10	1.0E+12	9.9E+09
Rubidium	1.0E+10	2.8E+10	7.4E+09
Molybdenum	1.0E+10	2.0E+11	9.5E+09
Ruthenium	5.0E+08	3.8E+11	5.0E+08
Tellurium	1.0E+10	1.0E+10	5.0E+09
Cesium	1.0E+10	7.7E+09	4.4E+09

^a Defined as the ratio of the amount of an isotope in the feed to the amount of that isotope in the off-gas.

^b The overall process consist of calcining liquid waste at about 500°C, mixing the calcined solids with a glass frit, and heating the calcine-glass frit mixture to about 1000°C to form a glassy product.

^c All tritium and iodine are volatilized in the calcination step.

Table 1a. Series of fission products released annually from treating high-level liquid waste with a fluidized-bed calciner and incorporation of the calcine into glass.^a

Fission product isotope	Annual release rate (Ci/yr)			Fission product isotope	Annual release rate (Ci/yr)		
	Time in waste storage tank				Time in waste storage tank		
	None ^b	189 days ^c	5 years		None ^b	189 days ^c	5 years
H-3	3.42E+04	3.38E+04	2.56E+04	Te-123m	4.84E-08	2.91E-03	5.90E-13
Cm-77	2.70E-39	3.99E-41	0	-125u	8.63E-04	8.16E-04	2.28E-04
Ge-72	3.88E-39	5.80E-41	0	-127m	1.27E-03	7.42E-04	6.71E-09
As-77	7.36E-44	1.06E-45	0	-127	1.26E-03	7.35E-04	6.64E-09
Se-77m	2.21E-46	2.71E-48	0	-129m	2.48E-04	6.28E-05	4.31E-21
-79	5.24E-08	5.26E-08	5.26E-08	-129	1.59E-04	4.05E-05	2.76E-21
Br-82	9.24E-48	1.04E-49	0	-132	7.35E-21	1.83E-22	0
Rb-86	3.68E-08	5.23E-19	2.13E-38	I-129	2.50E-02	2.51E-02	2.51E-02
-87	3.43E-12	3.43E-12	2.43E-12	-131	7.77E-03	4.78E-04	0
Sr-89	6.64E-03	2.42E-03	6.53E-14	-132	1.86E-14	9.02E-15	0
-90	9.97E-03	9.91E-03	8.75E-03	Cs-134	5.90E-02	5.41E-02	9.99E-03
Y-90	9.97E-03	9.92E-03	8.75E-03	-135	7.50E-08	7.50E-08	7.50E-08
-91	1.21E-02	4.84E-03	2.18E-12	-136	2.36E-07	2.34E-08	0
Zr-93	2.48E-07	2.49E-07	2.47E-07	-137	3.18E-02	3.17E-02	2.82E-02
-95	2.24E-02	9.62E-03	3.37E-11	Ba-137m	1.32E-02	1.32E-02	1.17E-02
Nb-93m	2.60E-08	2.90E-08	7.82E-08	-140	2.29E-06	2.24E-07	0
-95m	4.76E-04	2.05E-04	7.15E-13	La-140	2.64E-06	2.58E-07	0
-95	4.45E-02	1.99E-02	7.31E-11	Ce-141	2.38E-03	5.77E-04	6.29E-21
Mo-99	2.78E-24	5.91E-26	0	-143	4.64E-48	4.87E-50	0
Tc-99m	2.54E-24	5.48E-26	0	-144	8.91E-02	7.15E-02	8.29E-04
-99	1.92E-06	1.91E-06	1.91E-06	Pr-143	4.55E-06	4.76E-07	0
Ru-103	9.48E-02	2.76E-02	3.68E-16	-144	8.91E-02	7.15E-02	8.29E-04
-106	1.02E+00	8.57E-01	2.73E-02	Nd-147	1.60E-07	1.35E-08	0
Nb-103m	4.79E-03	1.40E-03	1.86E-17	Pm-147	1.30E-02	1.22E-02	3.24E-03
-105	1.74E-44	1.99E-46	0	-148m	1.61E-04	4.94E-05	4.10E-18
-106	5.15E-02	4.33E-02	1.38E-03	-148	1.29E-05	3.96E-06	3.29E-19
Pd-107	1.54E-08	1.54E-08	1.54E-08	-149	1.07E-30	1.81E-32	0
Ag-109m	4.58E-13	3.96E-13	2.42E-14	Sm-151	1.72E-04	1.72E-04	1.65E-04
-110m	2.93E-04	2.29E-04	1.54E-06	-153	1.40E-34	2.08E-36	0
-110	3.82E-05	2.97E-05	2.00E-07	Eu-152	1.56E-06	1.53E-06	1.15E-06
-111	1.86E-11	1.07E-12	0	-154	9.00E-04	8.90E-04	7.17E-04
Cd-109	4.58E-13	3.96E-13	2.42E-14	-155	8.06E-04	7.31E-04	1.08E-04
-113m	1.37E-06	1.36E-06	1.06E-06	-156	1.95E-06	2.23E-07	0
-115m	2.91E-06	9.09E-07	1.51E-19	Gd-153	2.87E-06	2.21E-06	1.19E-08
-115	2.20E-32	3.75E-34	0	Tb-160	2.56E-05	1.18E-05	2.81E-13
In-114m	1.21E-08	4.26E-09	4.35E-20	-161	5.42E-14	2.87E-15	0
-114	1.16E-08	4.12E-09	4.18E-20	Dy-166	2.36E-25	6.12E-27	0
-115m	2.40E-32	4.10E-34	0	Ho-166m	9.09E-11	9.11E-11	9.07E-11
Sn-117m	2.52E-10	2.69E-11	0	-166	3.52E-25	9.14E-27	0
-119m	1.29E-06	1.00E-06	6.33E-09	Total			
-121m	7.21E-11	7.20E-11	6.88E-11	F.P.s	3.42E+04	3.38E+04	2.56E+04
-123	4.09E-04	2.55E-04	1.01E-08	Total			
-125	3.61E-10	2.58E-11	0	F.P.s			
-126	7.52E-08	7.50E-08	7.50E-08	less H-3	1.62E+00	1.27E+00	1.28E-01
Sb-122	2.13E-27	4.55E-29	0				
-124	4.85E-06	1.97E-06	1.36E-15				
-125	1.05E-03	9.84E-04	2.73E-04				
-126m	7.52E-08	7.50E-08	7.50E-08				
-126	7.42E-08	7.44E-08	7.42E-08				
-127	3.45E-19	1.03E-20	0				

^a Plant operates 300 days per year and treats the high-level liquid waste from a 5 MTU/day fuel reprocessing plant. The waste contains the fission-product nuclides given in Table 1a. Decontamination factors expected in the overall treatment (calcination plus glass formulation) are listed in Table 2.

^b Represents immediate solidification.

^c Represents the time required to fill a 300,000-gal tank (having a cooling capacity of 3.7×10^7 Btu/hr) with the waste from reprocessing spent fuel at the rate of 5 MTU/day.

Table 1b. Series of actinides released annually from treating high-level liquid waste with a fluidized-bed calciner and incorporation of the calcine into glass^a

Actinide isotope	Annual release rate (Ci/yr)			Actinide isotope	Annual release rate (Ci/yr)		
	Time in waste storage tank				Time in waste storage tank		
	None ^b	189 days ^c	5 years		None ^b	189 days ^c	5 years
Tl-207	1.64E-13	1.88E-13	6.38E-13	U-232	9.58E-17	1.04E-11	1.96E-11
-208	1.11E-10	1.02E-10	2.16E-11	-233	5.97E-14	1.08E-13	1.09E-12
-209	9.94E-17	1.01E-16	1.07E-16	-234	9.82E-10	9.88E-10	1.17E-09
Pb-209	4.52E-15	4.57E-15	4.87E-15	-235	2.29E-11	2.29E-11	2.29E-11
-210	1.26E-16	1.51E-16	1.09E-15	-236	3.70E-10	3.69E-10	3.69E-10
-211	1.64E-13	1.89E-13	6.40E-13	-237	3.52E-09	3.46E-09	2.73E-09
-212	3.09E-10	2.85E-10	6.04E-11	-238	4.61E-10	4.59E-10	4.59E-10
-214	2.83E-15	3.16E-15	9.35E-15	-240	5.79E-24	1.89E-22	3.72E-21
Bi-210	1.26E-16	1.49E-16	1.09E-15	Mp-237	4.31E-08	4.31E-08	4.33E-08
-211	1.64E-13	1.89E-13	6.40E-13	-238	1.73E-12	2.77E-14	0
-212	3.09E-10	2.85E-10	6.04E-11	-239	2.45E-06	2.45E-06	2.44E-06
-213	4.52E-15	4.57E-15	4.87E-15	-240m	5.79E-22	1.89E-22	3.72E-21
-214	2.83E-15	3.16E-15	9.35E-15	Pu-236	3.79E-10	3.56E-10	1.05E-10
Po-210	8.64E-17	1.06E-16	9.46E-16	-238	3.58E-06	6.89E-06	1.37E-05
-211	4.91E-16	5.66E-16	1.92E-15	-239	4.73E-07	4.71E-07	4.71E-07
-212	1.97E-10	1.83E-10	3.86E-11	-240	6.88E-07	6.96E-07	8.40E-07
-213	4.42E-15	4.47E-15	4.76E-15	-241	1.46E-04	1.44E-04	1.14E-04
-214	2.83E-15	3.16E-15	9.35E-15	-242	1.91E-09	1.91E-09	1.91E-09
-215	1.64E-13	1.89E-13	6.40E-13	-243	3.03E-16	3.05E-14	3.05E-14
-216	3.09E-10	2.84E-10	6.04E-11	-244	5.82E-24	1.89E-22	3.74E-21
-218	2.83E-15	3.16E-15	9.35E-15	Am-241	2.53E-05	2.53E-05	2.61E-05
At-217	4.52E-15	4.57E-15	4.87E-15	-242m	1.27E-06	1.27E-06	1.24E-06
Fr-221	4.52E-15	4.57E-15	4.87E-15	-242	1.27E-06	1.28E-06	1.24E-06
-223	2.45E-15	2.81E-15	8.95E-15	-243	2.45E-06	2.45E-06	2.44E-06
Re-223	1.64E-13	1.89E-13	6.40E-13	-244	7.54E-25	2.45E-25	4.86E-24
-224	3.09E-10	2.84E-10	6.04E-11	-245	3.91E-15	3.21E-15	5.69E-17
-225	4.55E-15	4.57E-15	4.87E-15	Om-242	2.11E-03	1.45E-03	1.64E-06
-226	2.83E-15	3.16E-15	9.35E-15	-243	5.67E-07	5.63E-07	5.05E-07
-228	5.85E-19	6.86E-19	2.21E-18	-244	3.12E-04	3.09E-04	2.55E-04
Ac-225	4.52E-15	4.57E-15	4.87E-15	-245	4.36E-08	4.38E-08	4.38E-08
-227	1.75E-13	2.00E-13	6.40E-13	-246	8.45E-09	8.47E-09	8.45E-09
-228	5.85E-19	6.86E-19	2.21E-18	-247	3.03E-14	3.05E-14	3.05E-14
Th-227	1.66E-13	1.91E-13	6.30E-13	-248	9.24E-14	9.24E-14	9.24E-14
-228	3.06E-10	2.81E-10	6.01E-11	-249	3.85E-19	5.26E-20	7.23E-51
-229	4.58E-15	4.57E-15	4.87E-15	-250	1.21E-20	1.21E-20	1.21E-20
-230	2.85E-12	2.85E-12	2.89E-12	Bk-249	2.61E-10	2.13E-10	3.80E-12
-231	2.29E-09	4.14E-11	2.29E-11	-250	1.21E-20	1.21E-20	1.21E-20
-232	4.42E-18	4.43E-18	4.52E-18	Cf-249	4.94E-13	6.09E-13	1.11E-12
-234	4.61E-08	8.79E-09	4.59E-10	-250	4.18E-12	4.14E-12	3.17E-12
Pa-231	3.15E-12	3.16E-12	3.16E-12	-251	3.09E-14	3.09E-14	3.08E-14
-232	1.39E-56	1.40E-58	0	-252	4.76E-17	4.44E-12	1.20E-12
-233	4.33E-08	4.33E-08	4.33E-08	-253	1.24E-16	1.68E-17	2.32E-48
-234m	4.61E-08	8.79E-09	4.59E-10	-254	1.19E-17	4.87E-18	4.01E-27
-234	4.61E-11	8.88E-12	4.59E-13	Es-253	1.95E-15	3.24E-16	1.35E-42
Total				Actinides	2.60E-03	1.94E-03	4.20E-04

^a Plant operates 300 days per year and treats the high-level liquid waste from a 5 MTU/day fuel reprocessing plant. The waste contains the actinide nuclides given in Table 1b. Decontamination factors expected in the overall treatment (calcination plus glass formation) are listed in Table 2. Actinides are considered to be particulates.

^b Represents immediate solidification.

^c Represents the time required to fill a 100,000-gal tank (having a cooling capacity of 1.7×10^7 Btu/hr) with waste from the reprocessing of spent fuel at the rate of 5 MTU/day.

CONCLUSIONS

The most critical area of any waste solidification process in terms of potential for radiological dose is the removal of volatile and particulate radioactivity from the off-gas of the calciner and/or glass-forming vessel prior to release to the atmosphere. The DFs for particulates are generally adequate with most off-gas cleanup systems that have been used or proposed. The volatile radionuclide of primary concern in waste solidification is ruthenium (in particular, ^{106}Ru).¹⁷ With an estimated DF of 5.0×10^8 (Table 2), the ^{106}Ru expected to be released from the WSF (Table 3a) amounts to 3.4, 2.9, and 0.091 mCi/day for immediate solidification, a freshly filled waste tank (189 days), and five years of tank storage, respectively. By way of comparison, it may be noted that the Final Safety Analysis Report of the Barnwell Nuclear Fuel Plant (BNFP) Separations Facility implies that 4.6 mCi/day of ^{106}Ru might be released from the stack of the separations facility¹⁸ and states that such a release meets all standards and specifications promulgated by both State and Federal regulatory agencies.¹⁹ Obviously, the amount of ruthenium that would be acceptable for release from a facility depends on many factors such as stack height, meteorological conditions, and distance to site boundary. For example, it has been estimated²⁰ that a maximum of 7 Ci/day could be released by the WCF in Idaho and still meet Federal regulations (Appendix B of 10 CFR 20).

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APPENDIX: ESTIMATION OF GLASS-FORMATION AND OVERALL DFs

With the assumption that the glass formed is similar in composition to that in ref. 16, the fraction of rubidium, molybdenum, ruthenium, tellurium, and cesium lost from the glass-forming vessel may be estimated from the information presented in ref. 16. The procedure is illustrated in the following discussion. From Fig. 20 in ref. 16, the total weight loss from the glass at 1000°C in 12 hr amounts to about $2.2 \times 10^{-2} \text{ g/cm}^2$. In this study one 0.9-ft-diam x 10-ft-long pot is filled in 12 hr. The exposed surface area of glass (cross-sectional area of pot) is roughly 590 cm^2 . Thus, approximately 13 g of material $[(2.2 \times 10^{-2} \text{ g/cm}^2) \times (590 \text{ cm}^2)]$ is estimated to be lost from the pot. From Fig. 22 in ref. 16, the fraction of cesium in this lost material (1000°C for 12 hr) is shown as about 0.58. Therefore, on the order of 7.5 g of cesium ($13 \text{ g} \times 0.58$) would be lost from the pot. From ref. 13, the amount of cesium contained in a pot would be approximately $5.9 \times 10^3 \text{ g}$. Accordingly, the fraction of cesium lost from a pot is calculated to be $(7.5 \text{ g}) / (5.9 \times 10^3 \text{ g})$ or 1.3×10^{-3} . The fractions of rubidium ($0.26 \text{ g} / 7.2 \times 10^3 \text{ g}$), molybdenum ($0.39 \text{ g} / 7.6 \times 10^3 \text{ g}$), ruthenium ($0.13 \text{ g} / 5.0 \times 10^3 \text{ g}$), and tellurium ($1.3 \text{ g} / 1.3 \times 10^3 \text{ g}$) were calculated in a similar manner.

The DF for glass formation is obtained by multiplying the DF of the pot (i.e., the reciprocal of the fraction lost) by the DF of the calciner off-gas system for each element. For example, with cesium this amounts to multiplying $1 / 1.3 \times 10^{-3}$ or 7.7×10^2 by 1×10^7 to give a DF for glass formation of 7.7×10^9 .

The overall DF for calcination plus glass formation is obtained rigorously from an overall material balance on each element. However, since the amount of each element in the liquid waste is for all practical purposes equal to the amount of that element in the calcine and this in turn is equal to the amount of that element in the glass, the overall DF is very nearly the reciprocal of the sum of the reciprocals of the individual DFs; that is,

$$\frac{1}{\text{overall DF}} \approx \frac{1}{\text{calcination DF}} + \frac{1}{\text{glass formation DF}} .$$

For example, with cesium

$$\frac{1}{\text{overall DF}} = \frac{1}{1 \times 10^{10}} + \frac{1}{7.7 \times 10^9} \text{ or overall DF} = 4.4 \times 10^9 .$$

All the DFs obtained in this manner are listed in Table 2.