RADIATION TRANSPORT IN HIGH-LEVEL WASTE FORM

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ABSTRACT

The waste form selected for vitrifying high-level nuclear waste stored in underground tanks at West Valley, NY is borosilicate glass. The maximum radiation level at the surface of a canister filled with the high-level waste form is prescribed by repository design criteria for handling and disposition of the vitrified waste. This paper presents an evaluation of the radiation transport characteristics for the vitreous waste form expected to be produced at West Valley and the resulting neutron and gamma dose rates. The maximum gamma and neutron dose rates are estimated to be less than 7500 R/h and 10 mRem/h respectively at the surface of a West Valley canister filled with borosilicate waste glass.

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Introduction

The high-level waste resulting from reprocessing spent uranium fuel from several nuclear reactors using the PUREX process at the Nuclear Fuel Services site in West Valley, New York is stored as neutralized liquid waste in a carbon steel tank. This waste has separated into a hydroxide sludge settled at the bottom of the tank and a salt solution on top of the sludge. Spent fuel containing thorium was reprocessed using the THOREX process and resulting waste is stored in a stainless steel tank.

The West Valley Nuclear Services Company has been given the task of solidifying these liquid high-level waste into borosilicate glass for disposal in a federal repository. The waste glass produced at the site has to meet certain repository requirements specified in the Waste Acceptance Preliminary Specification[1] (WAPS) document. The amount of glass forming chemicals to be added to the waste such that the resulting waste glass stored in stainless canisters meets the WAPS has been determined during the product development activity. As a part of activity an estimation of the radionuclides contained in the canistered waste and its consequences during handling and disposal have also been determined.

This paper deals with the determination of radiation dose and other effects resulting from radionuclides in canistered waste glass. Methods and results of some of the analyses performed during the process and product development are discussed.

Source Determination

Radio-chemical analysis of samples taken from the tanks is the primary method of determining the type and strength of the radionuclides in the high-level wastes stored at the site. This is an ongoing program in which several samples of the waste have been obtained and analyzed at several laboratories. The results are compared and a list of radionuclides and their concentrations have been generated and are updated every year. In addition the spent fuel reprocessing steps have been simulated using the ORIGEN-2 computer code[2] to compute the radionuclides expected to be present in the waste. The modeling results are also used to derive the strength of the radionuclides not measurable in radiochemical laboratory. From this information a conservative estimate of the radiation sources inventory expected in the waste was determined. The list is shown in Table 1.

Several glasses have been prepared with varying oxide composition and tested for compliance with the WAPS as well as processability requirements. The type and amount of oxides needed to bind the radionuclides in a glass matrix was determined from product evaluations. Table 2 shows chemical oxide content of the high-level waste glass expected to be produced at West Valley. The total amount of waste glass that will be produced at the WVDP is about 475,000 kg, which will be stored in about 260 stainless steel canisters. The radionuclide inventory in each canister was calculated by dividing the total inventory by the number of canisters expected. Using this as sources in a canister, the radiation intensity surrounding the canister can be evaluated.

Methods of Analyses

The gamma flux at the surface of a canister filled with high-level waste glass is determined by the multigroup, one-dimensional, discrete ordinate transport code with anisotropic scattering, ANISN[3]. Numerical solution of the transport equation by the discrete ordinates method requires both cross sections and source data at the discrete energy levels. Angular dependence of scattering cross sections were represented by orthogonal Legendre polynomial expansion while other nuclear parameters were treated explicitly for a few directions. Finite difference approximation has been used to discretize spatial dependence. Multigroup gamma sources, at various times as required, were determined by using the isotope generation and depletion code ORIGEN-2. ORIGEN-2 is a reactor physics code that simulates spent fuel reprocessing steps, where necessary, and computes various high-level waste characteristics. Radionuclide data such as half-life, scattering, nuclear reaction cross sections and energy of emitting radiations provide the necessary information for computing the heat generation rate, gamma spectrum and neutron sources and sinks in the canistered waste. The multigroup gamma source was then modeled in ANISN as a fixed source in glass former oxides and the appropriate group dependent cross section and S_4 level symmetric quadrature sets were input to compute gamma fluxes at the surface of the canister. The gamma fluxes were then converted to dose rates by the appropriate energy and absorption dependent conversion factors.

The neutron sources in the waste glass are not sufficient enough to warrant any rigorous neutron transport calculation. The amount of fissile material is small and the waste glass contains a significant amount of neutron absorbing boron. For a conservative estimate of neutron dose at the surface of the canister, the net neutrons (production of spontaneous fission and alph-n reaction - absorption by boron) produced and its flux at the surface were calculated and converted to a dose rate with appropriate conversion factor.

The other effects anticipated from a high-level waste loaded canister which can be estimated from these calculations are the generation of heat by absorbing radiation and the generation of helium within the canisters by the emitted alpha particles.

The estimated canister heat generation rate was determined by multiplying the radionuclide concentration by its recoverable energy values. This information is necessary to specify the temporary storage cell design requirements so that the maximum center line temperature of the high-level waste canister is below the limit specified in the Waste Acceptance Specification.

The potential for pressurization of a canister filled with waste glass was analyzed by assuming that all the alpha particles released from the wastes would accumulate over a hundred years in the canister as helium gas. The resulting pressure increase was calculated using normal canister fill conditions.

Results and Conclusion

Heat generation, neutron production and absorption and alpha emission rates in a canistered waste were computed by decaying the radionuclides in the canister to the time of production and the time of shipment to repository using the ORIGEN-2 code.

Using the multigroup (18 groups) gamma sources generated in ORIGEN-2, in ANISN-W code the multigroup gamma fluxes at the boundary of 2 ft diameter high-level waste canister were determined. The resulting integrated gamma dose rate is calculated to be 6900 R/hr. An independent calculation of the gamma dose rate using the QADMOD-G[4] code was made for comparison purposes. The independent calculation, being less rigorous, yielded a dose rate of 7400 R/hr. To estimate the uncertainty in these calculations an analysis of the gamma dose rate at the top of the 8D-2 tank was made using the two codes and compared with the measured dose rate. The calculated dose rate was within 10% of the measured dose rate.

The neutrons sources (neutrons per second) from alpha-n reactions and spontaneous fission are 2.753E+05 and 4.432E+06 and decrease to 2.536E+05 and 2.220E+06 respectively after twenty years in storage. The presence of B-10 in significant quantity in the canistered waste accounts for neutron absorption at the rate of 1.094E+06 neutrons/sec. The net neutron dose rate at the surface of the canister is less than 10 mrem/hr which is negligibly small.

The heat generation rate in a nominal canister (85 percent full) is about 324 watts and decreases to 50 watts in about 100 years. Assuming an uncertainty of 15 percent attributed to process variability, a maximum heat generation rate of 374 watts is expected in a canister.

If every alpha particle emitted by radionuclides contained in a canistered waste is converted into a helium atom, it would amount to about 2.89E-02 liters per year. The increase in pressure, over a storage period of 100 years, would be about 0.48 psi assuming a void space of 10 percent of canister volume.

From these analyses it is concluded that the effects of radiation in the HLW canisters are well within the limits identified in the Waste Acceptance Preliminary Specifications.

REFERENCES

1. Office of Civilian Radioactive Waste Management, "Waste Acceptance Preliminary Specifications for The West Valley Demonstration Project High-Level Waste Form," DOE/RW-0261, Revision 1. U.S. Department of Energy, Washington, DC, January, 1990.

2. A. G. Croff, "ORIGEN-2 A Revised and Updated Version of The Oak Ridge Isotope Generation and Depletion Code," ORNL, 5621, Oak Ridge National Laboratory (1980).

3. ANISN: "Multigroup One-Dimensional Discrete Ordinates Transport Code with Anisotropic Scattering," CCC-254, Radiation Shielding Information Center, Oak Ridge National Laboratory.

4. QADMOD-G: "Point Kernal Gamma Ray Shielding Code," CCC-369, Radiation Shielding Information Center, Oak Ridge National Laboratory.

Table 1
Estimated Radionuclide Content (curies)
in West Valley Wastes, 1987 Baseline

Nuclide	PUREX Supernatant	PUREX Sludge	THOREX	Total	Nuclide	PUREX Supernatant	PUREX Sludge	THOREX	Total
3-H	9.74E+01	0.00E+00	1.74E+00	9.91E+01	223-Fr	0.00E+00	1.26E-05	1.04E-01	1.04E-01
14-C	1.37E+02	0.00E+00	1.30E-01	1.37E+02	223-Ra	0.00E+00	9.14E-04	7.52E+00	7.52E+00
55-Fe	0.00E+00	1.00E+03	5.63E+02	1.56E+03	224-Ra	0.00E+00	1.19E-01	9.76E+00	9.88E+00
60-Co	0.00E+00	4.70E+00	1.14E+03	1.14E+03	225-Ra	0.00E+00	6.61E-06	2.07E-01	2.07E-01
59-Ni	0.00E+00	8.56E+01	2.03E+01	1.06E+02	228-Ra	0.00E+00	4.81E-09	1.48E+00	1.48E+00
63-Ni	8.89E+02	5.34E+03	2.51E+03	8.74E+03	225-Ac	0.00E+00	6.61E-06	2.07E-01	2.07E-01
79-Se	5.68E+01	0.00E+00	3.35E+00	6.02E+01	227-Ac	0.00E+00	9.14E-04	7.52E+00	7.52E+00
90-Sr	2.89E+03	6.74E+06	4.54E+05	7.20E+06	228-Ac	0.00E+00	4.81E-09	1.48E+00	1.48E+00
90-Y	2.89E+03	6.74E+06	4.54E+05	7.20E+06	227-Th	0.00E+00	9.01E-04	7.42E+00	7.42E+00
93-Zr	2.56E-01	2.56E+02	1.62E+01	2.72E+02	228-Th	0.00E+00	1.19E-01	9.76E+00	9.88E+00
93m-Nb	1.59E-01	1.59E+02	1.02E+01	1.69E+02	229-Th	0.00E+00	6.61E-06	2.07E-01	2.07E-01
99-Tc	1.60E+03	0.00E+00	1.04E+02	1.70E+03	230-Th	0.00E+00	1.45E-02	4.38E-02	5.83E-02
106-Ru	1.10E-01	1.10E+02	6.24E-01	1.11E+02	231-Th	6.41E-03	8.94E-02	5.17E-03	1.01E-01
106-Rh	1.10E-01	1.10E+02	6.24E-01	1.11E+02	232-Th	0.00E+00	5.87E-09	1.64E+00	1.64E+00
107-Pd	1.09E-02	1.09E+01	1.14E-01	1.10E+01	234-Th	5.71E-02	7.97E-01	7.11E-06	8.54E-01
113m-Cd	2.41E+00	2.41E+03	3.75E+01	2.45E+03	231-Pa	0.00E+00	2.86E-04	1.52E+01	1.52E+01
121m-Sn	1.76E-02	1.76E+01	5.99E-01	1.82E+01	233-Pa	0.00E+00	2.30E+01	3.02E-01	2.33E+01
126-Sn	1.01E-01	1.01E+02	1.01E+02	3.11E+00	234a-Pa	5.71E-02	7.97E-01	7.11E-05	8.54E-01
125-Sb	4.90E+01	1.51E+04	2.89E+02	1.54E+04	232-U	3.13E-01	4.36E+00	2.74E+00	7.41E+00
126-Sb	1.42E-02	1.42E+01	4.35E-01	1.46E+01	233-U	4.98E-01	6.94E+00	2.09E+00	9.53E+00
126m-Sb	1.01E-01	1.01E+02	3.11E+00	1.04E+02	234-U	2.80E-01	3.90E+00	2.17E-01	4.40E+00
125m-Te	1.20E+00	3.70E+03	7.08E+01	3.78E+03	235-U	6.41E-03	8.94E-02	5.17E-03	1.01E-01
129-I	2.10E-01	0.00E+00	1.80E-01	3.90E-01	236-U	1.91E-02	2.67E-01	9.80E-03	2.96E-01
134-Cs	1.39E+04	0.00E+00	3.10E+02	1.42E+04	238-U	5.71E-02	7.97E-01	7.11E-05	8.54E-01
135-Cs	1.56E+02	0.00E+00	5.47E+00	1.61E+02	236-Np	0.00E+00	9.35E+00	1.23E-01	9.47E+00
137-Cs	7.26E+06	0.00E+00	4.75E+06	7.74E+06	237-Np	0.00E+00	2.30E+01	3.02E-01	2.33E+01
137m-Ba	6.87E+06	0.00E+00	4.49E+05	7.32E+06	239-Np	0.00E+00	3.43E+02	4.49E+00	3.47E+02
144-Ce	2.09E-05	9.21E+00	1.39E-01	9.35E+00	236-Pu	1.36E-02	8.24E-01	1.09E-02	8.49E-01

Table 1
Estimated Radionuclide Content (curies)
in West Valley Wastes, 1987 Baseline (Cont.)

Nuclide	PUREX Supernatant	PUREX Sludge	THOREX	Total	Nuclide	PUREX Supernatant	PUREX Sludge	THOREX	Total
144-Pr	2.09E-05	9.21E+00	1.39E-01	9.35E+00	238-Pu	1.27E+02	8.00E+03	4.80E+02	8.61E+03
146-Pm	4.77E-02	1.53E+01	5.07E-01	1.59E+01	239-Pu	2.54E+01	1.61E+03	1.54E+01	1.65E+03
147-Pm	5.71E+02	1.85E+05	9.11E+03	1.95E+05	240-Pu	1.87E+01	1.18E+03	8.09E+00	1.21E+03
151-Sm	5.03E-01	8.15E+04	4.78E+03	8.63E+04	241-Pu	1.46E+03	9.23E+04	8.50E+02	9.46E+04
152-Eu	4.57E-02	3.77E+02	4.82E+01	4.25E+02	242-Pu	2.54E-02	1.61E+00	1.19E-02	1.65E+00
154-Eu	1.44E+01	1.19E+05	2.53E+03	1.22E+05	241-Am	0.00E+00	5.30E+04	2.41E+02	5.32E+04
155-Eu	2.37E+00	3.54E+04	8.44E+02	3.26E+04	242-Am	0.00E+00	2.93E+02	6.76E+00	2.99E+02
207-Tl	0.00E+00	9.12E-04	7.50E+00	7.50E+00	242m-Am	0.00E+00	2.94E+02	6.79E+00	3.01E+02
208-Tl	0.00E+00	4.28E-02	3.51E+00	3.55E+00	243-Am	0.00E+00	3.39E+02	7.83E+00	3.47E+02
209-Pb	0.00E+00	6.61E-06	2.07E-01	2.07E-01	242-Cm	0.00E+00	2.438E+02	5.95E+00	2.49E+02
211-Pb	0.00E+00	9.14E-04	7.52E+00	7.52E+00	243-Cm	0.00E+00	1.44E+02	2.34E-01	1.44E+02
212-Pb	0.00E+00	1.19E-01	9.76E+00	9.88E+00	244-Cm	0.00E+00	8.56E+03	1.37E+01	8.57E+03
211-Bi	0.00E+00	9.14E-04	7.52E+00	7.52E—00	245-Cm	0.00E+00	8.62E-01	2.00E-02	8.82E-01
212-Bi	0.00E+00	1.19E-01	9.76E+00	9.88E+00	246-Cm	0.00E+00	9.87E-02	2.29E-03	1.01E-01
213-Bi	0.00E+00	6.61E-06	2.07E-01	2.07E-01					
212-Po	0.00E+00	7.62E-02	6.25E+00	6.33E+00					
213-Po	0.00E+00	6.47E-06	2.03E-01	2.03E-01					
215-Po	0.00E+00	9.14E-04	7.52E+00	7.52E+00					
216-Po	0.00E+00	1.19E-01	9.76E+00	9.88E+00					
217-At	0.00E+00	6.61E-06	2.07E-01	2.07E-01					
219-Rn	0.00E+00	9.14E-04	7.52E+00	7.52E+00					
220-Rn	0.00E+00	1.19E-01	9.76E+00	9.88E+00					
221-Fr	0.00E+00	6.61E-06	2.07E-01	2.07E-01					

Table 2
Nominal Oxide Content of the WVDP Glass
As a Function of Source
(kilograms)

Oxide	PUREX*	THOREX	Zeolite	Glass-Forming Additives	Final Glass
Al_2O_3	4100	1000	9800	15700	30600
B_2O_3	30	270	0	60900	61200
CaO	1700	10	460	0	2170
Fe_2O_3	52800	2800	2000	0	57600
K_2O	0	120	19400	4300	23820
Li_2O	4	0	0	9600	9604
MgO	400	10	440	3400	4250
MnO	2700	40	0	1200	3940
Na_2O	7000	240	11500	19200	37940
P_2O_5	5700	5	0	0	5705
SiO_2	7100	60	38000	150600	195760
ThO_2	0	17100	0	0	17100
TiO_2	1	0	330	3500	3831
UO_2	2700	4	0	0	2704
ZrO_2	0	0	0	11000	11000
OTHER+	7400	6	5	0	7411
TOTAL	91635	21665	81935	279400	474635

* Washed PUREX sludge
\+ Oxide components that individually, constitute less than 0.5 percent of the glass