PRELIMINARY ASSESSMENT OF DISPOSAL LIMITS FOR VITRIFIED LOW-LEVEL-MIXED-WASTES IN RCRA LANDFILLS

James R. Cook, and Dennis F. Bickford
Westinghouse Savannah River Company
Savannah River Technology Center
Aiken, SC 29808

ABSTRACT

A generic RCRA-type landfill design has been used to estimate the inventory of mixed waste that could be disposed of at a humid site using a vitrified waste form. Contaminant release was modeled as a solubility limited phenomena. Limits for a number of metals, organics, and radionuclides are presented.

SUMMARY

The U.S. Department of Energy's Mixed Waste Integrated Program includes development of a generic delisting petition for mixed Resource Conservation and Recovery Act (RCRA) and low level radioactive wastes. Thus, a generic modified RCRA landfill is being evaluated for disposal of both hazardous and mixed radioactive and hazardous wastes that have been vitrified to pass the Land Disposal Restrictions (LDR). The landfill would include multiple barriers to restrict and limit the release of hazardous and radioactive substances to the environment. Included are preliminary upper limits for both radionuclide and hazardous chemical inventories which could be allowed in the landfill. Thus, the work presented here will aid in the development of both Waste Acceptance Criteria and Operating Limits for Mixed Waste RCRA Landfills. Forty isotopes and nineteen hazardous chemicals which are typical of DOE waste streams are considered. Preliminary limits for those constituents which impact groundwater are recommended, as well as those radioactive species which could affect intruders.

BACKGROUND

RCRA landfills might be used to simultaneously meet the performance objectives of DOE Order 5820.2A (1) and the requirements of the Resource Conservation and Recovery Act (2) (RCRA). Vitrification at temperatures above 1000°C provides destruction and removal of hazardous organics and can provide primary confinement for radioactive and inorganic wastes. Synthetic liners and engineered low-permeability clay barriers can provide secondary and tertiary confinement, respectively. In addition, at final closure a layer of highly impermeable clay can be placed over the landfill to minimize infiltration of water. This paper discusses the initial results of calculations of the ability of the combination of vitrification treatment and an engineered landfill system to isolate the combined hazardous and radioactive materials, and maintain the local environment within acceptable standards without active intervention. Where the generic model required site specific data, the corresponding data for the Savannah River Site (SRS) has been used.

DOE Order 5820.2A sets forth the requirements for radioactive waste (and therefore, the radioactive portion of mixed waste) disposal within the DOE complex. Each disposal facility is required to have a *site-specific performance* assessment to demonstrate that the facility will meet the performance requirements stated in the Order. Waste acceptance criteria, which guarantee that the waste emplaced in the facility is within the bounds of that analyzed by the performance assessment, are also required. In this analysis, wastes with transuranic nuclide concentrations of 100 nCi/g or less are considered low level waste. Vitrified wastes contain no free liquids, and vitrified fine particulate wastes, such as ashes, are immobilized by the glass matrix.

RCRA is a prescriptive regulation in that it requires waste treatment and engineered disposal facilities utilizing a double liner and leachate collection system. A Generic Mixed Waste/RCRA Landfill is being evaluated for compliance with the requirements of both RCRA and DOE Order 5820.2A. This analysis provides preliminary limits on the amounts of individual radionuclides and hazardous chemicals which can be placed the Landfill.

Two computer codes were used in this work, the HELP code and PATHRAE. Each of these codes is described, and the methodology used to apply them to this problem is explained. The Hydrologic Evaluation of Landfill Performance (HELP) code was developed by the U. S. Army Corps of Engineers for the U. S. Environmental Protection Agency.(3,4,5,6) The purpose of the code is to evaluate various designs for shallow land burial waste disposal systems in terms of their effect on the overall water balance. Up to fourteen layers may be used, including closure caps and liners. Input data required by the code consist of the physical dimensions of the waste site and each of the layers, hydraulic properties of the layers, and climatic information. The code itself provides default hydraulic properties for a number of possible cap and liner materials. Climatic data consists of ten years of daily rainfall data from the SRS F-Area meteorologic tower and other data provided with the code for Augusta, Georgia.

Output from the code is the water balance resulting from the particular design and climate. Water balance means that the amount of water exiting the overall system must equal that which enters the system as precipitation. This exiting water is partitioned into evapotranspiration back into the atmosphere, surface runoff, lateral drainage, and infiltration. In general, evapotranspiration and lateral drainage are beneficial to the closure system. Surface runoff can cause erosion of the cap system and should be minimized by engineered construction features. Similarly, infiltration causes leaching of the buried waste, so it should be reduced to the extent possible.

The HELP code has been the subject of several verification studies by the U. S. Army Corps of Engineers,(6, 7) and it has undergone a sensitivity analysis by Oak Ridge National Laboratory.(8) The methodology used in the sensitivity analysis also performs a very rigorous check on the actual computer code to assure that it is internally self consistent.

The purpose of the PATHRAE (9, 10) computer code is to calculate doses and health effects which might be caused by disposal of waste material in the near surface environment. Radioactive decay chain effects are included. The code was developed for the U. S. Environmental Protection Agency, and is accepted by that organization. PATHRAE was selected for use in the Environmental Impact Statement on Waste Management Activities and Groundwater Protection at the Savannah River Site.(11) Input to the code consists of a number of parameters which describe the waste and the characteristics of the disposal site and the surrounding area. The use of site specific information about the waste and the site conditions increase confidence in the results produced by the code.

The PATHRAE code can calculate doses caused by a number of exposure pathways: groundwater transport to a surface stream, groundwater use in a nearby well, surface erosion and subsequent waste exposure, trench overflow (bathtub effect), food grown on the site, biointrusion into the waste, direct exposure, inhalation of dust on-site, inhalation of radon gas, and atmospheric transport of particulates off-site. The code assumes that the waste inventory is evenly distributed throughout the waste volume. For the groundwater transport pathways a specified fraction of the inventory is leached from the waste each year and transported vertically through the unsaturated zone, and then horizontally to hypothetical wells at locations at 1 meter, 100 meters, and to a nearby stream. The water velocity through the unsaturated zone is calculated in the code from input values for infiltration and soil porosity. The velocity in the water table is a code input, the value of which is taken from three-dimensional numerical modeling by Parizek and Root. (12)

The velocity of each radionuclide considered is calculated based on the partition coefficient (commonly called Kd) and the water velocity. A quantity called the retardation factor is calculated from the partition coefficient and used to modify the velocity of a species relative to the water velocity. For example, uranium with a partition coefficient of 40 has a retardation factor equal to 320, meaning that uranium is transported 320 times more slowly than the groundwater. Reference 13 provides the bases for these values.

The Erosion pathway considers the effects of erosion of the cover material and transport and deposition in a nearby stream. The pathway called the Bathtub Effect calculates doses due to disposal trenches overflowing if geologic and precipitation conditions are such that this phenomenon might occur. The Food Grown on Site pathway assumes that at some time in the future someone moves onto the disposal facility and constructs a home with a basement and a water well. Excavation of the basement and well construction brings some portion of the buried waste to the surface, which is then used to grow food in a small garden. The person is thus exposed to waste constituents from the food he eats. The Biointrusion pathway is essentially the same as the Food Grown on Site except that no home is constructed. The Direct Exposure pathway considers the effects of penetrating gamma radiation (RAD version) or skin contact (HAZ version) from the buried waste both to workers during the operational phase and to someone occupying the disposal site and disturbing the waste sometime in the future. The Dust pathway considers inhalation of resuspended dust by an intruder at the waste disposal facility due to house construction and well drilling. The Radon Inhalation (RAD version) or Chemical Vapor (HAZ version) pathway calculates exposures due to gaseous contaminants in a structure built over a disposal facility. The Atmospheric Transport pathway considers off-site transport of materials due to dust resuspension, incineration, or trench fire.

In each of these pathways a hypothetical person is exposed to materials released from the disposal facility. The PATHRAE code calculates the dilution, dispersion, or attenuation provided by the waste site and the environment, and thus the concentration of each contaminant to which the person will be exposed. For the ingestion and inhalation pathways, dose conversion factors from the International Commission on Radiological Protection (ICRP) (14,15) are used to calculate annual doses from curie concentrations. Dose conversion factors for direct gamma exposure are taken from the PRESTO data base. (16) The PRESTO methodology is used for the food chain analysis in the HAZ calculations.

As stated above, the PATHRAE code was used as the basis for dose calculations for the Groundwater Protection and Waste Management Environmental Impact Statement for SRS issued by DOE in 1987. The model was qualified by, 1) review of code documentation, history of use, and previous validation and verification studies, 2) comparison of model results to alternate models using different boundary conditions, 3) comparison of model predictions to measured concentrations, and 4) sensitivity analysis to identify critical input parameters.(17)

DOSE CRITERIA

Radionuclides

The dose criteria set forth in DOE Order 5820.2A were used in this study for radionuclides. The Order states that the dose to an inadvertent intruder be no more than 500 mrem for a single acute exposure, such as digging into or drilling through the buried waste. For continuous exposure, that is living on the waste site and growing food there, the limit is 100 mrem/yr. No member of the general population should receive more than 25 mrem/year by all pathways. In addition, the Order states that the disposal facility must meet all applicable local, state and federal regulations for groundwater protection. Though no such regulations exist at this time for radionuclides, a limit of 4 mrem/yr for groundwater at the edge of the disposal facility at all times during operation and after closure was used.

For this study the 100 mrem/yr limit was used and applied at 100 years after site closure to estimate intruder doses. Studies by Kennedy and Peloquin (18) and Aaberg and Kennedy (19) have shown that the 100 mrem/yr limit for continuous exposure always results in lower allowable waste concentrations than the 500 mrem short term exposure limit. For times less than 100 years, the general population was considered to be at the SRS boundary. After 100 years the general population was assumed to be at the edge of the disposal site. The times, locations and dose limits for each type of exposure considered in this report are summarized in Table I.

Chemicals

The regulations governing disposal of hazardous materials are prescriptive, in that they state the treatment and

TABLE I
Locations, Times, and Radionuclide Dose Limits Used

Performance Objective		Location and Time of Compliance: Waste Site	Waste Site Boundary	SRS Site Boundary
Groundwater Protection	4 mrem/yr[a]	NA	All times	NA
General Population	25 mrem/yr[b]	NA	> 100 years	0-100 years
Intruder	100 mrem/yr[b]	> 100 years	NA	NA
	500 mrem[b]	> 100 years	NA	NA

[a]Proposed EPA limit (20).
[b]DOE Order 5820.2A (1).

TABLE II
Groundwater Protection Limits

Species	Limit (mg/L)	Source
Arsenic	0.050	Final DWS (21)
Barium	2.000	Final DWS (21)
Benzene	0.005	Final DWS (21)
Cadmium	0.010	Final DWS (21)
Chromium	0.100	Final DWS (21)
Cyanide	0.200	Proposed DWS (22)
Dichloromethane	0.005	Set by SRS
Lead	0.015	Final DWS (21)
Mercury	0.002	Final DWS (22)
Nickel	0.100	Final DWS (22)
Nitrate	10.0	Final DWS (21)
PCB's	0.0005	Final DWS (23)
Selenium	0.050	Final DWS (21)
Silver	0.050	Final DWS (21)
Tetrachloromethane	0.005	Final DWS (21)
Tetrachlorethylene	0.005	Final DWS (23)
1,1,1-Trichlorethane	0.200	Final DWS (21)
Trichlorethylene	0.005	Final DWS (20)
Xylene	10.0	Final DWS (23)

disposal technologies to be used. For the purpose of setting disposal limits in this report, protection of groundwater to the levels given by current or proposed regulations was adopted. The criteria used are shown in Table II. (20-23)

METHODOLOGY

Conceptual Waste Treatment and Disposal Method

The waste resulting from incineration, stabilization and wastewater treatment will be vitrified into glass or glass/ceramic monoliths, with the objective and effect of removing and destroying organics, and reducing the leachability of inorganic waste constituents. The release mechanism used in this study was that the waste constituents would be released only as the glass matrix dissolved in infiltrating water. Once released from the glass matrix, the waste materials would then be sorbed onto the clay surrounding the glass monoliths. Solubility of the glass was taken to be 7.8 E-05 moles/l (24). Kd values for the clay were taken from Sheppard and Thibault (25), and the method described by Baes and Sharp (26) was used to estimate release of sorbed material from the clay.

Vitrified monoliths approximately 8 inches in diameter and one foot tall were assumed to be deposited in a two-foot-square horizontal grid array with interstices packed with kaolin. One foot of packed kaolin is placed between successive layers of the monoliths. The landfill is 200 meters square by 6 meters deep. In the PATHRAE model no credit was taken for containers.

Conceptual Site and Disposal Facility Parameters

The initial modeling step in this work was to use the HELP code to estimate the long-term infiltration rate to be expected through a clay barrier surrounding the disposed vitrified waste. Parameters for the PATHRAE code were taken from values used in the Environmental Information Documents (EIDs) for new radioactive waste disposal (27) and new hazardous and mixed waste disposal (28) with a few exceptions: 1) For the groundwater pathway the hypothetical person consumed 730 liters of water per year (2 liters per day). In the EIDs this value was 365 liters per year (1 liter per day); 2) The exposure from drinking groundwater was calculated using well characteristics of a monitoring well rather than a domestic well, i.e., a fifteen foot screen zone rather than the 33 feet used in the EIDs. This is consistent with the protection criteria and the limits given in Table II and III) The exposure from groundwater considered only drinking water. Other uses, such as irrigation and watering food and milk producing animals were included in the intruder cases. This is consistent with the groundwater protection criteria; 4) The atmospheric transport pathway, where contamination is spread by a trench fire to offsite individuals, was not considered credible for vitrified waste.

Each radionuclide and chemical hazard was initially assigned an arbitrary large inventory. The results for each pathway and each constituent were examined and compared with the performance objectives given above. Inventories were adjusted so that the calculated dose for the most limiting pathway for each constituent would equal the performance objective for that pathway. Only a few of the ten pathways considered by PATHRAE were found to be significant: groundwater, direct gamma, resuspended dust and food grown on site.

Since uranium and the transuranics have decay chains which include other radioactive species, the RADDECAY program was used to determine if any daughter radionuclides would be produced in sufficient quantities to exceed the inventory limits calculated for them as parents. The decay chains which were investigated are:

Cm-244	=>	Pu-240	=>	U-236
Pu-240	=>	U-236	=>	Th-232
Am-243	=>	Pu-239	=>	U-235
Pu-240	=>	U-236	=>	Th-232
Am-243	=>	Pu-239	=>	U-235
Pu-241	=>	Am-241	=>	Np-237
Pu-242	=>	U-238	=>	U-234
Pu-238	=>	U-234		

RESULTS AND DISCUSSION

The results of this study are shown in Table III. Many of the radionuclides are shown as having a limit of 1E+09 Curies, meaning that even with an initial inventory of 1 billion curies the performance objectives are not exceeded. The property that these "unlimited" nuclides have in common is a short half life. Because the barriers prevent radionuclide release for 100 to 300 years, isotopes with half-lives less than about 5 years decay to insignificant amounts before they can reach the environment.

The remaining isotopes fall into two categories, those which are limited by the groundwater pathway and those which are limited by intruder pathways. This difference in the method of exposure has implications for the manner in which facility limits should be implemented. Radionuclides which impact the groundwater system must first be leached out of the vitrified wasteforms by percolating water and then be transported through the vadose (unsaturated) zone. This mechanism therefore integrates over the entire disposal facility, so that the overall facility inventory is the controlling factor. Intrusion scenarios, however, involve only a small fraction of the waste disposed in the facility, that which is disturbed by excavation and drilling. This means that individual waste package concentrations are the controlling factor.

Consideration of daughter ingrowth from radioactive decay led to limits being set for Pu-241 (ingrowth of Np-237), Am-241 (ingrowth of Np-237), and Pu-240 (ingrowth of U-236).

TABLE III
Preliminary Landfill Disposal Limits for Vitrified Mixed Waste

Chemical	Inventory Limit (kg)	Nuclide	Inventory Limit (Ci)
Arsenic	2.04E+04	C-14	1.54E+01
Barium	1.00E+06	Cf-252	1.00E+09
Benzene	1.69E+00	Co-58	1.00E+09
Cadmium	5.39E+02	Co-60	1.00E+09
Chromium	2.88E+04	Cs-134	1.00E+09
Cyanide	7.12E+01	Cs-137	1.59E+06
Dichloromethane	1.66E+00	Eu-154	5.06E+08
Freon	3.38E+00	Eu-155	1.00E+09
Lead	1.88E+05	H-3	5.12E+03
Mercury	1.59E+04	I-129	1.08E-01
Nickel	2.56E+04	Nb-94	5.12E+04
Nitrate	1.00E+06	Nb-95	1.00E+09
PCB's	7.63E-01	Ni-59	3.73E+05
Selenium	8.00E+03	Pm-147	1.00E+09
Silver	9.8E+03	Rb-87	7.14E+07
Tetrachloromethane	1.73E+00	Rh-106	1.00E+09
Tetrachloroethylene	1.79E+00	Ru-106	1.00E+09
1,1,1-Trichloroethane	6.80E+01	Sb-125	1.00E+09
Trichloroethylene	1.70E+00	Se-75	1.00E+09
Xylene	1.00E+06	Se-79	1.65E+04
		Tc-99	2.34E+01
		Th-232	5.54E+01
		U-234	5.04E+02
		U-235	1.71E+02
		U-236	1.73E+02
		U-238	1.86E+02
		Np-237	3.81E-01
		Pu-238	5.00E+07
		Pu-239	1.90E+07
		Pu-240	9.15E+04
		Pu-241	5.67E+04
		Pu-242	3.33E+02
		Am-241	1.89E+03
		Am-243	7.69E+05
		Sr-90	5.00E+07
		Y-90	5.00E+07

Many of the chemical hazards are destroyed or removed during vitrification. All chemical constituents were limited by the groundwater pathway, since there are no requirements for an intruder analysis for hazardous chemicals.

Using SRS as a typical DOE Site, typical DOE Site inventories are lower than the Preliminary Landfill Inventories given in Table III. No radionuclides are within an order of magnitude of the calculated limit, when the vitrification treatment is considered.

When the chemical constituents are considered, the Preliminary Landfill Inventory Limits of Table 3 are much higher than the typical actual inventories of DOE Sites, provided that the wastes are vitrified, or otherwise thermally treated. The high organic destruction and removal efficiencies of the vitrification process are particularly beneficial, since more traditional solidification methods have severe restrictions on many organics.

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