PERFORMANCE ASSESSMENT OF DOUBLE-SHELL TANK WASTE AT HANFORD

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ABSTRACT

The low-level liquid waste stored in double-shell tanks at Hanford will be solidified within a cementitious matrix (grout) and disposed of in subsurface vaults. This paper discusses activities related to the preparation of a site-specific performance assessment for grout disposal, as required by Department of Energy Order 5820.2A.

Site-specific data are being used in the preparation of a performance assessment for the planned grout disposal system at Hanford. The assessment will estimate the incremental increase in radiological dose to future populations who, after loss of institutional control at the site, use groundwater downgradient of the disposal site. Increases in nonradiological species in water from a hypothetical well also will be estimated. Two-dimensional transport models are being used to estimate contaminant concentrations in groundwater. Sensitivity studies on various parameters are in progress. The performance assessment will be updated as additional data become available.

INTRODUCTION

It is planned to dispose of 44 million gallons of low-level liquid wastes at Hanford by solidifying the waste within a cementitious (grout) matrix[1]. The grout will be produced in the Grout Treatment Facility (GTF) at Hanford. The GTF, which was constructed during 1986 through 1988, consists of a Dry Materials Facility, Transportable Grout Equipment, waste feed tank, and disposal vaults[2]. Waste from a million-gallon feed tank will be mixed with grout formers, creating a slurry that will be pumped to below-grade vaults. Each vault will hold 1.4 million gallons of the slurry and will take about one month to fill. The slurry will harden within several days after production. After the grout has set, any excess liquid will be pumped from the vault and the remaining void will be filled with nonradioactive grout. A closure cover will be placed over the vaults, and eventually a long-term protective barrier will be placed over the entire disposal site.

This paper discusses the performance assessment process that is being carried out to guide the disposal system design and to meet the requirements of the Department of Energy (DOE) Order 5820.2A.

PERFORMANCE ASSESSMENT PROCESS

Performance assessment is an iterative process. For the application described here, the disposal system is defined based on conceptual or definitive designs. The performance of the system, or the long-term environmental impacts, are estimated based on scenarios prescribed by regulations or dictated by the proposed environmental setting. The estimated long-term impacts are compared with regulations. If the estimated impacts are significantly below regulatory criteria and the assessment is defensible, then the plans for disposal can proceed. If not, there may be options to improve the design of the disposal system, improve performance of the waste form, or reduce conservatism in modeling. Reduction of conservatism in models usually requires the use of more complex codes and additional data.

At Hanford, the first scoping analysis for the disposal of the low-level fraction of double-shell tank wastes used one-dimensional models with many of the parameters estimated because better defined data were not available. As additional data became available, subsequent performance analyses became more sophisticated and two-dimensional models were used.

In 1987, a performance assessment was prepared for the disposal of very low activity wastes from N Reactor operations at Hanford[3]. Following this evaluation, the same methodology for performance assessment was used to evaluate a simplified grout disposal system for the projected 44 million gallons of low-level wastes. In this evaluation, diffusional release of contaminants from the grout was modeled. Using one-dimensional flow and transport models, contaminant concentrations as functions of arrival times at hypothetical downgradient groundwater receptor points were determined for various contaminants. The dose to potential users of the groundwater was calculated.

Results of this scoping analysis showed that an improvement in system performance was required. The addition of a long-term infiltration barrier was projected to cause some reduction in peak doses and significant retardation of the contaminant plume. However, the reduction of the peak contamination still did not meet proposed release criteria. Thus, an additional engineered feature was proposed to reduce the release of contaminants from the vault. This feature, called a diffusion barrier, would isolate the vaults from advecting water in the surrounding soil. It was

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also believed that some improvement could be realized by using more complex models, which could eliminate conservative assumptions that were needed in the one-dimensional analysis. These combined enhancements were anticipated to allow a comfortable margin of safety in meeting release goals.

Since the initial performance evaluation in 1987, efforts have been initiated to obtain the required data and computer codes to permit a more refined projection of the disposal system performance.

DISPOSAL SYSTEM DESIGN

The Hanford Reservation is in a semi-arid environment in Washington State. The surface of the disposal site for grouted low-level waste is about 90 meters above the water table. The soils at the disposal site are primarily sands and silty sands.

The disposal system consists of grout, subsurface concrete vaults, diffusion barriers, interim barriers, and a protective barrier (Fig. 1). Each component will be discussed further.

The reference grout consists of liquid waste and grout-forming solids blended at a ratio of 9 lb of solids per gallon of waste. The major waste components are sodium hydroxide, sodium nitrate, and sodium nitrite. The grout-forming solids are 47 wt% blast furnace slag, 47 wt% class F fly ash, and 6 wt% Portland I-II cement. Prior to each production campaign, the processability of waste in the feed tank and the acceptability of the product will be verified in laboratory tests.

The disposal site is estimated to consist of 44 concrete vaults. The internal dimensions of the vaults are approximately 125 ft long by 50 ft wide by 34 ft deep. The base of a vault is about 4.5 ft thick, the top is 2 ft thick, and the thickness of the sides varies from 2 to 4 ft. Under each vault is a lined concrete catch basin to satisfy requirements stipulated by the Resource Conservation and Recovery Act (RCRA). Each vault will be filled with grouted waste to about the 30-ft level. The remaining space will be filled with non-radioactive grout.

A continuous layer of asphalted material, which serves as a hydraulic/diffusion barrier, will surround each vault. The relatively high unsaturated conductivity of the soils surrounding the asphalted material will divert liquid around the vault and essentially eliminate advecting water flow to the vault. The low moisture content and the hydrophobic

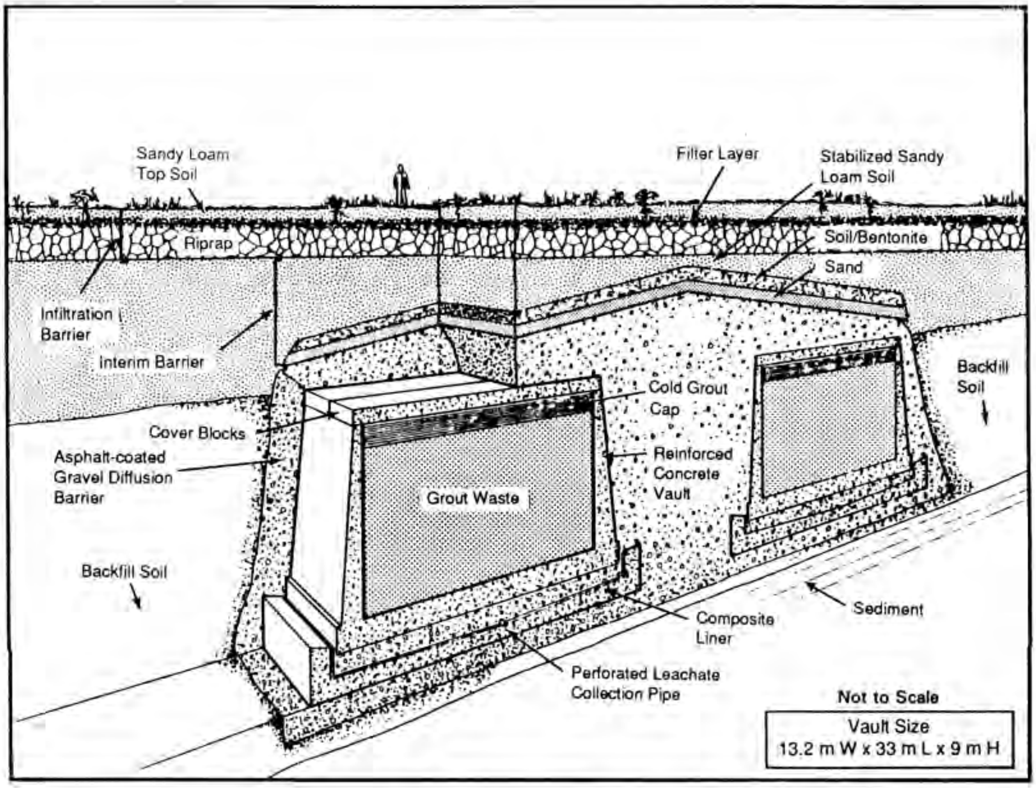

Fig. 1. Vault Conceptual Design.

nature of the asphalt results in a low effective diffusion coefficient across the surface of the barrier.

Above a pair of vaults is a clay/soil barrier that meets the RCRA requirements and diverts advecting water away from the gravel diffusion barrier.

Finally, a long-term protective barrier[4] is planned for placement above the entire disposal site to provide low infiltration rates and to resist intrusion by animals and plant roots. To deter human intrusion, the protective barrier will include large granite monuments with warning markers at various intervals along the border, as well as ceramic disks with warnings buried in the top surface of the barrier.

SCENARIOS/CRITERIA

Two exposure scenarios are being considered: use of water from a hypothetical downgradient well by the future population (full-garden scenario), and use of water from the Columbia River by the future population. Each scenario is discussed below.

Full Garden Scenario - This scenario estimates the impacts to a maximum-exposed individual who resides just downgradient of the grout disposal site and uses well water for drinking, irrigating crops for humans and livestock, and watering livestock. His garden provides 25% of his food intake. This scenario has been justified based on the significant amount of farming by irrigation in the region around Hanford. Estimates from this scenario are compared with the standard of 25 mrem/yr (all pathways) prescribed by the DOE order. Water from the well is also compared with existing and proposed drinking water standards. To account for potential impacts from other disposal actions at Hanford, grout disposal has been apportioned 20% of the dose and drinking water standards.

River Scenario - This scenario estimates the impacts to future populations that use the Columbia River for drinking water, agriculture, fishing, and recreation. The criterion for this scenario is also the 25 mrem/yr all- pathways dose.

DATA

The data used in the performance assessment stems from numerous experiments, chemical analyses, radiological analyses, and the open literature. In addition, some assumptions were necessary for cases in which experiments have not been completed or long-term performance isn't known. Strategy for key data are described below.

Inventory - The inventory of contaminants consists of those in the waste and the dry materials used for solidification. The waste inventory is based on a projected total waste volume of 44 million gallons. At this time, analysis of three million gallons of waste has been completed[5]. The bounding inventory for the performance assessment is calculated based on the average measured waste concentrations adjusted by statistical methods. As additional samples of waste are analyzed, the average concentrations, standard deviations, and projected upper bound waste inventory will be revised. If the inventory of key species increases during this process, the impacts to the performance assessment will be determined.

The inventory of contaminants in the dry blend is based on analyses of the various components. As materials for each production campaign are received, additional analyses will be performed to reduce the uncertainty of this portion of the inventory.

Current analyses have identified technetium-99 and iodine-129 as the key impact radionuclides in the inventory. Selenium-79 also provides small contributions. The controlling chemical is nitrate, assuming that all the nitrite is converted into nitrate in the soil column and the groundwater. All of these species of concern are anionic in the grout waste form, with little or no tendency to sorb onto Hanford sediments. Inventories of key radionuclides are listed in Table I and the total chemical inventory is listed in Table II. The performance assessment also accounts for organics; however, impacts due to organics are extremely low and therefore are not discussed here.

Grout - The principal data used in the performance assessment for the grout is the diffusivity of various contaminants within the grout. These data have been collected over several years for grouts made using both actual and simulated double-shell tank waste. In addition, a data base is being built for numerous species in other types of grouts to help support the values used in the performance assessment[6]. These data have been collected via methods such as the ANS 16.1[7] and MCC-1[8] leach tests.

Diffusion Barrier - The effective diffusivity of components through the diffusion barrier is a key parameter in the performance assessment. Effective diffusivity has been determined for gravels and asphalt-coated gravels using an electrical conductivity technique. Additional long-term verification tests are proposed in the future.

The ability of the diffusion barrier to divert water from the grout waste form is also essential in assessing the long-term performance of the disposal system. The capacity to divert advecting water has been studied in physical tests and with computer models.

Infiltration barrier - Data on the performance of the infiltration barrier is being collected under a separate, long-term program. The goal of the program is to develop a barrier that limits infiltration to an average rate of 0.05 cm/yr. The value used in the baseline case for performance assessment is 0.1 cm/yr. Sensitivity to this parameter is being examined.

Sorption - Many components in the grouted waste are adsorbed by Hanford soils. Because of the relatively deep

TABLE I

Inventory of Key Radionuclides for Grouted Waste[5]

Radionuclide	Mean Concentration, (a) Ci/L	Upper-Bound Inventory, (b) Ci
^{14}C	8.4E-07	1.9E+02
^{79}Se	6.7E-06	4.2E+03
^{94}Nb	1.0E-05	5.8E+03
^{99}Tc	7.7E-05	1.5E+04
^{129}I	1.7E-07	5.1E+01
^{237}Np	5.8E-08	3.5E+01

(a) Based on mean composition of three tanks.
(b) Based on 95% confidence level; 167 million liters (44 million gallons) of waste.

soil column and the low infiltration rate, short-lived fission products such as cesium and strontium will decay to insignificant levels before they can diffuse from the grout and travel through the soil column to the groundwater. The transuranics also strongly sorb to the soils. The combination of slow release from the waste form, due to solubility control and sorption once released, makes the transuranic elements insignificant factors in the pathways involving groundwater. Data on sorption has come from batch sorption tests and sorption tests with grout leachates flowing in columns of soil.

TABLE II

Summary of Inorganic Chemicals in Grouted Waste

	Waste		Dry Blend		Total Inventory kg
kg	% of Total	kg	% of Total		
Ag	<1.7E+3	79	<4.2E+2	21	2.2E+3
As	<1.5E+4	87	2.1E+3	13	1.7E+4
Ba	<1.5E+3	1	2.7E+5	99	2.7E+5
Cd	6.5E+3	92	<5.6E+2	8	7.2E+3
Cl	6.7E+5	100	0.0E+0	0	6.7E+5
Cr	1.3E+5	90	1.5E+4	10	1.5E+5
Cu	1.5E+3	7	2.2E+4	93	2.3E+4
F	1.8E+5	100	0.0E+0	0	1.8E+5
Fey	5.5E+3	0	4.7E+6	100	4.7E+6
Hg	3.6E+3	91	<3.7E+2	9	4.0E+3
Mn	3.6E+3	1	6.5E+5	99	6.6E+5
NO_2	9.4E+6	100	0.0E+0	0	9.4E+6
NO_3	3.1E+7	100	0.0E+0	0	3.1E+7
Pb	3.8E+4	94	2.3E+3	6	4.0E+4
Se	<1.6E+4	98	3.4E+2	2	1.7E+4
SO_4	7.0E+5	16	3.6E+6	84	4.3E+6
Zn	<3.4E+3	18	1.5E+4	82	1.9E+4

Hydrologic parameters - The hydrologic properties of the soil and gravel components in the system have been determined from conventional permeability tests and water retention tests. The soils were obtained from one split-spoon sampler coring at the grout disposal site. Data from additional cores will be obtained to supplement the database.

COMPUTER CODES

As stated earlier, initial performance assessment activities were based on relatively simple models. The release of contaminants from the grout inventory was modeled as either a diffusion- or solubility-controlled process. If a diffusion barrier was present, the release through the diffusion barrier was determined based on an analytical solution of the combined system, assuming pseudo-steady-state concentration profiles.

The release term was then plugged into a one-dimensional flow/transport code called TRANSS[9], which provided contaminant arrival profiles at the saturated zone. The contaminants in the saturated zone were transported to the receptor point using VTT[10]. Contaminant concentrations in the water at the receptor point were determined and doses to users were calculated for various scenarios using the codes DITTY[11] and MAXI[12].

The most recent analyses have used more sophisticated two-dimensional models for unsaturated flow and transport. The flow code used was TRACR3D, which was developed at Los Alamos National Laboratory[13] (LANL). This code was shown to be capable of solving flow problems that were prone to numerical instability due to the large differences in hydraulic properties of the various system components. Verification and validation of this code is in progress at LANL.

The unsaturated transport code selected was S-301, which was developed at the Winfrith Atomic Energy Establishment[14]. This code was selected over several other codes because it was shown to compare most favorably to both advection- and diffusion-dominated problems with analytical solutions.

Output from the unsaturated transport code is used to provide contaminant fluxes for modeling the aquifer. The flow and transport in the aquifer were analyzed using a three-dimensional code called SLAEM[15].

Finally, contaminant concentrations in the well water for the full garden scenario and contaminant fluxes into the

Columbia River are input to the radiological dose code GENII[16].

SENSITIVITY AND UNCERTAINTY

Areas of uncertainty primarily include assumptions regarding long-term performance of various components of the system. Some specific assumptions that have been made are that the grout properties remain stable for 10,000 years, the diffusion barrier remains effective in diverting advecting water from the grout over 10,000 years, non-isothermal characteristics of the disposal system will not considerably alter the performance of the system, and the infiltration barrier remains effective over 10,000 years. Experiments are being conducted to help support some of these assumptions.

Determining the sensitivity of the system performance to various parameters is essential to understanding the relative importance of various components of the system and to help defend long-term performance assessment. The sensitivity of results for the full garden scenario to variations in the effective diffusivity of species in the diffusion barrier, effective diffusivity of species in the grout, well location, and water infiltration rates is being analyzed. Results will be included in the next draft of the performance assessment document.

REFERENCES

1. J. M. ALLISON and T. B. BERGMAN, "The Hanford Grout Treatment Facility -Environmental Assurance in Low-Level Waste Disposal," Proceedings of the International Topical Meeting on Nuclear and Hazardous Waste Management, Spectrum '88, American Nuclear Society, Inc., La Grange Park, Illinois, pp. 130-132 (1988).
2. R. H. GUYMAN, L. D. VANSELOW, and G. D. CAMPBELL, "The Grout Treatment Facility--Processing Facilities for Low-Level Waste Immobilization and Disposal," Proceedings of the International Topical Meeting on Nuclear and Hazardous Waste Management, Spectrum '88, American Nuclear Society, Inc., La Grange Park, Illinois, pp. 95-98 (1988).
3. G. H. SEWART, W. T. FARRIS, D. G. HUIZENGA, A. H. McMAKIN, G. P STREILE, and R. L. TREAT, "Long-Term Performance Assessment of Grouted Phosphate/ Sulfate Waste from N Reactor Operations," PNL-6152, Pacific Northwest Laboratory (1987).
4. U.S. DEPARTMENT OF ENERGY, "Final Environmental Impact Statement: Disposal of Hanford Defense High-Level, Transuranic and Tank Wastes," DOE/EIS-0113, Vol. 3, U.S. Department of Energy (1987).
5. W. G. RICHMOND, "Methods and Data for Use in Determining Source Terms in the Grout Disposal Program," Westinghouse Hanford Company Engineering Data Transmittal 101203 (November 14, 1988).
6. R. J. SERNE, W. J. MARTIN, S. B. McLAURINE, S. P. AIRHART, V. L. LeGORE, and R. L. TREAT, "Laboratory Leach Tests of Phosphate/Sulfate Waste Grout and Leachate Adsorption Tests Using Hanford Sediment," PNL-6019, Pacific Northwest Laboratory (1987).
7. AMERICAN NUCLEAR SOCIETY, "Measurement of the Leachability of Solidified Low-Level Radioactive Wastes by a Short-Term Test Procedure," ANS 16.1, American Nuclear Society (1986).
8. J. E. MENDEL (editor), "Test Methods Submitted for Nuclear Waste Materials Handbook," PNL-3990, Rev. 6, Pacific Northwest Laboratory (1985).
9. C. S. SIMMONS, C. T. KINCAID, and A. E. REISENAUER, "A Simplified Model for Radioactive Transport: The TRANSS Code," PNL-6029, Pacific Northwest Laboratory (1986).
10. A. E. REISENAUER, "Variable Thickness Transient Ground-Water Flow Model," PNL-3160-2, Pacific Northwest Laboratory (1979).
11. B. A. NAPIER, R. A. PELOQUIN, and D. L. STRENGE, "DITTY - A Computer Program for Calculating Population Dose Integrated Over Ten Thousand Years," PNL-4456, Pacific Northwest Laboratory (1986).
12. B. A. NAPIER, R. A. PELOQUIN, W. E. KENNEDY, Jr., and S. M. NEUDER, "Intruder Dose Pathway Analysis for the ONSITE/MAXI Computer Program," NUREG/CR-3620 (PNL-4054), U.S. Nuclear Regulatory Commission (1984).
13. B. J. TRAVIS, "TRACR3D: A Model of Flow and Transport in Porous Fractured Media," LA-9667-MS, Los Alamos National Laboratory (1984).
14. R. S. WIKRAMARATNA and C. L. FARMER, "The S301 High Resolution Miscible Simulator 2-D Version with User-Specified Velocities," S301-2/1, Oil Recovery Projects Division, Winfrith Atomic Energy Establishment, Dorset, United Kingdom (1987).
15. O. D. L. STRACK, "Groundwater Mechanics," Prentice-Hall, Englewood Cliffs, New Jersey (1989).
16. B. A. NAPIER, R. A. PELOQUIN, D. L. STRENGE, and J. V. RAMSDELL, "Hanford Environmental Dosimetry Upgrade Project. GENII - The Hanford Environmental Radiation Dosimetry Software System," PNL-6584, Vols. 1-3, Pacific Northwest Laboratory (1988).