OPERATIONAL AND LONG-TERM PERFORMANCE ASSESSMENT FOR HANFORD GROUT

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ABSTRACT

The Department of Energy and Rockwell Hanford Operations are preparing for the construction of a Transportable Grout Facility to immobilize selected radioactive liquid wastes now stored at Hanford. Oak Ridge National Laboratory is supporting the program by developing initial grout formulations for the facility. Pacific Northwest Laboratory is verifying the required operational characteristics of the grout formulation and evaluating the long-term performance of the grouted waste form.

Preliminary assessments of the operational characteristics of the grout formulation show that the grout meets established criteria. Preliminary performance assessments indicate that the grouted waste form will provide long-term environmental protection. A series of laboratory and field tests are planned and ongoing to verify these assessments.

BACKGROUND AND INTRODUCTION

Plans have been made at Hanford to dispose of millions of gallons of low-level radioactive liquid waste by solidification in grout.[1] Grout is a mixture of liquid wastes and grout-forming solids that includes portland cement, fly ash, and clays. The mixture is designed to be efficiently processed, easily pumped, and subsequently hardened into a solid waste form that provides long-term environmental protection. The Transportable Grout Facility (TGF) will convert low-level liquid wastes to a grout slurry that can be pumped to shallow land sites for disposal. Once emplaced in the disposal sites, the grout will harden into massive blocks, effectively isolating the waste (Fig. 1).

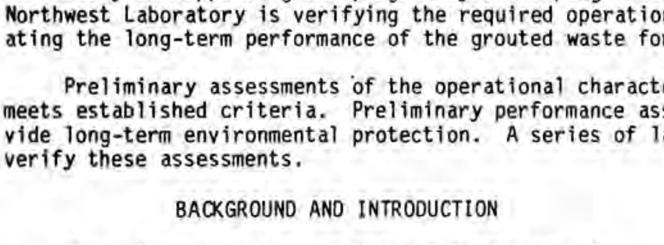

Fig. 1. Grout Monolith and Soil Barrier System for Disposal of Hanford Facilities Waste

High-pressure grout injection technology has been successfully applied at Oak Ridge National Laboratory during the last 20 years.[2] Grout technologies are also under development at Savannah River Laboratory in South Carolina[3] and in the Federal Republic of Germany. Cement is commonly used to solidify low-level wastes generated throughout the world by the commercial nuclear power industry.

Several Department of Energy contractors are involved in the TGF project at Hanford. Rockwell Hanford Operations manages the project and will be responsible for operating the facility. Oak Ridge National Laboratory (ORNL) is providing initial grout formulations. Pacific Northwest Laboratory (PNL) is working with ORNL and Rockwell Hanford Operations to verify that these formulations can be safely and reliably processed and that they will yield a solid grouted waste form which will provide adequate long-term environmental protection.

A variety of wastes have been identified as candidates for grouting. Hanford Facilities Waste (HFW) has been selected as the feed stream for initial operation of the TGF beginning in 1986. HFW is expected to be comprised of a blend of wastes including the wastes produced from reactor containment decontamination and from spent fuel storage pool filtration systems at the Hanford N-Reactor. This waste stream will contain about 250 microcuries of radioactivity per liter. Primary radionuclides in HFW are cesium-137, strontium-90, and cobalt-60. The reference chemical analysis for HFW is shown in Table I. ORNL has provided a preliminary grout formulation based on this analysis. The ORNL formulation is shown in Table II. PNL has conducted a preliminary evaluation of the operational characteristics of grout produced according to this formulation. PNL has also conducted a preliminary assessment of the performance of this grout over the long term. This paper is a discussion of the results of these evaluations to date.

TABLE I

Reference Composition of Hanford Facilities Waste (HFW)

Component	Molarity
Na_2SO_4	0.015
$NaNO_2$	0.016
Na_3PO_4	0.15

TABLE II

Reference Grout Formulation for HFW*

Grout-Forming Solids	Amount (wt%)
Type I-II-LA Portland Cement	41
Centralia, WA ASTM Class F Fly Ash	40
Attapulgite-150 Drilling Clay	11
Indian Red Pottery Clay	8

*7.5 pounds of grout-forming solids are mixed with each gallon of HFW (900 kg solids per m^3 of HFW).

ASSESSMENT OF OPERATIONAL CHARACTERISTICS OF HFW GROUT

Operating criteria must be satisfied by the reference grout formulation as well as by expected variations in the formulation to ensure acceptable performance of the mixing, pumping, and distribution systems of the TGF as currently designed. Variations may be caused by differences in the following components: 1) the chemical and physical properties of the waste and grout formers, 2) the proportions of the waste and grout-formers in the grout mixture, and 3) the degree of mixing achieved during grout mixing and pumping. For the grout formula to be acceptable, it must be shown that the formulation has sufficient flexibility to accommodate potential variations and assure reliable operations.

The operational characteristics of HFW grout were assessed on the basis of criteria relating to grout rheology, angle of flow, strength, temperature, and the development of free-standing liquid. A test matrix was established as the first step in assessing the effects of variability around the reference formula (see Table III). The test matrix included grouts whose individual solids proportions varied by ±10% around target levels. It was assumed that the range of process control limits would contribute ±5% variation. Variability in the properties of the solids was assumed to add an additional ±5% variation. The test matrix also included grouts whose grout-formers blend-to-liquid waste ratios varied by about ±6.0%. Sources of this variability include process control limits and variability in mixing efficiency.

Grout Operating Criteria and Conformance Testing

Grout operating criteria and results of conformance testing are presented below. These criteria were based on the requirement that grout disposal operations be safely and efficiently conducted. Conformance testing involved the grouts shown in Table III. These grouts were mixed for three minutes in a Hobart N50 mixer using a wire whisk and were subsequently exposed to a battery of tests to determine conformance to the operating criteria.

- The pressure required to pump grout should not exceed 250 psig (1725 kPa).

  The TGF pump is designed to pump grout at pressures up to 350 psig (2415 kPa). Operating plans limit the normal operating pressure to 250 psig (1725 kPa). The maximum pumping distance is expected to be 3000 ft (914 m), although for HFW grout, the disposal sites will be located a maximum of ~1500 ft (457 m) from the TFG. Therefore, the grout pumping pressure should not exceed 16.7 psi (115 kPa) per 100 ft (30.5 m).

  Pressure drops were determined for pumping grout through 1500 ft (457 m) of 2 inch (5 cm) pipe assuming that the rheology of the grout is described by the power law model. Calculations were performed using the equations in Smith, 1976.[4] Rheological data were obtained using a Haake Rotovisco RV100® viscometer with a M500 measuring/drive unit and MVI sensor. Pressure drop data, as a function of pump rate, are plotted in Fig. 2. This plot shows that maximum pressure drops for the grout over a 30 to 70 gal/min (0.11 to 0.27 m^3/min) pumping range are well below 16.7 psi (115 kPa) per 100 ft (30.5 m) of 2 in. (5 cm) pipe, and thus meet this operating criterion.

- The grout should be pumped under turbulent flow conditions.

  Experience gained through the cementing of oil wells has demonstrated the advisability of pumping grout under turbulent flow conditions to minimize the potential for plugging. A Reynolds Number (N_{Re}) of greater than 2100 generally defines the transition region where laminar flow ends and turbulent flow begins. Using the power law model and rheological data obtained from the Haake viscometer, Reynolds Numbers were determined for various flow rates (Fig. 3). This figure shows that the grout would be pumped in turbulent flow as required.

- The pressure required to restart the flow of grout after a 10 minute loss of power should not exceed 250 psig (1725 kPa).

  Provisions have been made to add flush water to the grout pipe at 500 psi (3450 kPa). Engineering experience dictates that the pressure required to restart flow should be lower (i.e., 250 psi or 1725 kPa) to accommodate unexpected conditions. This is equivalent to a gel strength of 100 $lb_f/100\ ft^2$ (48 Pa) in 1500 ft (457 m) of 2 in. (5 cm) pipe. The pressure required to restart flow was determined by measuring gel strength after 10 minutes using a Fann 35A® viscometer. For the reference grout formula and its variations, the restart pressure ranged from 30 to 100 psi (207 to 690 kPa), which is well below the 250 psi (1725 kPa) limit.

- The grout should flow at an angle that is compatible with the design of the disposal site and the system for distributing grout within the site.

  Although the reference grout is very fluid when in motion, its viscosity increases dramatically as flow gradually ceases. When flow does cease, a gel strength develops that must be overcome with a force if the grout is to resume flowing. This force is related to the pressure required to reinitiate flow of grout in a pipe. The angle of flow of grout was measured while slowly pouring a large quantity of grout (130 gal. or 0.49 m^3) into a trench. The angle of flow was measured when 50%, 75%, and 100% of the grout was poured. Figure 4 shows that the grout angle of flow increased as additional grout was poured to a final angle of 3° above horizontal. Because there was no evidence that the rate of angle development was diminishing, it was concluded that the terminal angle of flow of the grout is probably greater than 3°. Additional tests are planned to more accurately determine the flow profile of grout in a disposal site.

TABLE III
Test Matrix for Hanford Facilities Waste (HFW) Grout

Grout Forming Solids*	Dry Blend Number						
	1	2	3	4	5	6	7
	(weight percent)						
Type I-II-LA Portland Cement	41	41	41	43	38	42	39
Centralia, WA ASTM Class F Fly Ash	40	41	39	38	42	38	42
Attapulgite-150 Drilling Clay	11	10	12	10	12	12	10
Indian Red Pottery Clay	8	8	8	9	8	8	9

*Grout forming solids mixed with HFW in ratios between 7.0 and 8.5 pounds per gallon (840-1020 kg/m^3).

Tradename of Haake Buchler Instruments, Inc., Saddlebrook, New Jersey

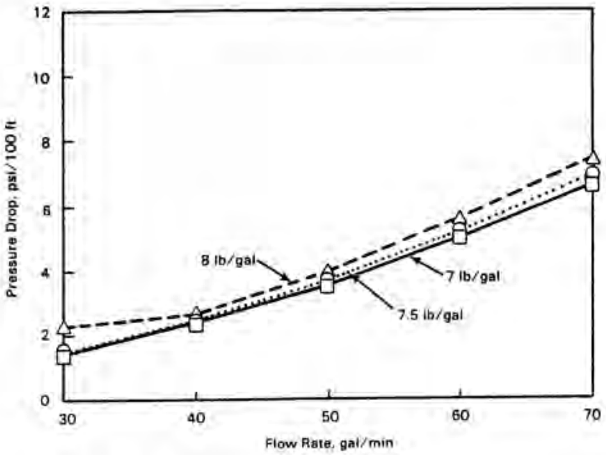

Fig. 2. Pressure Drop vs Flow Rate

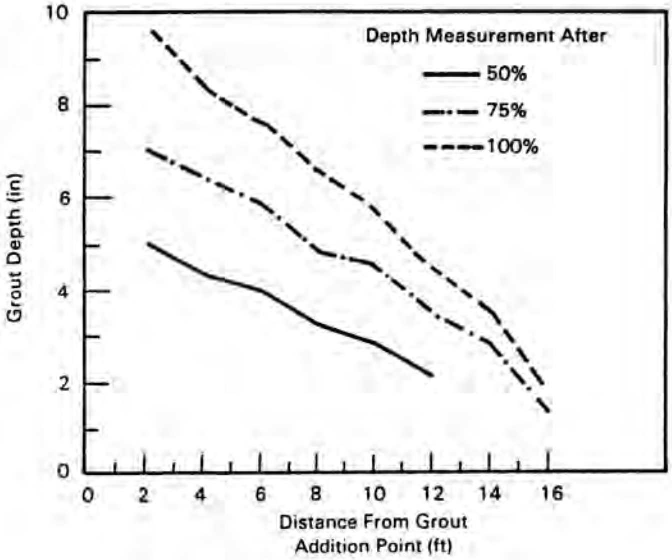

Fig. 4. Grout Depth vs Flow Distance

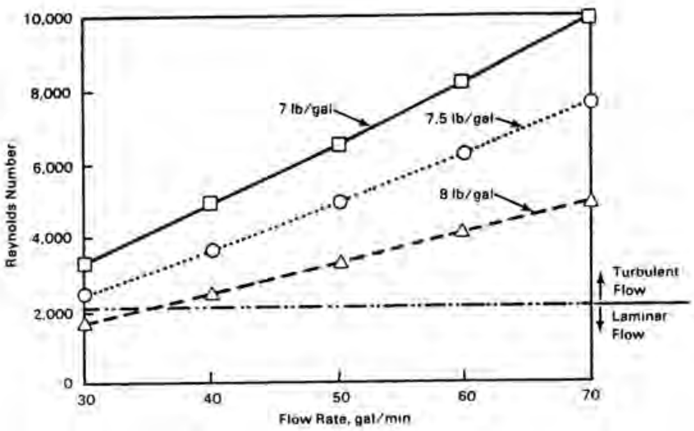

Fig. 3. Pumpability vs Flow Rate

- <u>The grout should develop a compressive strength of at least 50 psig (350 kPa) within 28 days.</u>

A compressive strength of 50 psi (350 kPa) is sufficient to ensure that the grout will not be crushed by its own weight and by that of a future 16 ft (5 m) thick barrier of soil. Compressive strength is measured using the ASTM C109 test. After 28 days, the compressive strength ranged from about 200 to 650 psi (1380 to 4480 kPa). The strength of the grout will increase somewhat as cement reactions continue.

- <u>The grout should reabsorb or promote evaporation of the free-standing liquid within 28 days.</u>

The objective of grouting is the disposal of liquid wastes in a solid waste form. Thus, it is undesirable for liquid waste to remain in the disposal site. Normally, a small amount of free-standing liquid develops on the surface of grout within a few minutes of pouring. The volume of liquid reaches its maximum within about 2 days. The liquid is then largely or completely reabsorbed as hydration reactions within the interior of the grout draw water inside. During the setting process, the grout must be maintained under totally humid or immersed conditions to minimize the formation of cracks. Even after the grout has set and cured, humid conditions must be maintained to prevent cracking.

The reference grout yielded no free-standing water after 28 days although grout mixed with 7 pounds of solids blend per gallon (840 kg/m^3) typically demonstrated about 2% free-standing liquid. If free-standing liquid is present on the grout, it must be removed and recycled through the TGF for disposal or be treated with other means.

- At no time shall the temperature of the grout exceed the boiling point of the liquid contained within the grout pores.

Boiling must be prevented to avoid a dangerous buildup of pressure inside the grout mass. Two sources of heat exist in the grout: reaction heat, largely from the hydration of cement, and heat from the decay of radionuclides. For HFW grout, decay heat is inconsequential due to the very low levels of heat-producing radionuclides in the waste and the moderately good thermal conductivity of grout and the surrounding soil that allows heat to diffuse. The heat of hydration released when portland cement reacts with water can be significant. Assuming worst case adiabatic conditions, the maximum temperature reached at the center of the casting is about 78°C (172°F). This is safely below the boiling point of the pore liquid (~121°C or ~250°C). The potential for other exothermic reactions between the waste and grout formers is also being investigated. Little, if any, effect of other exothermic reactions is expected, however.

Discussion of Predicted Operating Impacts

The results of the operational studies on HFW grout show that the grout formulation yields a grout that conforms to the criteria for acceptable operability. The analyses in this study were based on laboratory-scale tests. To add confidence to the conclusions reached in this study, large-scale tests involving a 1/4-scale grout process system are being conducted. An important objective of large-scale testing is to refine the predictability of laboratory procedures so that laboratory testing can be applied with confidence using samples of actual waste.

LONG-TERM PERFORMANCE ASSESSMENT

The second phase of the evaluation of the HFW grout formulation involved an assessment of the long-term environmental protection provided by the disposed grout. The primary performance objective of a waste disposal system is to isolate from human contact radionuclides and hazardous chemicals that occur in biologically significant concentrations and, thus, provide for the long-term protection of public health and safety. The radiological performance of disposed HFW grout was assessed by evaluating the effects of leaching by percolating groundwater and the potentially disruptive effects of indigenous plants and animals which live over the grout disposal site. The radiological effects were evaluated over a 1,000 year period. Certain low probability events dismissed from consideration include meteorite impact, magmatic activity, flooding, and magnitude 7 or greater seismic events. The effects of percolating groundwater and indigenous plants and animals are analyzed below.

- Waste Migration Through Groundwater Recharge

Water percolating through the waste site could cause radionuclides to move slowly from the site and into the groundwater where it would be available for removal via a well. The quantity of water available for percolation (recharge water) is dependent on the climate and the vegetation over the waste site. A conservative water recharge rate of 5 cm/year was selected for the assessment of this event.(5) This recharge rate is based on a hypothetical change to a wetter climate (50% increase in precipitation). Retardation coefficients (K_d) of 26 and 0.39 were assumed for cesium-137 and strontium-90, respectively, the two significant radionuclides in HFW. Drinking and irrigation water was assumed to be withdrawn from the Columbia River and a well located 2 km down gradient from the disposal site for comparison to regulatory limits. For this event, the potential maximum one year radiation dose to an individual was calculated to be $3x10^{-9}$ rem. This extremely low dose would occur in the year 2770 A.D. and would result primarily from exposure to strontium-90. For perspective, the average annual dose due to exposure to naturally-occurring radiation (0.1 rem) is 30 million times higher. Moreover, the actual water recharge rate at Hanford under current climatic conditions does not exceed 0.5 cm per year. At this rate, percolating water does not reach the water table for about 1,000 years. By this time, cesium-137 and strontium-90 have decayed to innocuous levels.

- Biotic Transport

Biotic transport of waste would involve the penetration of the waste disposal site by indigenous plants and animals. Wastes would subsequently be brought to the surface where they could cause direct exposure and/or lead to ingestion of contaminated foods. The depth of plant and animal intrusion in an arid environment such as at Hanford is generally about 2 m. Significant mobilization of the HFW should be prevented by the 5 m of soil cover over the grout. Therefore, no radiological doses are projected for this event.

Discussion of Long-Term Environmental Protection

Radiological doses received as a result of percolating groundwater and actions of indigenous plants and animals are extremely small. Chemical hazards are currently being assessed in a follow-up effort. Due to the low concentrations of relatively innocuous chemical wastes present in HFW, however, no significant impacts are expected.

CONCLUSIONS

Grout produced according to a preliminary grout formulation for HFW was shown to meet criteria for acceptable processability and provide long-term radiological safety. Although additional study is required to verify operational performance on a large scale and to address chemical waste issues, it is expected that grout will be proven to provide a safe, effective means of disposal for HFW.

ACKNOWLEDGEMENT

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