PHYSICAL INTERACTIONS BETWEEN HANFORD SINGLE-SHELL TANK WASTES AND FILL MATERIAL

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ABSTRACT

Spent fuel reprocessing operations at the U. S. Department of Energy Hanford Site in the period 1944 to 1972 generated radioactive waste currently stored in 149 underground single-shell tanks. One proposed method, in-place stabilization, for final disposal of the wastes in the single-shell tanks involves filling the tanks with basalt gravel to support the tank dome, followed by emplacement of a specially constructed earth barrier over the tank farms. Well-established soil mechanics instruments and procedures are applicable to the study of possible physical interactions between radioactive waste in Hanford Site single-shell tanks and fill (basalt gravel) material. Consolidation tests with simulated sludge waste indicate that 90% of the final consolidated volume at expected gravel loads will be attained a few days after filling the tank.

INTRODUCTION

Spent fuel reprocessing operations at the U. S. Department of Energy Hanford Site in the period 1944 to 1972 generated radioactive waste currently stored in 149 underground single-shell tanks. (All single-shell tanks at the Hanford Site have now been removed from service; double-shell tanks are now used to contain liquid high-level wastes.) This waste consists of sludges (hydrated metal oxides and hydroxides) and salts that crystallized when concentrated liquid wastes cooled. One proposed method, in-place stabilization, for final disposal of the wastes in the single-shell tanks involves filling the tanks with basalt gravel to support the tank dome, followed by emplacement of a specially constructed earth barrier over the tank farms.[1]

When basalt gravel is added to single-shell tank wastes, both chemical and physical interactions may occur. The latter may involve consolidation of waste from the weight of gravel on top of it or settling of the gravel through the waste to the bottom of the tank or a combination of the two major interactions. Settling of the gravel to the bottom of the tanks would result in a stable configuration suitable for supporting the tank dome when structural failure eventually occurs. In the case of waste consolidation, structural failure could cause the waste to consolidate farther; the net results could be a breach of the engineered barrier placed on top of the tank.

This paper discusses preliminary results of experiments conducted with simulated single-shell tank waste to investigate physical interactions between waste and basalt gravel.

SIMULATED SINGLE-SHELL TANK SLUDGES

The Hanford single-shell tanks contain solid radioactive waste resulting from neutralization and/or evaporation of neutralized high-level waste (HLW) produced by the three different processes [Bismuth Phosphate ($BiPO_4$), Redox, and PUREX] used to recover plutonium and uranium from irradiated uranium metal. Neutralized HLW consists of a solid (sludge) phase and a supernatant liquid phase. The sludge phase -- mainly hydrated oxides of iron and other metals which precipitated when the NaOH was added to the HLW -- settled to the bottom of the single-shell tanks. The liquid phase has been evaporated to produce solid waste (mainly $NaNO_3$) which, in some cases, has been deposited on top of the sludge.

Table I lists the composition of the simulated sludges used to obtain the results reported in this paper. These compositions are based upon current best estimates of the amounts and types of materials present in the various sludges.

TABLE I
Compositions of Simulated Single-Shell Tank Sludges

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	Composition, Wt%		
Component	Sludge 1 [(a)]	Sludge 2 [(b)]	Sludge 3 [(c)]
$Al_2O_3 \cdot xH_2O$	57.6	9.5	(d)
$NaNO_3$	17.5	11.2	17.3
$Fe_2O_3 \cdot xH_2O$	6.4	21.3	11.2
H_2O	5.0	50.0	50.0
SiO_2	4.7	3.1	5.4
$Cr_2O_3 \cdot xH_2O$	4.4	(d)	1.7
$Ca(OH)_2$	2.1	1.5	(d)
$Mn(OH)_2$	2.0	1.5	(d)
MnO_2	0.8	1.7	(d)
Na_2SO_4	0.5	0.3	0.3
$BiPO_4$	(d)	(d)	13.3
NaF	(d)	(d)	0.8

(a) Representative of sludge produced from Redox process HLW.
(b) Representative of sludge produced from PUREX process HLW.
(c) Representative of sludge produced from $BiPO_4$ process HLW.
(d) Not present.

CONSOLIDATION TESTS

Approach

Well-known soil mechanics theories and procedures[2,3] are applicable to estimating the extent waste in single-shell tanks will be compressed by the weight of basalt gravel placed on top of them. For example, soil consolidation theories developed to predict the extent buildings constructed on a site underlain by clay strata will settle are applicable to consolidation of wastes in tanks. Clays are finely textured, and water flows very slowly through clay strata. Sludges in the single-shell tanks are also finely textured; procedures and equipment used to measure and/or predict consolidation of clay strata ought also to be applicable to prediction of consolidation of radioactive sludges.

Consolidation of sludges in single-shell tanks will result in the expulsion of liquids from the pores of the sludge solids. Because of the low permeability of the fine textured sludge, the initial load is borne by the interstitial liquid radioactive waste. The load is then slowly transferred to the solid particles causing them to reorient to a more dense material as the interstitial liquid transfers to the gravel. A coefficient of consolidation which predicts the time required to consolidate wastes when a load is applied to sludge confined in a waste tank can be measured by applying known loads to samples of various sludges. A typical bench-scale consolidation test apparatus is shown in Fig. 1.

Fig. 1. Apparatus Used to Measure Consolidation of Simulated Single-Shell Tank Sludges

Test Results

Figures 2 and 3 show results of consolidation tests with the three simulated sludges of the compositions shown in Table I. These data were obtained with a Model S 2855 Pneumatic Digicon Consolidation Apparatus (Geotest Instrument Corp.). This instrument is designed for pneumatic loading of 6.35 cm diameter, 2.54 cm thick samples. The sample is confined in a Teflon-lined (E. I. du Pont de Nemours Co.) load cell fitted at both top and bottom with a sintered bronze disc for drainage of liquids. During a consolidation test, the valve at the bottom of the test cell is closed so that liquids can drain only from the top. The load on the sample is applied incrementally with each load being twice the previous load.

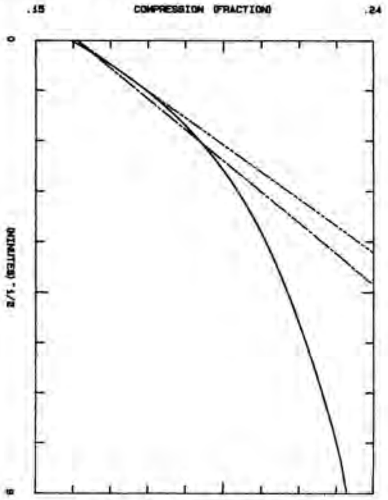

Fig. 2. Consolidation Data for Type 2 (Table I) Sludge

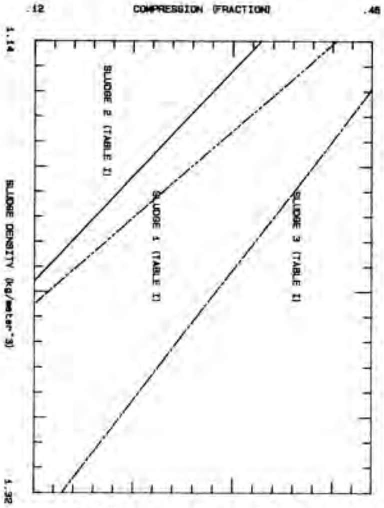

Fig. 3. Consolidation of Simulated Single-Shell Tank Sludges as a Function of Their Density

The load expected in single-shell tanks filled with basalt gravel is about 140 kPa. If structural failure occurs (i.e., dome collapse) and the present soil overburden settles onto wastes in the tanks, the total load will be about 280 kPa. These loads were used as reference points to establish starting initial experimental loads. Successive loads were each one-half of the previous load until the lower limit of the consolidation test apparatus was reached. Each load was applied for a minimum of eight hours.

Consolidation tests were performed at 333 K (60°C) and 363 K (90°C) as well as at approximately 298 K (approximately 25°C). Tests were performed at the higher temperature to simulate the insulating effect of the basalt gravel fill material and engineered barriers over the tanks.

The data from such tests were used to determine the time required for 90% of the total consolidation for each type of sludge. A consolidation coefficient was also calculated. Figure 2 depicts the method used to calculate the consolidation coefficient. The compression of the sample is plotted versus the square root of time (the solid line in Fig. 2). A straight line tangent to the initial part of the compression curve is plotted (the double-dashed line in Fig. 2) and its slope determined. A second straight line is then plotted (the single-dashed line in Fig. 2) using the slope of the tangent line divided by 1.15 (slope/1.15). The intersection of the second straight line and the sludge compression line gives the point of 90% of total consolidation. From the time involved to reach 90% consolidation, the consolidation coefficient is calculated. Using the consolidation coefficient and the compression fraction associated with 90% consolidation, the amount of settling and the time involved can be predicted.

The extent of consolidation of each type of sludge is approximately a linear function (Fig. 3) of the density. The denser the sludge, the less it compresses. Table II lists equations which express the fraction (Y) of original sludge depth lost by consolidation as a function of the density of the sludge.

TABLE II

Consolidation of Single-Shell Tank Sludge Wastes

Predictor Equations

Consolidation Predictor

Sludge Type	Equation[a]
Redox Process	$Y = 3.72 - 2.98x$
PUREX Process	$Y = 3.06 - 2.38x$
$BiPO_4$ Process	$Y = 2.71 - 1.94x$

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[a] Y = Percent of original sludge depth lost by consolidation

x = Sludge density.

Higher consolidation temperatures do not affect the extent of consolidation of single-shell tank sludges but do accelerate the process (Fig. 4). At higher temperatures, the viscosity of interstitial fluids is decreased, and they flow faster through the pores of the sludge. The solid curved line in Fig. 4 is a plot of compression at ambient temperatures (298 K); the double-dashed line at 333 K (60°C); the single-dashed line at 363 K (90°C). The solid horizontal line at compression fraction 0.197 intersects each curve at approximately 90% of total consolidation.

Typically, the sludge in a Hanford Site single-shell waste tank has a density of 1.6×10^{-3} kg L^{-1}; the depth of the sludge layer is about 1.3 m. From the results presented in Figs. 2-4 and in Table II, 90% (0.21 m) of the total sludge consolidation will occur in 60 days; about 213 days will be required for 99% of the total consolidation to take place.

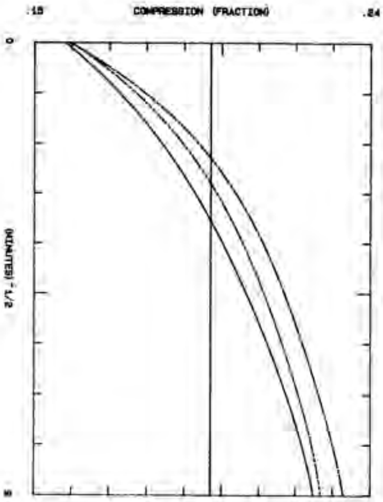

Fig. 4. Effect of Temperature on Consolidation of Simulated Sludge 2 (Table I)

Consolidation of Salt Cake Wastes

As noted previously, sludges in many single-shell tanks are overlain by salts precipitated when the alkaline liquid in the original HLW was evaporated and cooled. Tests are currently in progress to measure the consolidation of simulated salt cakes under applied loads. It is anticipated that, contrary to results with sludges, salt cakes will consolidate as soon as loads are applied. Instantaneous consolidation of salt cakes is expected because the large size of salt cake crystals will allow rapid movement of interstitial liquid through solid pores. Thus, applied loads will be borne directly by crystals of salt cake and not by interstitial liquids.

It is also anticipated that the extent of consolidation of salt cake will increase with increased temperature which will occur when layers of soil (engineered barriers) are placed over single-shell tanks. At increased temperature, the interstitial liquids will dissolve some of the salt cake and will thus reduce the volume (height) of the salt cake layer.

PENETRATION OF GRAVEL INTO WASTE

It is planned to use a permeameter such as those used in soil mechanics laboratories to study the extent to which basalt gravel will settle through the salt cake and/or sludge to the bottom of the tank. The permeameter will be used to determine the rate at which sludge or salt cake flows through a confined column of gravel as a function of the pressure across the columns. These measurements will permit calculation of both the yield point of the waste and its hydraulic conductivity. The yield point is the pressure at which plastic materials begin to flow. Hydraulic conductivity data will allow predictions to be made concerning the extent to which basalt gravel will settle through salt cake or sludge.

CONCLUSIONS

Well-established soil mechanics instruments and procedures are applicable to the study of possible physical interactions between radioactive waste in Hanford Site single-shell tanks and fill (basalt gravel) material. Consolidation tests with simulated sludge waste indicate that 90% of the final consolidated volume at expected gravel loads will be attained a few days after filling the tank.

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