Rheology of Savannah River Site Tank 42 HLW Radioactive Sludge

by

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DOE Contract No. DE-AC09-96SR18500

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**Keywords: DWPF, Rheology,
Sludge, Viscosity.**

**WSRC-RP-97-236
Rev. 0**

Retention Period: Permanent

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Waste Vitrification &
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STI, 703-43A**

MEMORANDUM

November 5, 1997

TO: J. R. Harbour, 773-43A

FROM: B. C. Ha, 773-43A *BEK*

**RHEOLOGY OF SAVANNAH RIVER SITE TANK 42 HLW
RADIOACTIVE SLUDGE**

SUMMARY

Knowledge of the rheology of the radioactive sludge slurries at the Savannah River Site (SRS) is necessary in order to ensure that they can be retrieved from waste tanks and processed for final disposal. At Savannah River Site (SRS), Tank 42 sludge represents one of the first HLW radioactive sludges to be vitrified in the Defense Waste

Processing Facility (DWPF). The rheological properties of unwashed Tank 42 sludge slurries at various solids concentrations were measured remotely in the Shielded Cells at the Savannah River Technology Center (SRTC) using a modified Haake Rotovisco viscometer.

The results indicated that the radioactive Tank 42 sludge slurries could be easily adjusted to meet the yield stress and viscosity limits of the DWPF process feed design basis. The DWPF requires a sludge feed slurry with a weight percent solids in the range of 13 to 19. The rheological properties of Tank 42 waste sludge at this weight percent solids range are within the yield stress design limits of 25 to 100 dynes/cm² and viscosity design limits of 4 to 12 centipoises.

The Bingham model parameters (yield stress and consistency) were correlated to a function of the insoluble solids content of Tank 42 sludge slurry. Results indicated that the critical insoluble solids content of Tank 42 sludge is 37 wt% on a dry basis.

INTRODUCTION

Knowledge of the rheology of the radioactive sludge slurries at the Savannah River Site (SRS) is necessary in order to ensure that they can be retrieved from waste tanks and processed in the Defense Waste Processing Facility (DWPF) for final disposal. The rheological properties of SRS Tank 42 and Tank 51 HLW radioactive waste sludges were investigated in the Shielded Cells at the Savannah River Technology Center (SRTC). Results of Tank 51 sludge have been reported previously.¹ The results determined whether the first radioactive waste sludge major batch can be delivered and processed by the current DWPF process equipment design. The data also formed a basis for validating experimental data of other studies with simulated sludges.

The high activity radioactive wastes stored as caustic slurries in tanks resulted from the neutralization of acid waste generated from

production of defense nuclear materials. As these wastes settle, they separate into an upper supernate layer and lower sludge layer. In the DWPF, the radionuclides from the sludge and supernate will be immobilized into borosilicate glass for long term storage and eventual disposal. In the DWPF, the sludge will initially be processed in the Sludge Receipt and Adjustment Tank (SRAT). The processed sludge slurry will then be transferred to the Slurry Mix Evaporator (SME) where frit will be added to form melter feed.

Tank 42 and Tank 51 radioactive sludges represent the first batches of HLW sludge to be processed in the DWPF. The rheological properties of Tank 42 and Tank 51 HLW radioactive sludges helped to verify DWPF equipment design basis, particularly process cell pump design. The rheological properties of the waste sludge also control mixing of glass frit and sludge to produce a homogeneous melter feed. This study shows that Tank 42 and Tank 51 waste sludges can be delivered to the DWPF, to be pretreated and vitrified. The rheology properties of Tank 42 radioactive sludge are presented in this paper. The results are successfully correlated to a function of the insoluble solids content.

EXPERIMENTAL SETUP

The rheological properties of Tank 42 sludge were measured using a Haake Rotovisco RV-12 concentric cylinder rheometer consisting of an M150 measuring drive unit and TI sensor system. The viscometer used the Searle technique, wherein the inner cylinder rotates and the outer cylinder remains stationary.

The shear stress was measured while the shear rate was varied from 0 to 300 sec^{-1} and back to 0 (using a Haake PG 142 programmer). The shear stress data of the samples were collected using an Apple Macintosh II containing a Strawberry Tree Computers' data acquisition system and the Strawberry's Analog Connection software. The shear stress and viscosity were calculated using Microsoft Excel spreadsheet software. The Haake viscometer was setup as follows:

Measuring Head: M150
Sensor System: TI
Final RPM: 128

Instrument Constants.

M = 2.34 cm/min.

G = 412 pascals•scale-grade•min/10⁻³

A = 0.966 pascals/scale-grade

The Haake viscometer was checked using 48.3 cp and 93.3 cp viscosity standards. The viscosity error was within $\pm 3\%$.

Rheological properties of the unwashed Tank 42 radioactive sludges were measured as a function of weight percent solids. Twenty five liters of this sludge were obtained from the SRS Tank and delivered to SRTC. Pristine (unwashed) sludge was used as the initial sample. The weight percent solids were adjusted by dilution with water or by concentration through drying. The slurry was stirred and then a measurement was made within five minutes at $23 \pm 1^\circ\text{C}$, the ambient temperature in the Shielded Cells. Temperature was monitored by a thermocouple during the experiments and did not increase more than 1°C before and after the measurements during the entire course of the experimental work. The process was repeated at least three times, before measurements of another slurry mixture were attempted. Effects of settling became significant, especially for high solids concentration slurries, 34 wt% solids or more, if the measurements are longer than five minutes.

The yield stress and consistency of the sludge slurries were determined by assuming a Bingham plastic fluid model. The general shape of the sludge slurry flow curve data supports the Bingham plastic fluid approximation (Figure 1). Yield stress and consistency

Figure 1. Tank 42 Sludge Containing 34 wt% Total Solids Behaves as a Bingham Plastic Fluid.

were determined using the intercept and slope of the flow curves, respectively. Figure 2 shows the apparent viscosities as a function of shear rates for Tank 42 HLW radioactive sludge slurry containing 34 wt% total solids.

DISCUSSION OF RESULTS

The current design bases for the DWPF sludge slurry properties are shown below:

Washed Sludge Slurry (DWPF Feed):

Solids Content, weight %:	13 - 19
Yield Stress, dynes/cm ² :	25 - 100
Consistency, centipoise:	4 - 12

Figure 2. Rheology Behavior of Tank 42 Sludge Containing 34 wt% Total Solids.

SRAT Slurry:

Solids Content, weight %: 18 - 25

Yield Stress, dynes/cm²: 15 - 50

Consistency, centipoise: 5 - 12

Rheological Properties of Tank 42 Sludge

Rheological properties of Tank 42 and Tank 51 radioactive sludge were measured as a function of weight percent total solids to ensure that the first DWPF radioactive sludge batch can be pumped and processed in the DWPF with the current design bases. The Tank 42 sludge sample, as received, contained 27 wt% total solids. The weight

percent total solids were adjusted by dilution with water or by concentration through drying.

For Tank 42 sludge, the yield stresses were 10, 11, 63, 90 and 381 dynes/cm² at 8, 17, 23, 27 and 34 weight percent total solids, respectively. The apparent viscosities were 10, 20, 35, 45 and 150 centipoises at 300 sec⁻¹ shear rate, respectively. These results agreed with results of non-radioactive sludge simulants. For example, a 15 wt% total solids non-radioactive sludge simulant has a yield stress of 12 dynes/cm², and an apparent viscosity of 12 centipoises at 300 sec⁻¹ shear rate. Table 1 shows a summary of results from rheological measurement of Tank 42 radioactive sludge.

	Wt. % solids	Yield Stress dynes/cm ²	Apparent Viscosity cp @ 300 sec ⁻¹
Tank 42 Sludge	8	10	10
Tank 42 Sludge	17	11	20
Tank 42 Sludge	23	63	35
Tank 42 Sludge	27	90	45
Tank 42 Sludge	34	381	150
non-radioactive sludge	15	12	12

Table 1. Rheological Properties of Tank 42 Sludge as a Function of Total Solids Concentration.

The above results indicated that sludge feed slurries could be easily adjusted to meet the yield stress and viscosity limits of the DWPF process feed design basis. The DWPF requires a sludge feed slurry within the weight percent solids range of 13 to 19. And, the rheological properties of 42 radioactive sludge at this weight percent solids range are within the yield stress limits of 25 to 100 dynes/cm² and viscosity limits of 4 to 12 centipoises.

Rheological properties of SRS Tank 21 radioactive sludge has been studied previously, as reviewed in reference 1. It was found that below 14 wt% insoluble solids, the yield stress of Tank 21 sludge was less than 40 dynes/cm², and the consistency was less than 10

centipoises.¹ This is consistent with what has been observed with Tank 51 sludge containing less than 18 wt% total solids.¹ Tank 42 sludge containing less than 17 wt% total solids, or 13 wt% insoluble solids, had only a quarter of the yield stress, 10 dynes/cm², but twice the consistency, 20 centipoises.

It was also found that, at 44 volume % total solids (or 17 wt% insoluble solids), the yield stress of Tank 21 radioactive sludge was in the range of 80 to 120 dynes/cm², and the consistency was from 15 to 20 cp.¹ The yield stress results compared very well to those of Tank 51 sludge.¹ However, Tank 42 sludge has a higher consistency, but a lower yield stress at a comparable solids concentration. Both, Tank 51 and Tank 21 radioactive sludges have been washed, but Tank 42 sludge was not washed at the time the rheology was measured. The differences in their rheological properties can be attributed to the sludge washing process.

During the Extended Sludge Processing (ESP), the concentrations of sodium, nitrite, and nitrate in the DWPF sludge feed are reduced to the appropriate levels to ensure the processability of the glass and to minimize NO_x emissions and production of ammonium nitrate in the vent system. Hay and Bibler² reported that the settling rate of Tank 42 sludge increased as density of the wash water decreased with each wash cycle. The higher consistency of Tank 42 sludge slurry compared to those of Tank 51 and 21 washed sludges is due to the higher density of the supernate. Viscosity was correlated to a function of the square root of density by Thomas *et. al.*^{3,4} for associated and nonassociated liquids. Although the viscosity model^{3,4} was developed for a liquid mixture rather than a solid-liquid slurry, it indicates that density has an additive effect on the consistency of the unwashed sludge compared to the washed sludge slurries.

Furthermore, the morphology of the wash sludge slurries was also changed during the wash cycles of the ESP process. Lower density and better packing of solid particles in the settled washed sludge

slurries increases the settling rate as indicated in Hay and Bibler's paper², and the yield stress as evidenced from the lower yield stress of Tank 42 unwashed sludge compared to washed Tank 51 and Tank 21 sludge slurries.

Rheological Model of Tank 42 HLW Radioactive Sludge

Marek⁵ modeled rheological properties of simulated SRS vitrification feeds using Bingham model parameters of yield stress and consistency correlated as a function of the insoluble solids content of the slurry. The theoretical model describes the apparent viscosity of a variety of slurries:⁶

$$\tau = \frac{e^{b_1 * C}}{(1 - \frac{C}{C_{max}})}$$

$$\mu = \frac{e^{b_2 * C}}{(1 - \frac{C}{C_{max}})}$$

where τ = yield stress

μ = consistency

C = insoluble solids concentration, wt%

C_{max} = critical or maximum insoluble solids concentration

b_1, b_2 = empirical parameters

These equations model the reduced viscosity of solids in liquid suspension as a function of the volume fraction of insoluble solids. Insoluble solids concentration is used instead of volume fraction of insoluble solids, since it is a more convenient analytical procedure.

The same rheological model was used to approximate the behavior of Tank 42 HLW radioactive sludge. The correlating equations with the resultant parameters were as follows:

$$\tau = \frac{e^{0.18 * C}}{(1 - \frac{C}{37.3})}$$

with $R^2 = 0.99$

$$\mu = \frac{e^{0.14 * C}}{(1 - \frac{C}{37.3})}$$

with $R^2 = 0.98$

Agreement of the correlation with the experimental data are shown in Figures 3 and 4. The model indicated that the critical insoluble solids concentration of Tank 42 sludge, as received unwashed, is about 37.3 wt%, which corresponds to 47.8 wt% total solids.

The correlation can be used to predict the maximum insoluble solids content of the SRAT and SME vessels containing Tank 42 sludge, as received unwashed. The yield stress design limit of the SRAT process slurry is 50 dynes/cm². For unprocessed Tank 42 sludge slurry in the SRAT vessel to be transferred out, the insoluble solids content has to be less than 18 wt%, or the total solids concentration less than 23 wt%. One of the goals of the SRAT processing cycle is to reduce the viscosity of the melter feed. The processed SRAT Tank 42 sludge slurry product will have a lower viscosity than the unwashed Tank 42 at the same weight percent solids.

Also, the yield stress design limit of the SME process slurry is 150 dynes/cm². To be transferred out of the SME, the insoluble solids content of an unprocessed Tank 42 sludge slurry has to be less than

Figure 3. Yield Stress Dependency on Insoluble Solids Content (Tank 42 HLW Radioactive Sludge).

23 wt%, or the total solids concentration less than 29 wt%. However, during the SME processing cycle, glass-forming frit will be added to prepare a melter feed. Addition of Frit 202 to Tank 51 sludge did not increase the yield stress and viscosity quite as much as increasing the sludge solid content.¹ It is believed that the finer particle sizes and larger particle size distribution of the sludge slurry has a larger effect on rheological properties than Frit 202. Even in these unlikely scenarios, the unprocessed Tank 42 radioactive sludge can still be pumped out of major process vessels if the solids content is adjusted.

Figure 4. Consistency Dependency on Insoluble Solids Content (Tank 42 HLW Radioactive Sludge).

CONCLUSIONS

Tank 42 HLW radioactive sludge will be part of the first batch of sludges to be processed in the DWPF. The rheological properties of Tank 42 and Tank 51 radioactive sludges indicated that sludge slurries can be retrieved and transferred to the DWPF for pretreatment and vitrification. The yield stresses of Tank 42 and Tank 51 sludge slurries fall within the DWPF equipment design basis¹ when the sludge slurries are adjusted so that the solids concentration are within the design limits. The apparent viscosity also falls within the DWPF design basis for sludge consistency for the design solids concentration range.

The rheological behavior of Tank 42 HLW radioactive sludge was approximated using a Bingham plastic model of the reduced viscosity of solids in liquid suspension as a function of the volume fraction of insoluble solids. The correlation indicated that the asymptotic insoluble solids concentration of Tank 42 sludge, as received unwashed, is 37 wt%.

ACKNOWLEDGMENTS

The author would like to thank the personnel of the SRTC Shielded Cells Operations for performing these experiments. I am especially thankful for R. W. Blessing, J. T. Mills, and R. H. Galloway for their help in setting up and operating the lab-scale apparatus. Their outstanding skills in operating the manipulators were essential to the success of these experiments.

This paper was prepared in connection with work done under Contract no. DE-AC09-89SR18035 with the U. S. Department of Energy.

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