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Physical Characterization of Radioactive Sludges in Selected Melton Valley and Evaporator Facility Storage Tanks

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OPERATED BY
MARTIN MARIETTA ENERGY SYSTEMS, INC.
FOR THE UNITED STATES
DEPARTMENT OF ENERGY

ORNL/TM-11653

Chemical Technology Division

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*Experimental work described in this report was performed by R. N. Ceo while he was a member of the Analytical Chemistry Division. His present affiliation is the Y-12 Plant, Oak Ridge, TN.

Date Published - October 1990

Prepared for the
Office of Defense Waste and Environmental Restoration
(Activity No. GF 73 01 01 0)

Prepared by the
OAK RIDGE NATIONAL LABORATORY
Oak Ridge, Tennessee 37831
operated by
MARTIN MARIETTA ENERGY SYSTEMS, INC.
for the
U.S. DEPARTMENT OF ENERGY
under contract DE-AC05-84OR21400

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ABSTRACT

Physical measurements were performed on typical radioactive sludge samples from selected Melton Valley Storage Tanks (MVSTs) and evaporator facility storage tanks at ORNL. These measurements included viscosity, particle size, density, sedimentation rate, and solids content. The techniques developed during this project are simple and use inexpensive apparatus to assay the range of physical properties spanned by the sample set.

The report provides data in support of (1) the design of the proposed Waste Handling and Packaging Plant, and (2) research and development activities in developing waste management alternatives.

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1. INTRODUCTION

The purpose of this study was to determine physical properties of the radioactive sludges in the waste storage tanks at the Oak Ridge National Laboratory (ORNL). Objectives include providing data in support of (1) the design of the proposed Waste Handling and Packaging Plant (WHPP) and (2) research and development (R&D) activities in developing waste management alternatives. This information is needed to design systems for processing these wastes for disposal. Relevant sludge properties include density, solids content, viscosity, sedimentation rate, and particle size. These characteristics will influence the selection of the technology for removing the sludges from the tanks, as well as the design of the piping, valves, pumps, and drivers for plant process systems.

This report describes the physical characterization of four samples that are typical of sludges in the Melton Valley Storage Tanks (MVSTs) and the storage tanks at the low-level waste (LLW) evaporator service facility in Bethel Valley. Methods used for sampling the tanks and results of the analyses of the wastes for chemical and radiochemical constituents are presented in a related report by Sears et al.[1]

Other research groups at ORNL have performed extensive characterization of the waste's chemical and radiochemical properties;[1,2] however, until this project, no detailed assay of physical properties had been done. These measurements were difficult to make for several reasons, including sample inhomogeneity, mutability of physical characteristics after sampling, the need to make several measurements from the same small samples, and the radiotoxicity of the samples. Several simple methods have been developed to make the necessary physical measurements. The project has provided important information to the plant's designers, and the methods reported here may guide others in making similar measurements. The report also summarizes limitations and sources of error associated with sampling and measurement.

2. SAMPLE HANDLING AND ANALYSIS PLAN

2.1 SAMPLE COLLECTION AND HANDLING

During this project, 21 sludge samples and 22 liquid samples were collected from six of the MVSTs (tanks W-24 through W-28 and W-31) and two of the storage tanks at the evaporator service facility (W-21 and W-23). The collection of the samples and the chemical and radiochemical characterization of the wastes are described in detail in ref. 1. Four sludge samples were selected as a representative set for the physical measurements described in this report based on apparent physical properties and estimated volumes.

A brief summary of the collection and handling of the samples is given in this section (for details, see ref. 1). All of the tanks are located in below-grade concrete vaults, and each is accessed by means of a 7.6-cm (3-in.)-diam pipe that penetrates the tank from the roof of the vault. Liquid samples were pulled by vacuum through Teflon tubing into the sample jars. A bottom-opening soft-sludge sampler was used to collect a core of sludge up to 51 cm (20 in.) deep. The device consists of a detachable handle assembly and a hollow probe of clear polyvinyl chloride (PVC) pipe (2.5 cm ID) that can be controlled from above by the operator. The sludge layer was usually more than 51 cm deep. Samples were collected at successively lower layers to obtain a vertical profile of the tank contents. After the sludge sample had been collected, the handle was removed and the PVC sample tube was capped, packaged, and transported to the analytical laboratory in a shielded carrier.

Sludge samples were unloaded from the carrier and placed in a hot cell. The samples were allowed to stand overnight in the PVC tubes to allow the solids to settle. The height of the sludge (solids) layer in the tank was then measured. Any liquid layer over the sludge was removed and the sludge (solids) transferred to a sample jar. The sample was stirred gently, and portions were removed for waste characterization studies. Composite sludge samples representative of a complete vertical core were made up for the chemical and radiochemical analyses.[1] The composite samples were sonicated to ensure complete mixing. The physical measurements described in this report were conducted on unsonicated samples that had been handled as little as possible. Information concerning the sludge samples used in this study is given in Table 1. (Additional information is available in ref. 1.) Dose rates (field survey) for the samples in the PVC sampling tubes ranged from 1.2 to 2.5 R/h. Radiation fields were considerably higher near small samples dried in the laboratory; dose

Table 1. Information for sludge samples

Tank	Sample	Date sampled	Radiation from sample (R/h)[a]	Sludge height in sampler (cm)	Region of sludge phase sampled	Depth of settled solids in tank (cm)
W-21	W21-S1	1/31/90	1.5	33.0	Top of sludge layer	66
W-23	W23-S1	1/31/90	2.5	48.2	Top of sludge layer[b]	132
W-26	W26-S3	12/5/89	1.2	47.0	Bottom of sludge layer	124
W-28	W28-S1	9/21/89	1.2	48.9	Complete vertical core	49

[a]Field survey of the loaded PVC samplers.

[b]A waste transfer was made in August 1989 with the more fluid sludge pumped from tank W-23 to the MVST. The residual sludge in tank W-23 was too thick to pump with the existing equipment.

rates of up to 50 R per hour per gram were observed. The increased dose was primarily due to beta particles from ^{90}Sr, which had been attenuated in the wet sludges. Sludge samples were handled in a hot cell; small subsamples were removed for analysis in radiochemical hoods and glove boxes. Every effort was made to avoid handling dried samples in order to reduce both exposure and spread of contamination.

Because only sludge directly under the access point can be sampled, the samples may not be representative of other locations in the tank and should be considered as merely an indicator of the tank contents.

2.2 ANALYSIS PLAN

Samples described in this report were used for chemical, radiochemical, and physical measurements, so strict sample conservation was imperative. An analysis plan was developed which would yield all the required physical data for a tank using 20 g of sludge and 60 mL of supernatant liquid. A flow chart illustrating the physical analysis plan is shown in Fig. 1. The residue from the total solids determination could be dissolved in acid and used in radiochemical assays to conserve sample; however, only the composite sludge samples (i.e., one sample per tank) were assayed (ref. 1) because of budget limitations.

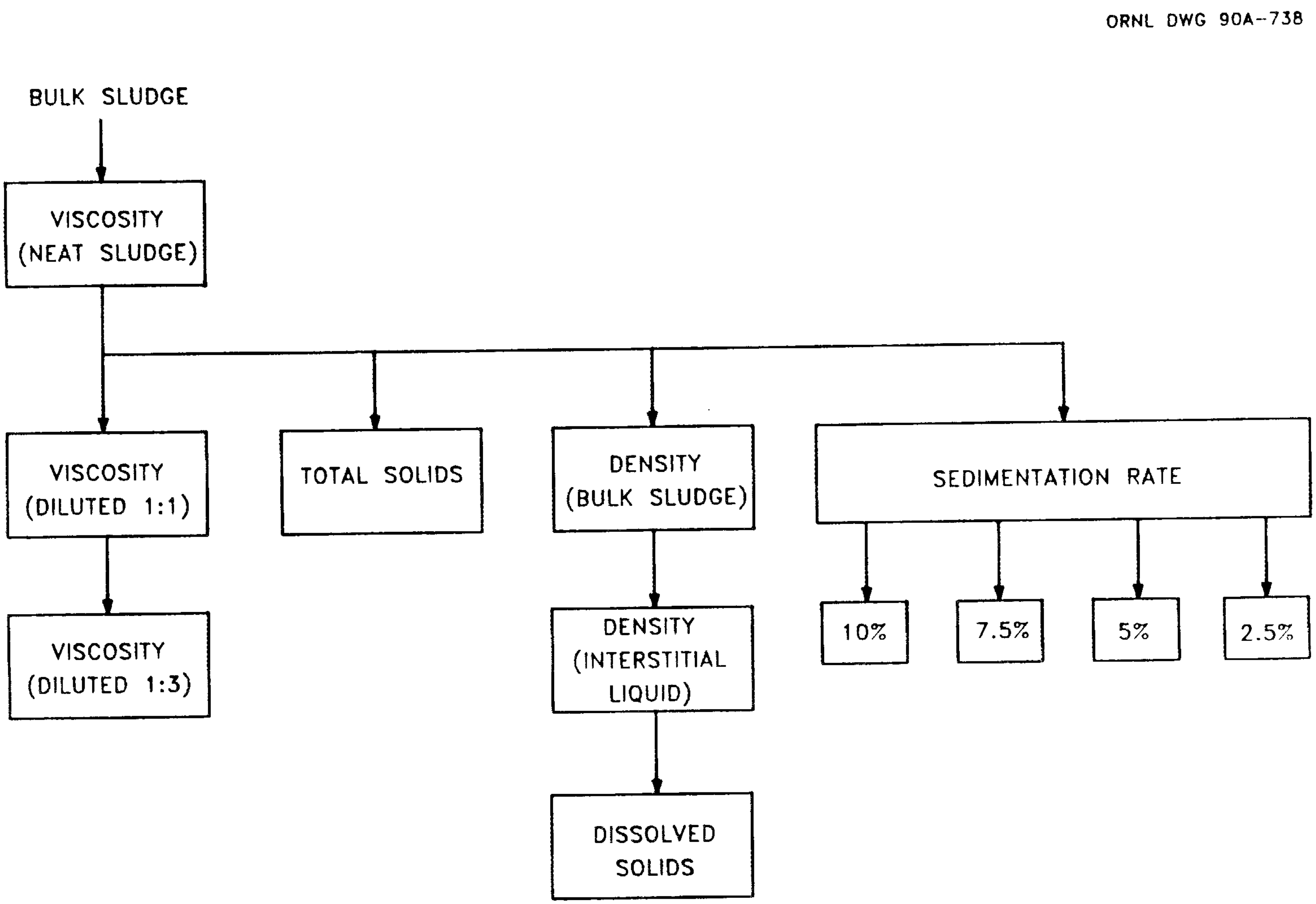

Fig. 1. Flow chart for physical measurements.

3. VISCOSITY

3.1 METHOD

Samples of the waste sludges and supernatant (bulk) liquids were assayed for viscosity using a digital Brookfield rotational viscometer[3] according to ASTM Standard D2196-86, Method B.[4] Sludges were tested using the small sample adapter and cylindrical spindles Nos. 18 and 34; liquids were tested using the low-viscosity adapter and cylindrical spindle No. 00.

Viscosities of liquid samples were measured as received; the sludges were measured as received and/or after dilution with bulk liquid from the same tank. All measurements were made at room temperature (21 ± 2°C). Viscosity and shear stress were tracked for each of several shear rates.

3.2 RESULTS

Results of the viscosity measurements are summarized in Table 2. Plots of shear stress vs shear rate are shown in Figs. 2—9 for several sludges. The "plastic viscosity" of a sludge, which is the change of shear stress with increasing shear rate, is the slope of the straight-line portion of the plot. The "yield stress" of a sludge, which is the applied stress necessary to initiate flow, is the y-intercept of the plot. Both quantities are shown graphically on the plots. The viscosity of a sludge can be reduced considerably if it is mixed with an equal volume of supernatant liquid from the same tank. The stated accuracy of the Brookfield rotational viscometer is ±1% of the instrument's full-scale reading. Measurements taken with less than 10% instrument response (i.e., >±10% error) are marked as solid black squares on the plots; other data points are >±1 and <±10%. A detailed listing of the viscosity data is presented in Appendix A, Tables A.1—A.4.

Viscosity is a dynamic property which varies with existing conditions. The bulk liquids in these waste tanks were fairly Newtonian (viscosity independent of shear rate), but the sludges exhibited non-Newtonian flow behavior, including pseudoplasticity (viscosity decreases as shear rate increases, see Appendix A, Tables A.1—A.4). Some time-dependent variation in shear stress at constant shear rate was observed, but insufficient data are available to permit positive statements about thixotropic behavior. One sludge (W26-S3) could not be clearly characterized as received. As shown in Fig. 5, the scatter in the shear-stress-vs-shear-rate data is too great to permit determination of the plastic viscosity and yield stress. On the

Table 2. Viscosities of selected waste tank samples

Property	W21-S1	W23-S1	W26-S3	W28-S1
Bulk liquid (cP)[a]	1.82	2.12	1.67	2.22
Neat sludge				
Plastic viscosity (cP)	56	b	c	7700[d]
Yield stress (dyn/cm^2)	57	-	-	22
Sludge diluted 1:1[e]				
Plastic viscosity (cP)	5.5[d]	95	70	130
Yield stress (dyn/cm^2)	2.2	44	105	66
Sludge diluted 1:3				
Plastic viscosity (cP)	-	-	-	55
Yield stress (dyn/cm^2)	-	-	-	20

[a]Bulk liquid samples W21-L3, W23-L1, W26-L2, and W28-L3 were taken from the same tanks as the sludge samples.

[b]Radiation field from undiluted sludge was too intense to permit viscosity measurements using sludge as received.

[c]There is too much scatter in shear-stress-vs-shear-rate data to determine the plastic viscosity or yield stress (see Fig. 5).

[d]Coagulated during test; not a true viscosity.

[e]Sludge diluted 1:1 by volume with bulk liquid taken from the same tank as the sludge sample.

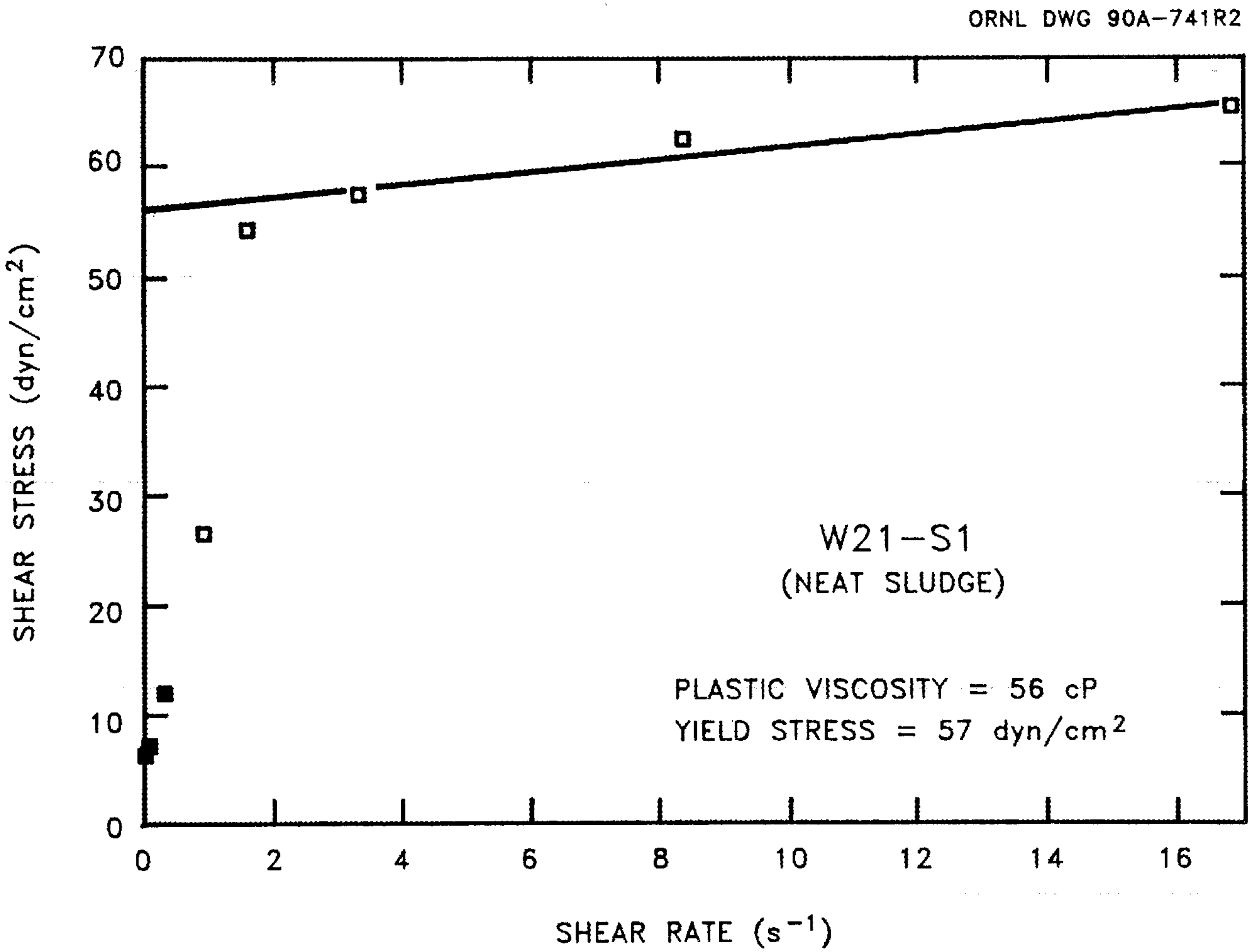

Fig. 2. Rheological properties of neat sludge sample W21-S1.

ORNL DWG 90A-740R2

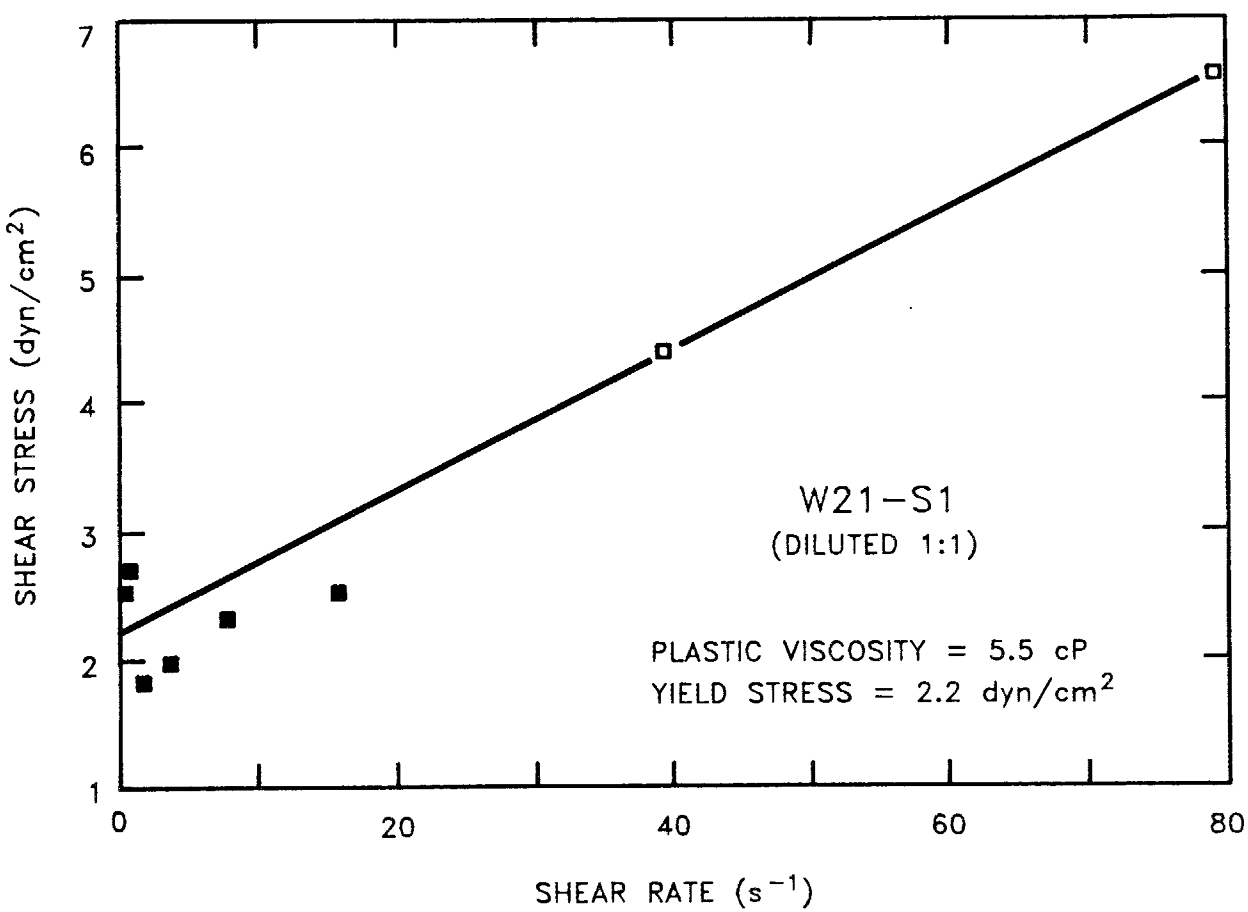

Fig. 3. Rheological properties of sludge sample W21-S1 diluted 1:1.

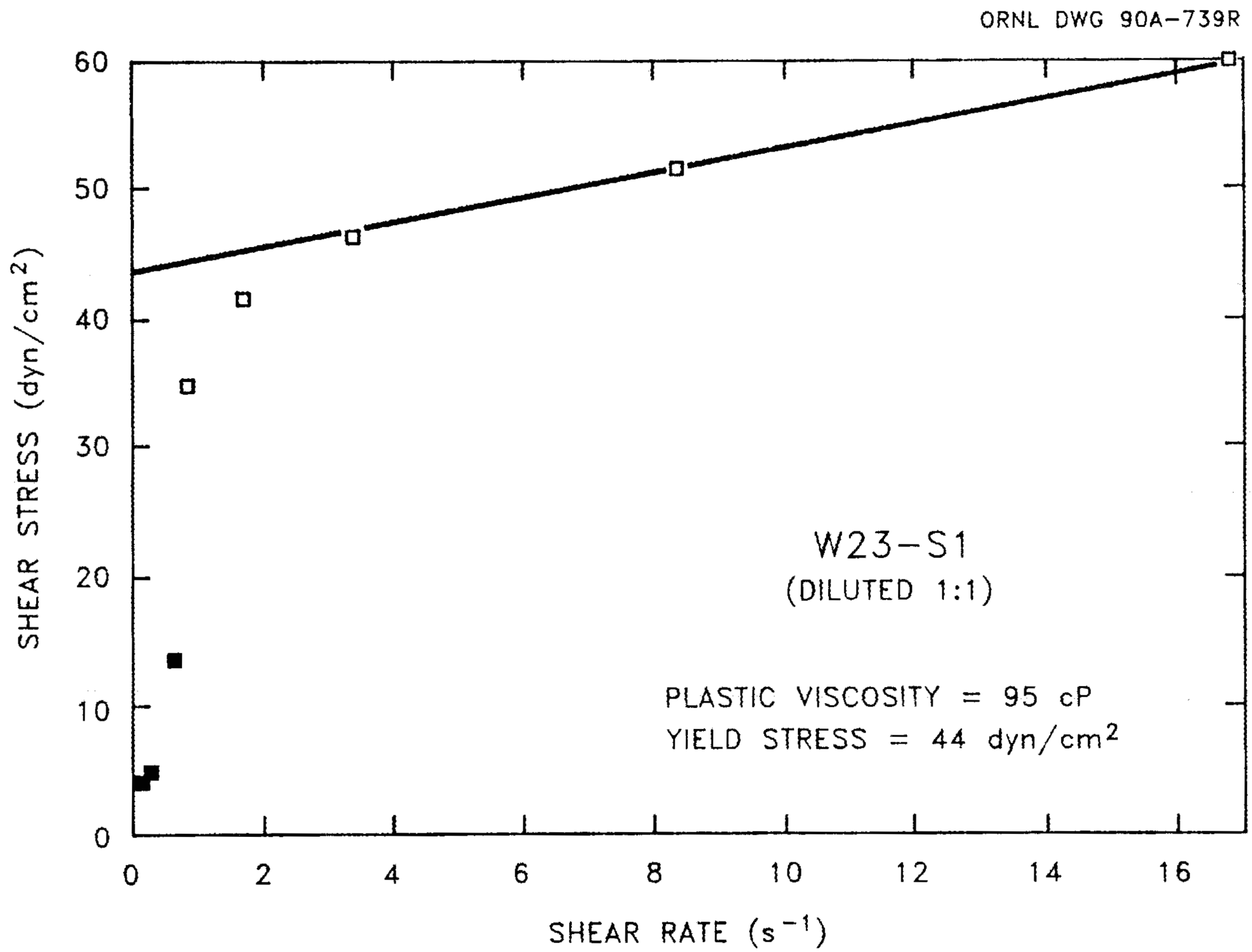

Fig. 4. Rheological properties of sludge sample W23-S1 diluted 1:1.

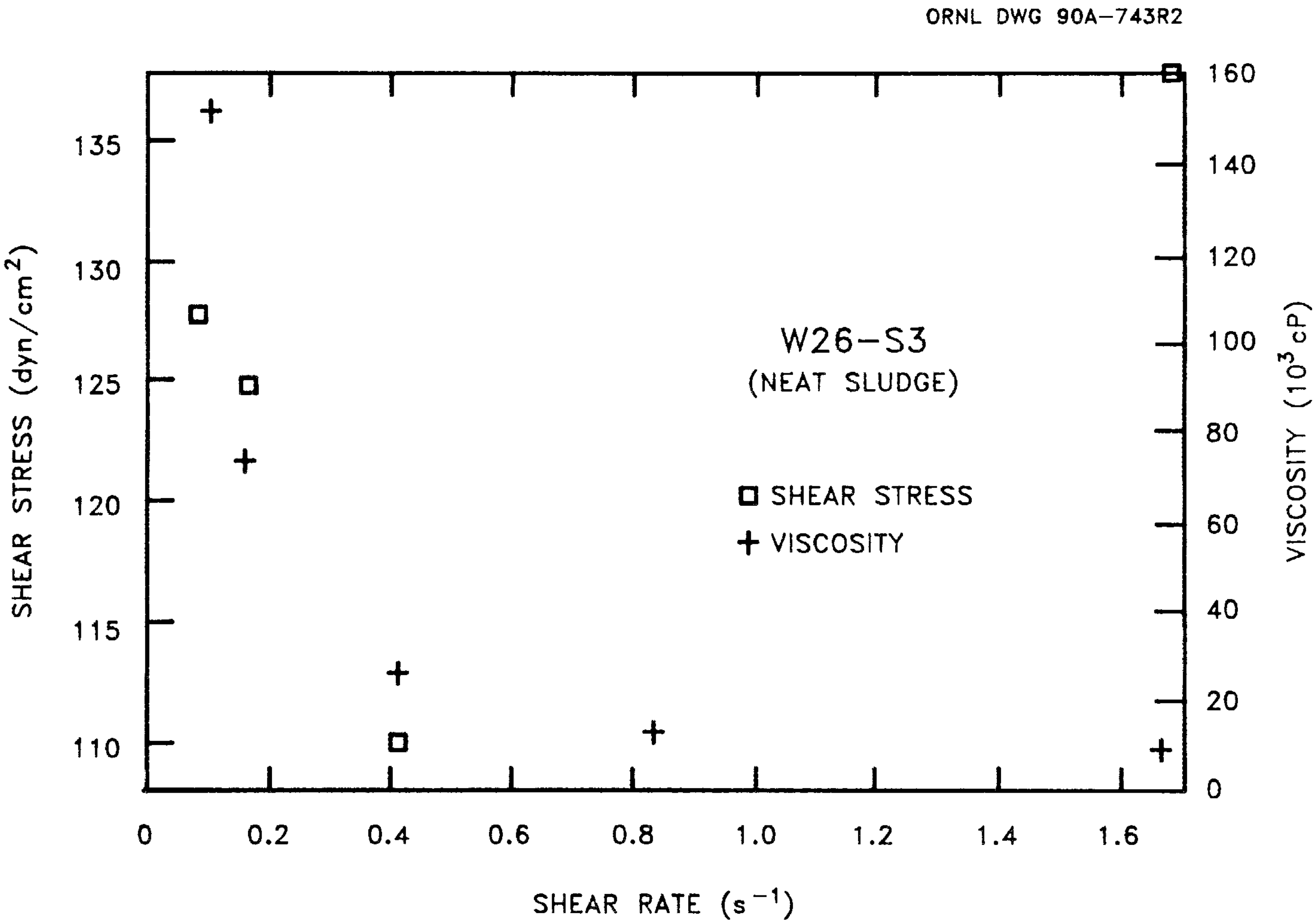

Fig. 5. Rheological properties of neat sludge sample W26-S3.

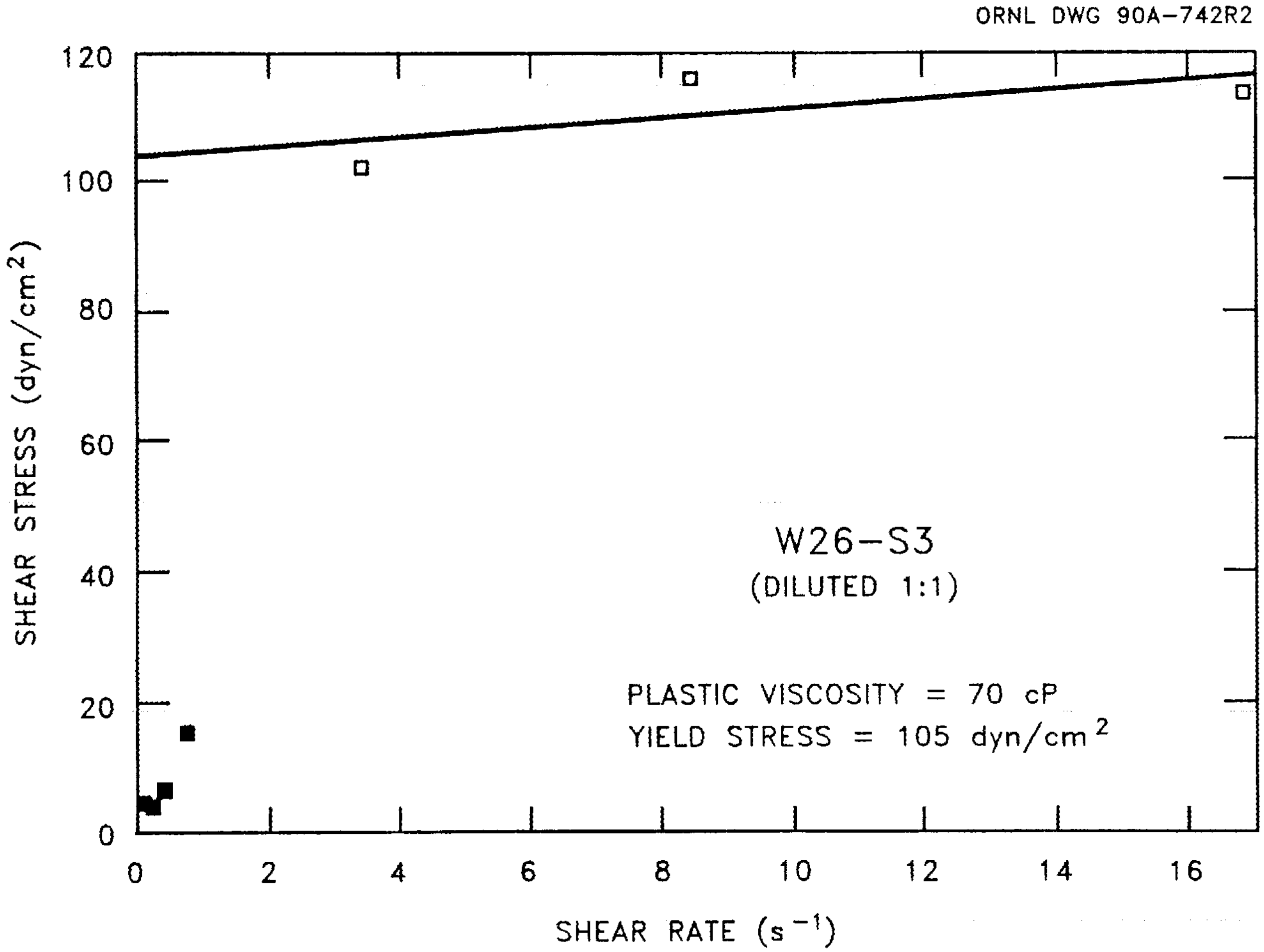

Fig. 6. Rheological properties of sludge sample W26-S3 diluted 1:1.

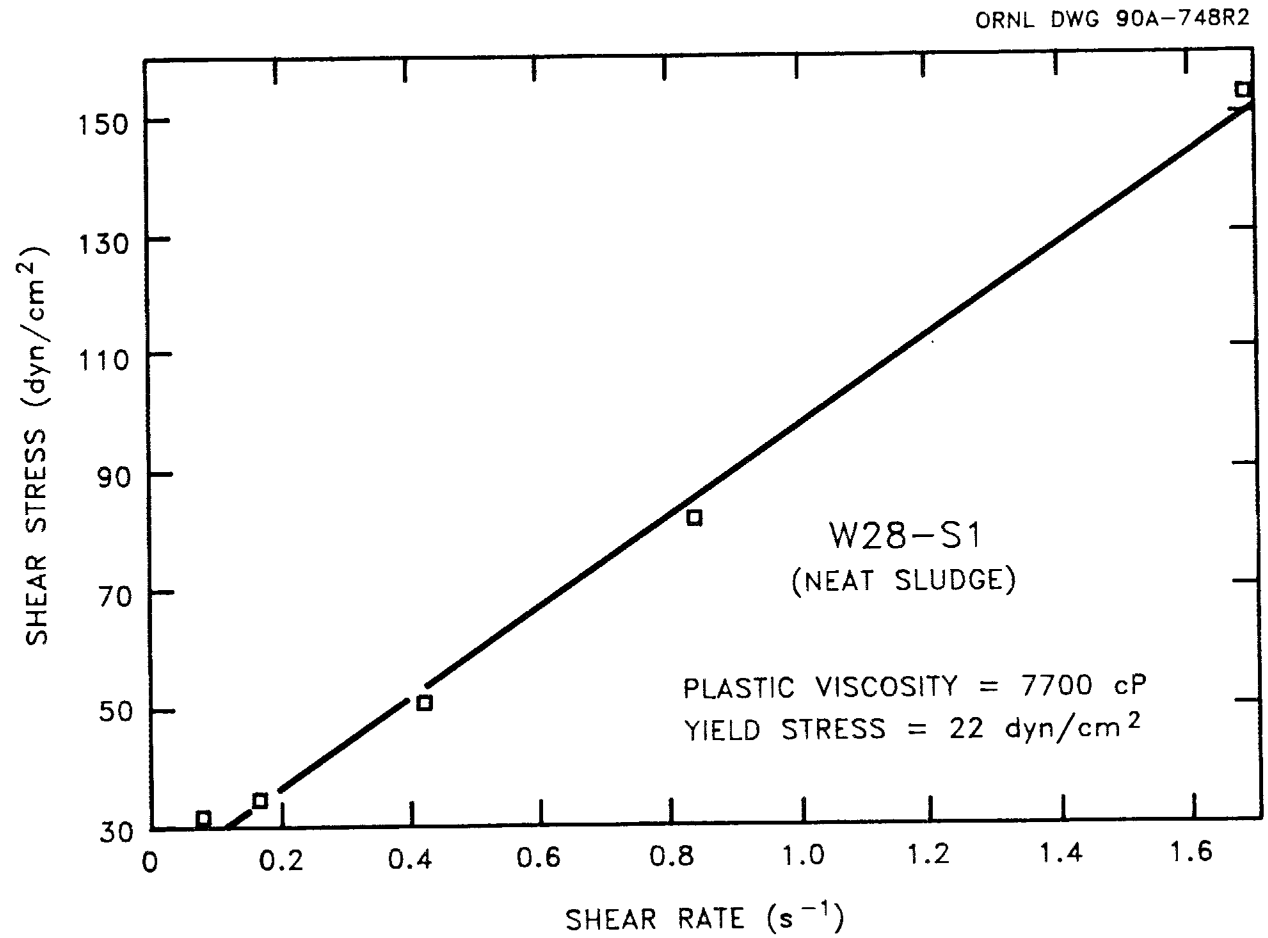

Fig. 7. Rheological properties of neat sludge sample W28-S1.

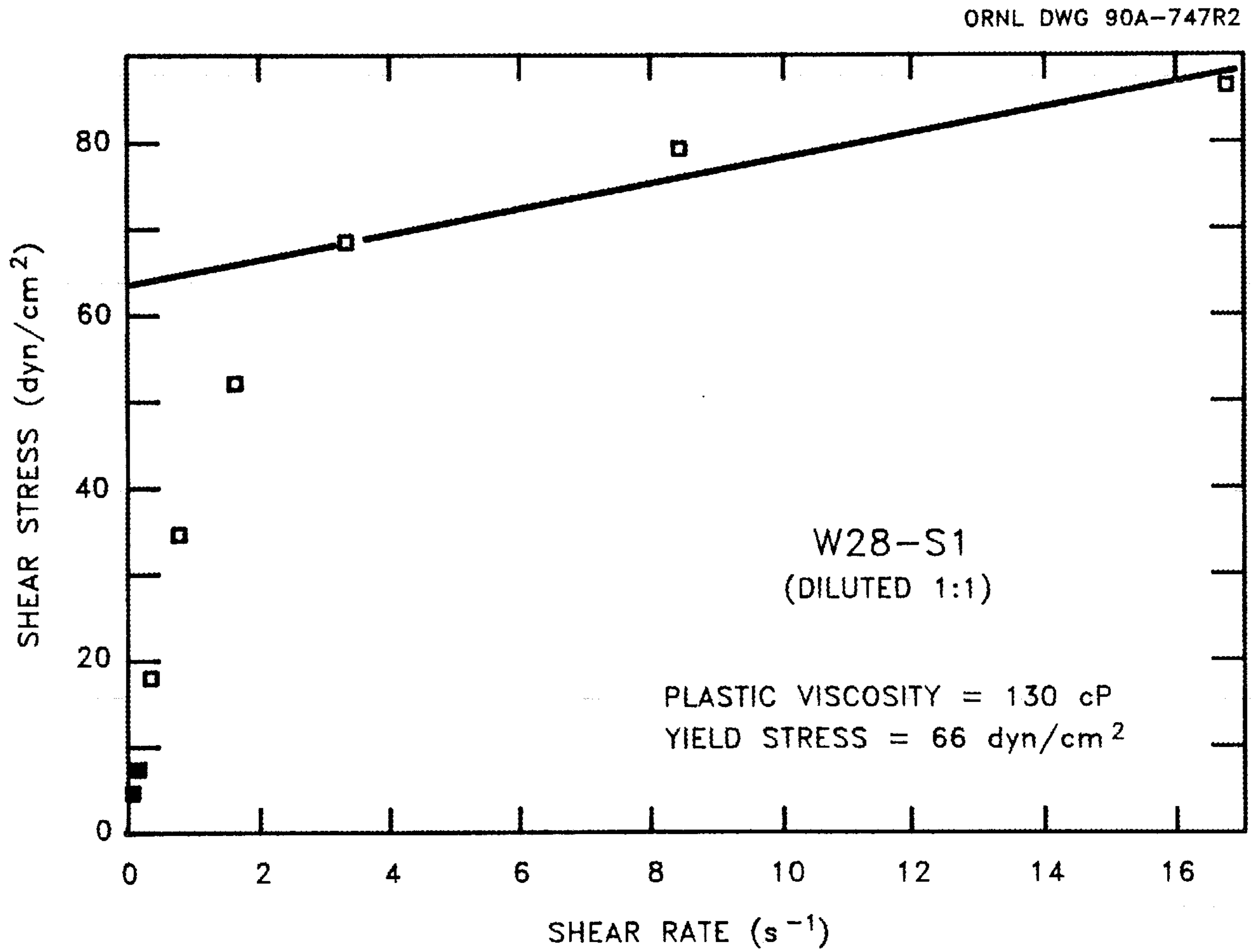

Fig. 8. Rheological properties of sludge sample W28-S1 diluted 1:1.

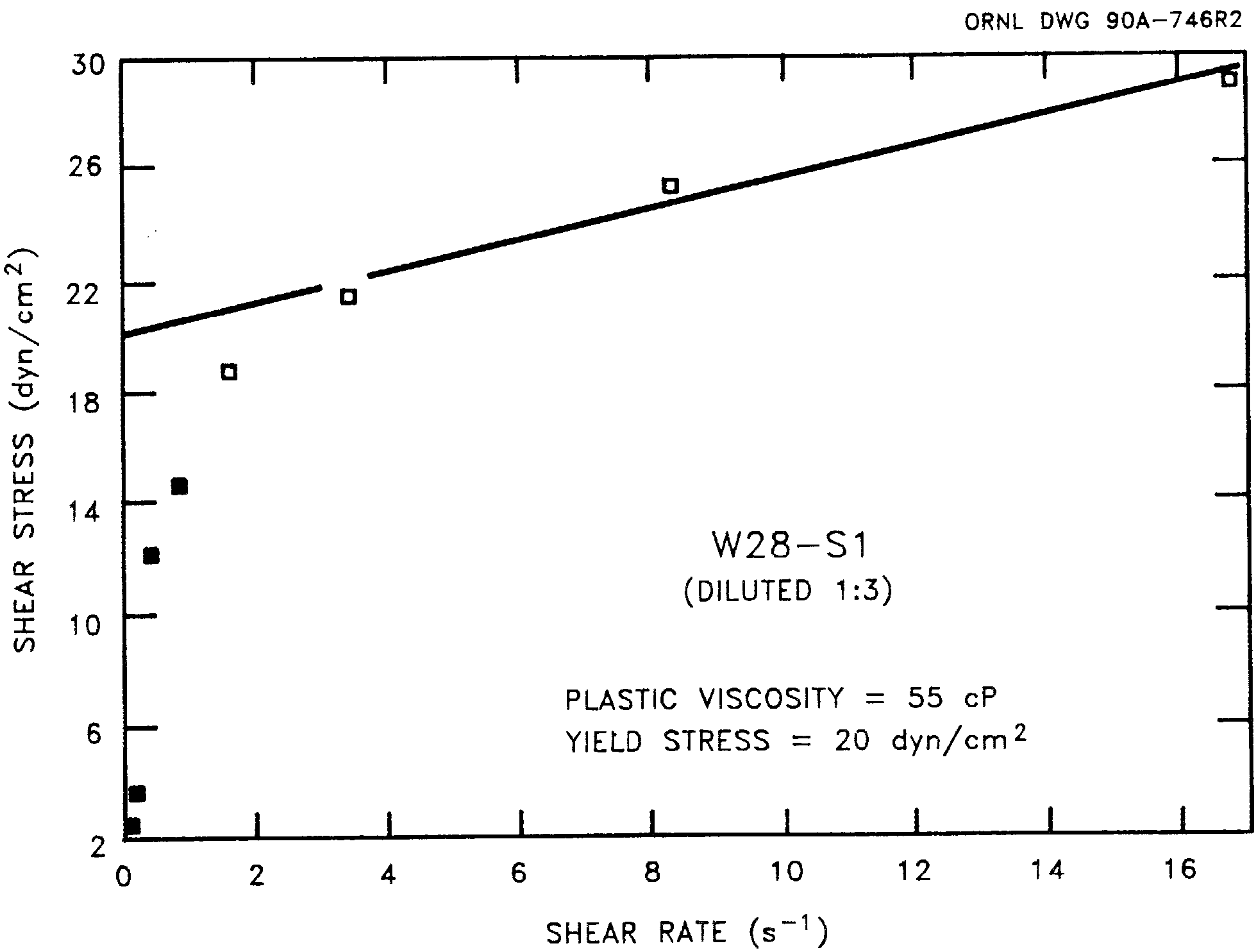

Fig. 9. Rheological properties of sludge sample W28-S1 diluted 1:3.

same plot, data are superimposed showing experimentally determined apparent viscosity over the same range of shear rates; these data show pseudoplasticity. The same sludge, diluted 1:1 with supernatant liquid, showed a more typical curve (Fig. 6).

Both the diluted sample W21-S1 and the neat sample W28-S1 coagulated appreciably during examination. The result was that a liquid layer formed between the sample and the cup, while the sludge rotated with the spindle almost without internal slippage. Therefore, data from these measurements cannot be considered true viscosities. Large granules in a sludge would frequently jam between the viscometer sample cup and spindle No. 18. Whenever possible, the smaller spindle No. 34 was used; however, the radiation field from sample W23-S1 was too intense to use such a large (12-mL) sample. These problems are amplified using small samples. More meaningful data would be obtained from troublesome samples using larger (500-mL) samples and special apparatus such as a T-bar spindle with a helical path. However, the high radiation level limits the sample quantity that can be collected in the field or handled in a radiochemical hood or glove box.

4. DENSITY AND SOLIDS MEASUREMENTS

4.1 METHOD

In principle, density and solids content are simple measurements to make; in this study, however, accurate values, particularly the percentages of dissolved and undissolved solids were difficult to obtain. The liquids in these tanks have high salt contents (about 4 M in nitrate), and most have high pH levels. If a sludge were simply vacuum filtered, the filter cake would retain some of the interstitial liquid. After being dried, the solids would contain an indeterminate mass of salts that had been dissolved in the original sample. If the filter cake were rinsed with water to remove the salts, some of the material that was insoluble in the original matrix might dissolve. The following method provides an indirect means of measuring the density and the solids content of the whole sludge, as well as of its dissolved and undissolved components.

Seven measurements were performed on each sludge sample to determine the density and the solids content of the whole sludge, its component interstitial liquid, and the component undissolved solids. Two portions of a sludge sample were required for these measurements. During these measurements, each sludge was inspected and visual and tactile impressions (i.e., color, texture) were recorded (see ref. 1).

The first portion of sludge, about 5 g, was packed into a short length of Teflon tubing. After the outside of the packed tubing had been wiped clean, the sample was lowered into a tared, graduated 15-mL glass centrifuge tube. The sludge was carefully extruded into the centrifuge tube using a glass rod as a piston so that no sludge was smeared on the inner wall of the centrifuge tube above the bulk of the sample. The centrifuge tube was then sealed with its screw cap and spun at a high rate (>4000 G) for 15 min to compact the solids and to displace any entrained air from the sludge. After centrifugation, a layer of clear liquid was visible above the compacted solids; the total volume of the sample was taken as the sum of the liquid and compacted solids volumes. The centrifuge tube was reweighed to determine the bulk sludge mass. The density of the bulk sludge was calculated by dividing the mass of the bulk sludge by the sample volume.

The separated interstitial liquid (see above) was withdrawn from the centrifuge tube and filtered through a 0.45-μm syringe filter. Then, 1.00 mL of the filtered liquid was pipetted into a tared 10-mL glass beaker. This beaker was weighed to determine the liquid mass and subsequently dried for 16 ± 1 h at 110 ± 5°C before reweighing to determine the mass of the residual dried salts. These measurements give the density (g/mL) of the interstitial liquid, the quantity of dissolved solids per milliliter of liquid, and the water loss per milliliter of liquid upon drying.

The second portion of sludge was used to determine the percentage of total solids in the bulk sludge. About 1 g of the sludge was placed into a tared 10-mL beaker. The beaker containing the sample was reweighed to determine the wet sludge mass and then dried for 16 ± 1 h at 110 ± 5°C. The beaker containing the dried solids was weighed.

4.2 RESULTS

Results of the density and solids measurements are summarized in Table 3. Details of the calculations are given in Appendix B. The density of the bulk sludge ranged from 1.34 to 1.44 g/mL, while that of the undissolved solids ranged from 1.68 to 2.44 g/mL. The densities of both the interstitial liquid and the bulk tank liquid (which was used in the sedimentation experiments described in Sect. 5) are shown in Table 3. The dissolved solids comprise 24 to 29 wt % of the bulk sludge and the undissolved solids 22 to 25 wt %.

The sludges were added to the tanks in layers at various times. Wastes have been transferred from one tank to another within the system, and some liquid wastes have been removed from the system for solidification. The interstitial liquid associated with a sludge does not necessarily have the same composition as the bulk liquid in the same tank. For example, the sodium/potassium ratio in the sludge is sometimes different from that in the bulk liquid; and in tank W-21, the pH of the bulk liquid was 0.8 while the pH of the sludge liquid was 7.* All of the other tank liquids are basic. Volume readings were accurate to ±0.1 mL, and mass readings were taken to the nearest 0.0001 g. Uncertainties for density and solids measurements are estimated at ±5%. No attempt was made to account for waters of hydration or crystallization in these measurements.

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*Tank W-21 receives liquid waste from the process waste treatment plant. Historically, it has served as a feed tank and as a concentrate storage tank for the low-level waste (LLW) evaporator and may contain these sludges. The other tanks, which are all basic, contain predominantly LLW concentrates.

Table 3. Density and solids measurements

Property	W21-S1	W23-S1	W26-S3	W28-S1
Density				
Bulk liquid (g/mL)[a]	1.2391	1.2423	1.2177	1.2852
Bulk sludge (g/mL)	1.34	1.44	1.36	1.40
Interstitial liquid (g/mL)	1.26	1.27	1.23	1.29
Undissolved solids (g/mL)	1.68	2.44	2.16	2.00
Sludge solids				
Total solids (wt %)	51.9	52.4	46.0	51.4
Dissolved solids (wt %)	28.2	27.5	23.6	29.4
Undissolved solids (wt %)	23.7	24.9	22.4	22.0

[a]Bulk liquid samples W21-L3, W23-L1, W26-L2, and W28-L3 were taken from the same tanks as the sludge samples.

5. SEDIMENTATION RATE

5.1 METHOD

The sedimentation rate of each sludge was determined by placing small portions (0.2 to 2 mL of the bulk sludge) into four 10-mL graduated mixing cylinders containing bulk liquid from the same tank so that the combined volume was 10.0 mL in each cylinder. The mixing cylinders were stoppered and their contents mixed gently by repeated inversion for 1 min. After mixing, the cylinders were righted and tapped sharply several times to release entrained air. As the solids settled, the positions of the liquid-slurry interface were recorded at 1-min intervals for a total of 30 min. When sedimentation tests were complete, the slurries were remixed, decanted into graduated centrifuge tubes, and centrifuged at a high rate (>4000 G) for 15 min, and the volume of compacted solids was measured. This provided a uniform basis for comparing undissolved solids contents.

5.2 RESULTS

Factors that influence the settling rates of individual particles include solids concentration, liquid density and viscosity, particle density, and particle radius. When a slurry is allowed to settle, the particles are initially separate and have zero average downward velocity. As the particles agglomerate and are accelerated by gravity, they settle faster until the force of gravity is balanced by the viscous drag exerted by the liquid they are falling through. The particles continue at constant velocity until they begin to pile up on the container bottom. A typical waste tank sludge sedimentation rate curve is given in Fig. 10.

Sedimentation data were recorded for several slurries of each sample, ranging from very dilute to partially compacted. (If a slurry was too dilute, coagulation was inefficient and the lighter particles settled poorly. If the slurry was too concentrated, the sludge was already partially compacted and the particles could not fall freely through the liquid.) Results were plotted for each dilution as settling rate vs elapsed time. Terminal velocities were plotted as their logarithms vs volume of compacted solids. The latter plots were extrapolated to infinite dilution to determine true sedimentation rates. Figure 11 shows a plot of the concentration dependence of the sedimentation rate for a typical waste tank sludge.

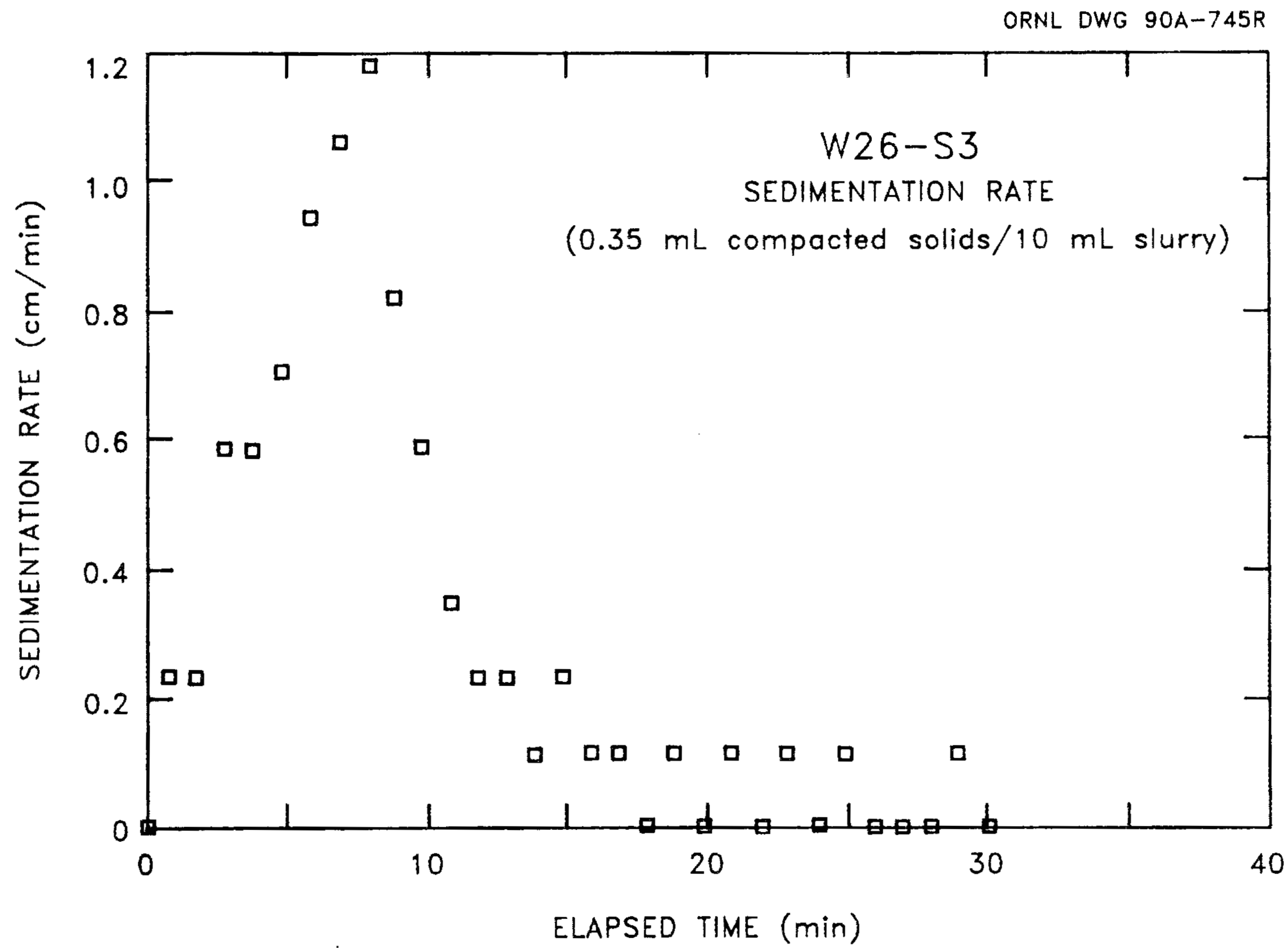

Fig. 10. Typical sedimentation rate curve.

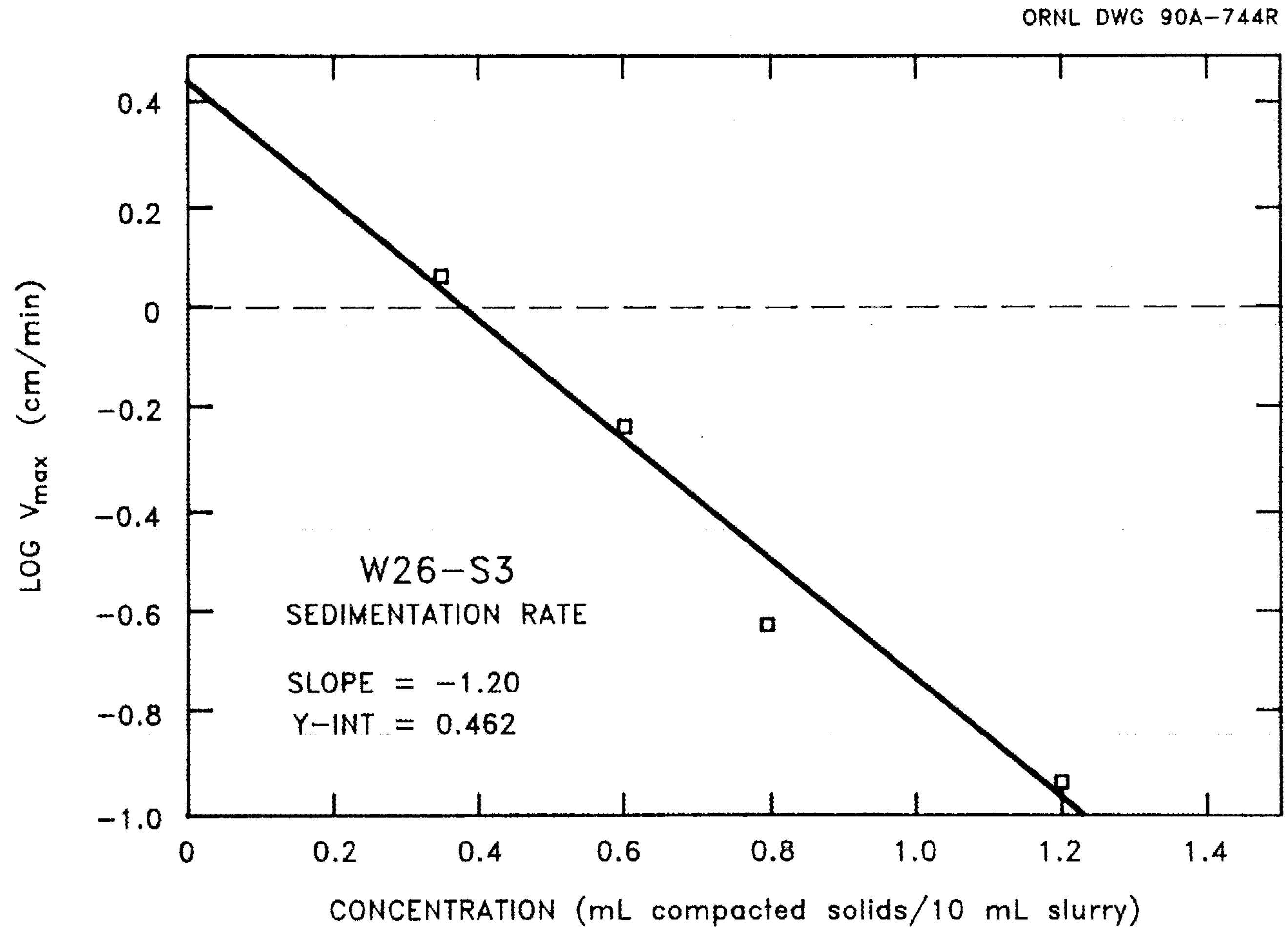

Fig. 11. Typical concentration dependence of sedimentation rate.

The sedimentation rates (at terminal velocity) for the sludge samples from three of the tanks are about 3 to 4 cm/min (Table 4). The rate for the sample W21-S1 is lower, about 1 cm/min.

Typical waste tank sludges have both granular and flocculent components; the flocculent component appears to make up about 90% of the sludge mass. The granules settle faster, as a rule, but are obscured from view by the floc. The sedimentation rates reported here are rates for the flocculent particles only. Data from these measurements are listed in Appendix A, Exhibits A.1—A.4.

Table 4. Floc sedimentation rate[a]

Sample	Terminal velocity (cm/min)
W21-S1	1.0
W23-S1	3.9
W26-S3	2.9
W28-S1	2.9

[a]Sedimentation rate in bulk liquid samples W21-L3, W23-L1, W26-L2, and W28-L3, which were taken from the same tanks as the sludge samples.

6. PARTICLE SIZE

During the sedimentation tests described in Sect. 5, sludge particles dispersed through the liquid during the initial agitation, then were seen to agglomerate when mechanical shearing stopped. An estimate of agglomerate size may be made by applying Stokes' law,[5] which describes spherical particles falling through a viscous fluid under the influence of gravity. It relates the terminal velocity of such a particle to the particle size, particle and liquid densities, and the liquid viscosity, as follows:

$$u = \frac{2gr^2(\rho - \rho')}{9\eta} \quad ,$$

where u = terminal velocity at zero concentration,
g = acceleration due to gravity,
r = radius of the falling particle,
ρ = particle density,
ρ' = liquid density,
η = liquid viscosity.

Agglomerate size was computed for each of the sludges using experimental values for liquid density and viscosity, particle density, and terminal velocity (Table 5). Application of Stokes' law yields only approximate particle sizes because of several factors: (1) sludge particle agglomerates are not spherical, (2) falling particles may be porous and carry some of the bulk liquid with them as they fall, and (3) terminal velocity was only measured for the slowest-settling fraction.

Table 5. Particle size estimates

Sample	Agglomerate radius (μm)
W21-S1	18
W23-S1	23
W26-S3	20
W28-S1	26

7. DISCUSSION

The design parameters for the WHPP will be influenced by the physical characteristics of the feedstock. Pump configuration and size depend on the rheological properties of the sludges to be pumped. An estimate of the rheology of the diluted sludge will be useful, since it may be necessary to dilute the sludge to mobilize it from the tank or to reduce the horsepower requirements for pump drivers. Knowledge of sedimentation behavior will help in choosing mass transport conditions. The viscosity of a sludge can be reduced considerably if it is mixed with an equal volume of supernatant liquid from the same tank. At this dilution, the solids are still partially compacted; such a mixture would be fairly stable during pumping and transport.

Table 6 summarizes some physical properties of selected waste tank sludges. Values for viscosity, sedimentation rate, and particle size listed here assume that the interstitial liquid associated with a sludge and the bulk liquid from the same tank have the same chemical composition; that is, no chemical or physical change occurred when the undissolved solids in a sludge were suspended in the bulk liquid from the same tank. This assumption is not always warranted; for example, in one tank, the pH of the bulk liquid was 0.8 and the pH of the sludge liquid was 7. Such chemical dissimilarity could have a profound impact on the physical properties of a sludge, particularly when the contents of a tank are mixed.

For the WHPP design, it has been proposed that solids be transported from the waste tank to the plant by using water as a transport fluid. (If some suspended solids dissolved in the transport medium, it might affect the physical properties.) Advantages of this strategy could include reduced exposure from beta and gamma radiation during sludge handling and separation of shorter-lived soluble radionuclides, such as ^{137}Cs and ^{90}Sr, from transuranic waste. A new set of experiments is being planned to assess the solubilities of sludge components in water under process conditions.

Table 6. Physical properties of selected waste tank samples

Physical property	W21-S1	W23-S1	W26-S3	W28-S1
Density				
Bulk liquid (g/mL)[a]	1.2391	1.2423	1.2177	1.2852
Bulk sludge (g/mL)	1.34	1.44	1.36	1.40
Interstitial liquid (g/mL)	1.26	1.27	1.23	1.29
Undissolved solids (g/mL)	1.68	2.44	2.16	2.00
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Total solids (wt %)	51.9	52.4	46.0	51.4
Dissolved solids (wt %)	28.2	27.5	23.6	29.4
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Viscosity				
Bulk liquid (cP)[a]	1.82	2.12	1.67	2.22
Neat sludge				
Plastic viscosity (cP)	56	b	c	7700[d]
Yield stress (dyn/cm^2)	57			22
Sludge diluted 1:1[e]				
Plastic viscosity (cP)	5.5[d]	95	70	130
Yield stress (dyn/cm^2)	2.2	44	105	66
Sludge diluted 1:3				
Plastic viscosity (cP)	-	-	-	55
Yield stress (dyn/cm^2)	-	-	-	20
Agglomerate radius (μm)	18	23	20	26
Floc Sedimentation Rate				
Terminal velocity (cm/min)	1.0	3.9	2.9	2.9

[a]Bulk liquid samples W21-L3, W23-L1, W26-L2, and W28-L3 were taken from the same tanks as the sludge samples.

[b]Radiation field from undiluted sludge was too intense to permit viscosity measurements using sludge as received.

[c]Rheological data are too scattered to determine plastic viscosity or yield stress (see Fig. 5).

[d]Coagulated during test; not a true viscosity.

[e]Sludge diluted 1:1 by volume with bulk liquid taken from the same tank as the sludge sample.

8. ACKNOWLEDGMENTS

The authors wish to express their appreciation to E. L. Youngblood and H. O. Weeren for their helpful comments in reviewing this report.

9. REFERENCES

1. M. B. Sears et al., Sampling and Analysis of Radioactive Liquid Wastes and Sludges in the Melton Valley and Evaporator Facility Storage Tanks at ORNL, ORNL/TM-11652, September 1990.

2. F. J. Peretz, B. R. Clark, C. B. Scott, and J. B. Berry, Characterization of Low-Level Liquid Wastes at the Oak Ridge National Laboratory, ORNL/TM-10218, December 1986.

3. Brookfield Engineering Laboratories, Inc., More Solutions to Sticky Problems, 1987.

4. ASTM Standard D2196-86, "Rheological Properties of Non-Newtonian Materials by Rotational (Brookfield) Viscometer," 1986.

5. S. Glasstone, Textbook of Physical Chemistry, D. Van Nostrand Co., New York, pp. 497-498, 1946.

APPENDIX A

LISTING OF PHYSICAL PROPERTIES DATA

APPENDIX A

LISTING OF PHYSICAL PROPERTIES DATA

The tables in this appendix list the raw numeric data obtained from the physical measurements described in this report. They also include observations made during the measurements.

Table A.1. Viscosities of bulk liquid and sludge, tank W-21[a]

Spindle (rpm)	Instrument response	Shear rate (1/s)	Viscosity (cP)	Shear stress (dyn/cm^2)
W21-L3 (liquid)				
-	-	72	1.8	1.3
W21-S1 (sludge), undiluted				
0.3	0.04	0.08	7600	6.4
0.6	0.05	0.16	4500	7.3
1.5	0.07	0.42	2800	12
3	0.13	0.84	2700	26
6	0.25	1.7	2700	55
12	0.35	3.4	1800	58
30	0.38	8.4	760	63
60	0.40	17	390	66
W21-S1 (sludge), diluted 1:1 using liquid W21-L3[b,c]				
0.3	0.07	0.40	720	2.5
0.6	0.08	0.80	320	2.7
1.5	0.07	2.0	110	1.8
3	0.06	4.0	51	2.0
6	0.07	7.9	30	2.3
12	0.08	16	16	2.5
30	0.11	40	12	4.4
60	0.16	79	8.3	6.6

[a]Measured by Brookfield rotational viscometer.

[b]The diluted sample appeared to coagulate so that solids adhered to and rotated with the spindle. Most slippage occurred within a thin liquid layer which formed between the solids mass and sample cup. Therefore, the affected measurements do not assay true viscosity.

[c]Measured using spindle No. 18.

Table A.2. Viscosities of bulk liquid and sludge, tank W-23[a,b]

Spindle (rpm)	Instrument response	Shear rate (1/s)	Viscosity (cP)	Shear stress (dyn/cm^2)
W23-L1 (liquid)				
-	-	76	2.1	1.6
W23-S1 (sludge), diluted 1:1 using liquid W23-L1				
0.3	0.02	0.08	4600	3.9
0.6	0.03	0.17	2700	4.5
1.5	0.08	0.42	3200	13
3	0.21	0.84	4200	35
6	0.25	1.7	2500	42
12	0.27	3.4	1400	47
30	0.31	8.4	630	53
60	0.36	16.8	360	60

[a]Undiluted sludge was too viscous to measure using the large spindle (No. 18). The radiation field near the undiluted sludge was too intense (700 mR/20 g) to allow the use of the smaller spindle (No. 34) and a larger sample volume.

[b]Measured by Brookfield rotational viscometer.

Table A.3. Viscosities of bulk liquid and sludge, tank W-26[a]

Spindle (rpm)	Instrument response	Shear rate (1/s)	Viscosity (cP)	Shear stress (dyn/cm^2)
W26-L2 (liquid)				
-	-	73	1.7	1.2
W26-S3 (sludge), undiluted				
0.3	0.75	0.08	156,000	128
0.6	0.73	0.17	74,000	125
1.5	0.63	0.42	27,000	110
3	0.62	0.84	13,000	108
6	0.80	1.7	8,200	138
12	Off scale			
W26-S3 (sludge), diluted 1:1 using liquid W26-L2				
0.3	0.03	0.08	5,900	5.1
0.6	0.03	0.17	2,800	4.8
1.5	0.04	0.42	1,500	6.4
3	0.09	0.84	1,900	16
6	0.40	1.7	4,000	67
12	0.61	3.4	3,100	103
30	0.68	8.4	1,400	117
60	0.67	16.8	680	113

[a]Measured by Brookfield rotational viscometer.

Table A.4. Viscosities of bulk liquid and sludge, tank W-28[a]

Spindle (rpm)	Instrument response	Shear rate (1/s)	Viscosity (cP)	Shear stress (dyn/cm^2)
W28-L3 (liquid)				
-	-	36	2.2	0.81
W28-S3 (sludge), undiluted[b]				
0.3	0.19	0.08	39,000	32
0.6	0.21	0.17	21,000	35
1.5	0.30	0.42	12,000	51
3	0.48	0.84	9,800	82
6	0.90	1.7	9,200	150
12	Off scale			
W28-S3 (sludge), diluted 1:1 using liquid W28-L3				
0.3	0.03	0.08	6,000	5.0
0.6	0.04	0.17	4,600	7.4
1.5	0.10	0.42	4,400	18
3	0.21	0.84	3,600	35
6	0.31	1.7	3,100	52
12	0.41	3.4	2,000	69
30	0.47	8.4	950	79
60	0.51	16.8	520	87
W28-S3 (sludge), diluted 1:3 using liquid W28-L3				
0.3	0.02	0.08	2,800	2.2
0.6	0.02	0.17	2,000	3.5
1.5	0.07	0.42	2,900	12
3	0.08	0.84	1,700	15
6	0.10	1.7	1,100	19
12	0.12	3.4	640	22
30	0.15	8.4	300	26
60	0.17	16.8	170	29

[a]Measured by Brookfield rotational viscometer.

[b]The undiluted sample appeared to coagulate so that solids adhered to and rotated with the spindle. Most slippage occurred within a thin liquid layer which formed between the solids mass and sample cup. Therefore, the affected measurements do not assay true viscosity.

Exhibit A.1. Sedimentation test data for sludge from tank W-21[a,b]

Dilution 1: 1.7 mL compacted solids/mL of slurry

Liquid/slurry interface height (cm)

Time (min)	0	1	2	3	4	5	6	7	8	9
0	7.6	7.5	7.4	7.4	7.3	7.2	7.2	7.1	7.0	6.9
10	6.9	6.8	6.7	6.6	6.6	6.5	6.4	6.4	6.3	6.2
20	6.2	6.1	6.1	6.1	5.9	5.9	5.9	5.8	5.8	5.7
30	5.6									

Dilution 2: 1.3 mL compacted sludge/mL of slurry

Liquid/slurry interface height (cm)

Time (min)	0	1	2	3	4	5	6	7	8	9
0	7.6	7.5	7.3	7.3	7.2	7.1	7.0	6.9	6.7	6.6
10	6.4	6.2	6.1	5.9	5.8	5.6	5.6	5.3	5.3	5.3
20	5.1	5.0	5.0	4.9	4.7	4.6	4.6	4.6	4.4	4.4
30	4.3									

Dilution 3: 0.9 mL compacted sludge/mL of slurry

Liquid/slurry interface height (cm)

Time (min)	0	1	2	3	4	5	6	7	8	9
0	7.6	7.4	7.0	6.8	6.6	6.4	6.2	5.9	5.6	5.3
10	5.0	4.7	4.4	4.0	3.8	3.7	3.4	3.2	3.0	3.0
20	2.9	2.7	2.7	2.7	2.6	2.5	2.4	2.4	2.3	2.3
30	2.2									

Exhibit A.1 (continued)

Dilution 4: 0.45 mL compacted sludge/mL of slurry

Liquid/slurry interface height (cm)

Time (min)	0	1	2	3	4	5	6	7	8	9
0	7.6	7.3	6.9	6.9	6.5	6.2	5.8	5.5	4.9	4.6
10	4.3	3.8	3.5	3.0	2.7	2.3	1.8	1.5	1.4	1.4
20	1.4	1.3	1.3	1.2	1.2	1.2	1.2	1.2	1.2	1.2
30	1.1									

[a]Sludge: sample W21-S1. Liquid: sample W21-L2.
[b]Position of clear liquid/slurry interface (in cm) from the bottom of a 10-mL graduated mixing cylinder.

Exhibit A.2. Sedimentation test data for sludge from tank W-23[a,b]

Dilution 1: 1.0 mL compacted solids/mL of slurry

Time (min)	0	1	2	3	4	5	6	7	8	9
0	11.9	11.9	11.9	11.8	11.8	11.8	11.7	11.7	11.7	11.7
10	11.4	11.4	11.4	11.4	11.4	11.4	11.4	11.4	11.4	11.4
20	11.4	11.3	11.2	11.2	11.1	10.9	10.8	10.7	10.7	10.6
30	10.5	10.5	10.2	10.2	10.1	10.0	9.9	9.8	9.8	9.6

Dilution 2: 0.8 mL compacted sludge/mL of slurry

Time (min)	0	1	2	3	4	5	6	7	8	9
0	11.9	11.9	11.9	11.7	11.7	11.5	11.4	11.2	11.1	10.9
10	10.7	10.6	10.5	10.4	10.1	10.0	9.8	9.6	9.5	9.4
20	9.2	9.0	8.9	8.8	8.6	8.4	8.3	8.1	8.0	7.9
30	7.7	7.6	7.4	7.3	7.1	7.0	6.9	6.8	6.7	6.7

Dilution 3: 0.6 mL compacted sludge/mL of slurry

Time (min)	0	1	2	3	4	5	6	7	8	9
0	11.9	11.9	11.8	11.7	11.4	11.2	10.9	10.7	10.5	10.2
10	10.0	9.8	9.5	9.3	9.0	8.8	8.4	8.2	8.1	7.9
20	7.6	7.5	7.3	7.1	6.9	6.8	6.5	6.4	6.3	6.2
30	6.0	5.8	5.7	5.6	5.5	5.4	5.2	5.2	5.1	5.0

Exhibit A.2 (continued)

Dilution 4: 0.5 mL compacted sludge/mL of slurry

Time (min)	0	1	2	3	4	5	6	7	8	9
0	11.9	11.9	11.7	11.4	10.7	9.9	9.2	8.4	8.0	7.4
10	6.9	6.4	6.1	5.7	5.5	5.2	5.0	4.9	4.8	4.8
20	4.6	4.5	4.4	4.3	4.3	4.0	4.0	3.9	3.8	3.8
30	3.7	3.6	3.6	3.5	3.3	3.3	3.2	3.1	3.1	3.1

[a]Sludge: sample W23-S1. Liquid: sample W23-L1.
[b]Position of clear liquid/slurry interface (in cm) from the bottom of a 10-mL graduated mixing cylinder.

Exhibit A.3. Sedimentation test data for sludge from tank W-26[a,b]

Dilution 1: 1.2 mL compacted solids/mL of slurry

Time (min)	0	1	2	3	4	5	6	7	8	9
0	11.9	11.9	11.9	11.9	11.9	11.9	11.8	11.8	11.8	11.8
10	11.8	11.8	11.8	11.8	11.8	11.8	11.8	11.8	11.7	11.7
20	11.7	11.7	11.7	11.7	11.7	11.7	11.7	11.7	11.7	11.7
30	11.5									

Dilution 2: 0.8 mL compacted sludge/mL of slurry

Time (min)	0	1	2	3	4	5	6	7	8	9
0	11.9	11.9	11.9	11.8	11.8	11.8	11.7	11.5	11.4	11.2
10	11.1	10.8	10.6	10.4	10.1	9.9	9.6	9.5	9.3	9.0
20	8.8	8.7	8.4	8.3	8.1	8.0	7.9	7.7	7.6	7.6
30	7.5									

Dilution 3: 0.6 mL compacted sludge/mL of slurry

Time (min)	0	1	2	3	4	5	6	7	8	9
0	11.9	11.8	11.7	11.3	10.9	10.5	9.9	9.5	9.0	8.6
10	8.2	8.0	7.6	7.4	7.1	6.8	6.7	6.4	6.3	6.1
20	6.0	5.8	5.7	5.6	5.5	5.5	5.4	5.4	5.2	5.2
30	5.1									

Exhibit A.3 (continued)

Dilution 4: 0.35 mL compacted sludge/mL of slurry

Time (min)	0	1	2	3	4	5	6	7	8	9
0	11.9	11.7	11.4	10.8	10.2	9.5	8.6	7.5	6.3	5.5
10	4.9	4.5	4.3	4.0	3.9	3.7	3.6	3.5	3.5	3.3
20	3.3	3.2	3.2	3.1	3.1	3.0	3.0	3.0	3.0	2.9
30	2.9									

[a]Sludge: sample W26-S3. Liquid: sample W26-L2.
[b]Position of clear liquid/slurry interface (in cm) from the bottom of a 10-mL graduated mixing cylinder.

Exhibit A.4. Sedimentation test data for sludge from tank W-28[a,b]

Dilution 1: 0.65 mL compacted solids/mL of slurry

Time (min)	0	1	2	3	4	5	6	7	8	9
0	11.9	11.9	11.9	11.9	11.8	11.7	11.4	11.4	11.1	10.8
10	10.4	10.1	9.9	9.5	9.2	9.0	8.8	8.6	8.3	8.2
20	8.0	7.9	7.6	7.4	7.3	7.1	6.9	6.7	6.4	6.3
30	6.3									

Dilution 2: 0.45 mL compacted sludge/mL of slurry

Time (min)	0	1	2	3	4	5	6	7	8	9
0	11.9	11.9	11.9	11.9	11.7	11.2	10.8	10.5	10.0	9.5
10	8.9	8.4	8.1	7.6	7.1	6.9	6.7	6.4	6.2	6.1
20	5.8	5.6	5.5	5.2	5.1	5.0	4.8	4.6	4.5	4.5
30	4.4									

Dilution 3: 0.31 mL compacted sludge/mL of slurry

Time (min)	0	1	2	3	4	5	6	7	8	9
0	11.9	11.9	11.9	11.9	11.8	11.4	11.3	11.2	9.5	8.3
10	6.5	5.2	4.5	3.8	3.6	3.3	3.2	3.1	3.0	3.0
20	3.0	2.9	2.9	2.7	2.6	2.6	2.6	2.6	2.6	2.5
30	2.5									

Dilution 4: 0.20 mL compacted sludge/mL of slurry

Note: This slurry was too dilute for solids to coagulate efficiently. The clear liquid/slurry interface was not visible, but the system appeared instead as a cloudy liquid which became less turbid with time.

[a]Sludge: sample W21-S1. Liquid: sample W21-L2.
[b]Position of clear liquid/slurry interface (in cm) from the bottom of a 10-mL graduated mixing cylinder.

APPENDIX B

CALCULATIONAL MODEL USED TO DETERMINE DENSITY AND SOLIDS VALUES

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CALCULATIONAL MODEL USED TO DETERMINE DENSITY AND SOLIDS VALUES

Two portions of each sludge sample were required for a set of measurements. The first portion (about 5 g) was placed into a tared, graduated 15-mL centrifuge tube and weighed. The sludge was centrifuged for 15 min at high speed before reading the total volume and calculating the bulk density:

$$Bulk\ density = \frac{Wet\ mass\ 1}{Wet\ volume\ 1}\ .$$

The separated interstitial liquid was drawn off from the centrifuged sample using a transfer pipet, then filtered through a 0.45-μm filter. One milliliter of the filtered solution was weighed to determine the interstitial liquid density:

$$Liquid\ density = \frac{Solution\ mass}{1\ mL}\ .$$

After being weighed, the 1-mL sample was dried at 110 ± 5°C for 16 ± 1 h and was then reweighed to determine loss of water and other volatiles:

$$Water\ loss\ (1\ mL\ liquid) = Solution\ mass - Residue\ mass.$$

A second portion (about 1 g) of the original sludge was taken. This portion was placed into a tared 10-mL beaker and weighed. The sludge was dried at 110 ± 5°C for 16 ± 1 h, then reweighed to determine the total solids content and loss of water and other volatiles:

$$Total\ solids = \frac{Dry\ mass\ 2}{Wet\ mass\ 2}\ ,$$

$$Water\ loss\ (sludge) = Wet\ mass\ 2 - Dry\ mass\ 2.$$

The dissolved solids content was calculated from the solution residue mass, the water losses from sludge and solution, and the wet sludge mass:

$$\text{Dissolved solids} = \frac{\text{Residue mass}}{\text{Wet mass 2}} \times \frac{\text{water loss (sludge)}}{\text{Water loss (1 mL liquid)}} .$$

The mass of interstitial liquid actually present in the second sludge portion is calculated from the liquid density and the ratio of water lost in drying the sludge to that lost in drying the liquid:

$$\text{Liquid mass} = \text{Liquid density} \times \frac{\text{water loss (sludge)}}{\text{water loss (1 mL liquid)}} .$$

The undissolved solids content was calculated by difference, as follows:

$$\text{Undissolved solids} = \text{Total solids} - \text{Dissolved solids.}$$

The undissolved solids density was also calculated as the ratio of differences in mass and volume:

$$\text{Floc density} = \frac{\text{Wet mass 2} - \text{Liquid mass}}{\frac{\text{wet mass 2}}{\text{bulk density}} - \frac{\text{water loss (sludge)}}{\text{water loss(1 mL liquid)}}} .$$

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