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# GRAVITY SETTLING OF HANFORD SINGLE-SHELL TANK SLUDGES

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## ABSTRACT

The U.S. Department of Energy plans to use gravity settling in million-gallon storage tanks while pretreating sludge on the Hanford site. To be considered viable in these large tanks, the supernatant must become clear, and the sludge must be concentrated in an acceptable time. These separations must occur over the wide range of conditions associated with sludge pretreatment. In the work reported here, gravity settling was studied with liter quantities of actual single-shell tank sludge from Hanford Tank 241-C-107. Because of limited sludge availability, an approach was developed using the results of these liter-scale tests to predict full-scale operation. Samples were centrifuged at various *g*-forces to simulate compaction with higher layers of sludge. A semi-empirical settling model was then developed incorporating both the liter-scale settling data and the centrifuge compression results to describe the sludge behavior in a million-gallon tank. The settling model predicted that the compacted sludge solids would exceed 20 wt % in less than 30 days of settling in a 10-m-tall tank for all pretreatment steps.

## INTRODUCTION

The U.S. Department of Energy's Hanford Site in southeastern Washington state has 177 underground storage tanks that contain wastes from approximately 50 years of reprocessing nuclear fuel and managing waste. The contents of these tanks will be disposed of either offsite as high-level waste (HLW) in a deep geologic repository or onsite in surface burial grounds as low-level waste (LLW). Because the cost to dispose of the HLW fraction is expected to be high, the waste will be pretreated before being immobilized to minimize the quantity of HLW generated.

The tanks at the Hanford Site contain a mixture of supernate, water-soluble salt cake, and water-insoluble sludge. The salt cake and supernate will be processed to remove cesium, and possibly technetium, and then immobilized as LLW. The tank sludges, on the other hand, contain the long-lived radionuclides and will be disposed of as HLW. To minimize their impact on the final HLW volume, those sludges may be retrieved from their current storage tanks and pretreated by enhanced sludge washing (ESW). The retrieval process will fluidize the sludge in the tank using a high-pressure and high-temperature spray. The sludge/water mixture will then be transferred to another large tank for ESW. The ESW process first washes the sludge with hot caustic (3 *M* NaOH) and then with inhibited water (0.01 *M* NaOH/0.01 *M* $NaNO_2$). These steps remove the caustic- and water-soluble metals from the sludge, leaving the radionuclides behind. The washes will be combined with the supernates as non-transuranic (TRU) LLW, and the sludge can then be immobilized as a smaller volume of HLW.

During each step in the ESW process, solid/liquid separation techniques will be required. The baseline process for these separations is gravity settling. Both the gravity settling and the ESW may be performed in Hanford's million-gallon double-shell tanks. If this is the case, gravity settling in these tanks must provide efficient separation in an acceptable period of time. The feed staging plan and flow sheets for the Hanford site assume that the compacted solids will exceed 20 wt % solids in less than 30 days (1).

Because of the difficulties in obtaining and handling large samples of sludge, a system was developed at Pacific Northwest National Laboratory (PNNL) to measure sludge-settling properties on a liter scale and use these data to extrapolate settling performance to a full-scale, million-gallon double-shell tank. The settling column must be tall enough to allow for accurate measurements of settling rate while having a sufficiently large diameter to prevent the walls from affecting the settling rate. Furthermore, once the settling and compaction-rate data are obtained, they must be extrapolated to provide values for a full-scale "settle/decant" system. PNNL developed a model to process these data and estimate the sludge compaction on a large scale.

In this paper, we report our investigation on the settling properties of sludge from Hanford Tank 241-C-107 (the 241 prefix, which is common to all Hanford waste tanks, will not be used hereafter). The settling rate is evaluated for various stages of the ESW process. The data are further used in a model to estimate the settling properties of sludge in a 10-m-tall Hanford double-shell tank.

## THEORETICAL ANALYSIS

The sedimentation rates of a pilot-scale process are useful only if they can be extrapolated to large production-scale systems. A transient sedimentation model has been developed that incorporates the primary features necessary for accurately predicting the sedimentation behavior of a tank-waste settling column. Both the sedimentation test results and the analytical data are used to determine the appropriate coefficients for each stage in the ESW procedure.

Hanford tank waste contains particles of widely varying size and composition. The smallest particles are less than a micron ($\mu$m) in diameter and may exhibit colloidal behavior, while the largest particles may be hundreds of microns in diameter. Depending on the chemistry of the solution, the small colloidal particles may aggregate to form large porous flocs. The rate of sedimentation of each individual particle or floc depends on its size and relative density. When sedimentation begins, the large, dense particles and flocs quickly settle to the bottom. Therefore, the small particles and low-density flocs control the rate of sedimentation.

Samples such as those used in this work, which have a relatively high solids loading, exhibit a sharp, well-defined interface that appears almost immediately between the clear supernatant liquid and an opaque region that contains the suspended solids. For such suspensions, the sedimentation velocity is monitored by noting the position of the interface as a function of time. As the sediment settles, the sediment layer becomes thinner, and the average solids loading in the sediment layer increases until the sediment compresses to its equilibrium solids-loading profile.

When the volume fraction of the solids ($\phi$) exceeds a specific value, known as the gel point, $\phi_g$, the agglomerates form a network, and the suspension takes on the form of a solid structure. Compressive stresses on the system can be transmitted via the network, and the structure can then, at least partially, support itself. In this case, the compression rate of the sediment is controlled by a combination of the hydrodynamic drag of the interstitial fluid being squeezed out of the network and particle bonds breaking and re-forming as the agglomerates are being crushed by the weight of the sediment above.

A computational sedimentation model predicts the solids density profile as a function of time, based on information obtained from both settling experiments and laboratory tests of the suspensions of interest. From this information, we can derive the height of the sediment as a function of time. To be accurate, this model must reflect two major aspects of the sedimentation process as follows:

- *Hindered Settling* – The settling rate of a suspension for a given particle distribution and solution chemistry depends only on the local solids loading. The rate is independent of the overall dimensions of the system. For example, if a 5 wt % particle suspension settles at 5 cm/h in a liter-scale column (assuming no wall effects), it will settle at 5 cm/h in a full-scale tank until it reaches the sediment layer. An expression must be developed that relates local solids loading to the hindered settling rate.
- *Sediment Compression* – As the total solids loading per unit area increases, the final height of the sediment increases. However, as the additional weight of solids is added, the sediment is compressed, resulting in a higher average solids density in the sediment. An expression must be developed that relates the local solids density to the compressive force on the sediment.



## Hindered Settling

The sedimentation model divides the system into two regions. In the upper region, the solids concentration is below the gel point, $\phi_g$, for that suspension, and the particle agglomerates interact only through hydrodynamic forces. The velocities, u, of the agglomerates in this region were taken from Buscall and White (2) as

$$u = \frac{u_0(1-\phi)}{r(\phi)} \tag{1}$$

where $u_0$ is the Stokes settling velocity at infinite dilution, and $r(\phi)$ is a dimensionless hydrodynamic interaction parameter. The term $(1 - \phi)$ results from the fact that the volume displacement of downward-flowing solids must be compensated by an equal upward volume flow of solution. The term converts the relative velocity of solids to solution into a reference-frame velocity. The Stokes settling velocity, $u_0$, for solid spherical particles is given by the expression

$$u_0 = \frac{2a^2 \Delta\rho g}{9\eta_s} \quad (2)$$

where a is the particle radius, $\Delta\rho$ is the solid-liquid density difference, and $\eta_s$ is the solution viscosity. For particle agglomerates containing many primary particles, the radius becomes the effective agglomerate radius, and the density is given by the relative density of the agglomerate. The hydrodynamic interaction parameter, $r(\phi)$, can take many forms, but one possible expression is

$$r(\phi) = (1 - \frac{\phi}{\phi_{ref}})^n \quad (3)$$

where $\phi_{ref}$ is a reference volume fraction. For particle agglomerates, the value for $\phi_{ref}$ must be greater than the gel point for that system.

Using experimental hindered settling data over a range of solids concentrations ($\phi$), the parameters $u_0$, $\phi_{ref}$, and n can be determined. The Stokes settling velocity expression generally is applied to monodispersed particle-size systems. However, since the interface height is controlled by the settling rate of the smallest particle size, these equations can also be applied to the polydispersed sludges studied in this work.

## Sediment Compression

As discussed above, when the particle volume fraction is sufficiently high, a network of connected aggregates forms, and the suspension takes on the form of a solid structure. In particular, compressive stresses on the system can be transmitted via the network throughout the system, and the structure then possesses the ability to support itself. As the network pressure, P, is increased, either mechanically with a piston or through gravitation forces, the network structure will resist further compression until the forces become strong enough that the structure begins to deform irreversibly. This network pressure at any vertical location is the relative weight per unit area of the sediment above that location. The relative weight, in turn, is calculated by multiplying the integral of the volume fraction of solids above the location of interest by the acceleration of gravity, g, and by the difference between the solid and liquid densities.

The compressive yield stress $P_y(\phi)$ is defined as the value of the network pressure at which the flocculated suspension at volume fraction, $\phi$, will no longer resist compression elastically and will start to yield and irreversibly consolidate. The compressive yield stress is an implicit function of many variables, including the size, shape, composition, and relative number of particles involved and the interparticle forces (which, in turn, depend on the solution chemistry). At concentrations less than the gel point, the aggregates are not connected and act as independent units. At the gel point, these aggregates become interconnected throughout the container to the extent that they are able to support a load. At concentrations greater than the gel point, the compressive yield stress is typically modeled using a power law curve of the following type:



$$P_y(\phi) = c\left[\left(\frac{\phi}{\phi_g}\right) - 1\right]^m \qquad \phi > \phi_g \tag{4}$$

with m varying between 4 and 10 (3).

The parameters c and m for the power-law curve may be determined using equilibrium sediment-height data when the network pressure (P) is equal to the compressive yield stress $P_y(\phi)$. The only data required are the solid and liquid densities, the overall weight or volume percent of particulate solids in the sediment, and the final sediment height. The primary disadvantage of relying only on standard sediment-height data is that the range is limited by the heights of the test columns used, which are typically much smaller than the full-scale applications that we wish to model.

The range of sediment compression data can be extended by measuring the sediment heights of samples that have been centrifuged at different speeds. For these centrifuge tests, the compressive yield stress is based on the integral of the relative artificial weight of the solids created by the centrifuge at each location in the sediment. These data, together with the equilibrium gravity sedimentation data, are used to determine the expression parameters. A computer program has been written to optimize the power-law parameters (c, $\phi_g$, and n or m) by performing a least-squares fit based on the sediment heights using a simulated annealing approach. The exponents are restricted to the ranges specified above.

## Overall Sedimentation Model

The sedimentation model divides the system into two regions. In the upper region, the solids concentration is below the gel point, $\phi_g$, for that suspension, and the particle agglomerates interact only through hydrodynamic forces. The velocities of the solids in this region are expressed in Equation (1).

In the lower region, the solids concentration is above the gel point, $\phi_g$, and the particle agglomerates interact through both hydrodynamic forces, represented by r($\phi$), and solid network pressure, P. The velocities of the agglomerates in this region are given by the expression taken from Buscall and White (2)

$$u = \frac{u_0(1-\phi)}{r(\phi)}\left(1 + \frac{\partial P/\partial z}{(\Delta\rho g\phi)}\right) \qquad (5)$$

where P is the network pressure at elevation z, and the term $\Delta\rho g\phi$ is the change in gravitational head per unit elevation. Note that for regions that have no network pressure, the last term is zero. For sediments that have reached equilibrium, the change in network pressure is equal to the negative of the change in gravitational head, resulting in a net velocity of zero.

In Equation (4), we described the compressive yield stress, $P_y(\phi)$, of a suspension. If the network pressure, P, is less than or equal to the compressive yield stress, the network is strong enough to support the weight of the sediment, and no change occurs. However, when the network pressure exceeds the compressive yield stress, the network consolidates irreversibly until the volume fraction, $\phi$, increases to the point where the yield stress equals the network pressure. This changes at a rate controlled by the dynamic compressibility, $\kappa(\phi)$. The network velocity is controlled by the expression from Buscall and White (2)

$$\frac{\partial u}{\partial z} = \frac{\kappa(\phi)}{\phi}\left[P - P_y(\phi)\right] \quad P \geq P_y(\phi) = 0 \;\; P < P_y\,(\phi). \qquad (6)$$

When Equation (6) is substituted into this expression, we obtain a second-order differential equation for the network pressure,

$$\frac{\partial}{\partial z}\left[\frac{(1-\phi)}{r(\phi)}\left(1+\frac{\partial P/\partial z}{(\Delta\rho g\phi)}\right)\right] = \frac{\kappa(\phi)}{u_0}\,[P - P_y(\phi)] \qquad (7)$$

where the right-hand term is zero when $P < P_y(\phi)$.

A computational model has been developed that combines the hindered settling model, the network pressure model, and the aggregation kinetics model (when appropriate) to predict the sedimentation behavior of suspensions. Both time and elevation are discretized using a finite-difference formulation. The following procedure is followed for each time step:

- The total solids volume fraction, $\phi$, is calculated at each elevation based on a measured value of the overall mixture and mass balance equations. If the total solids volume fraction exceeds the gel point ($\phi > \phi_g$), the node is considered part of the sediment layer. The elevation node that represents the top of the sediment layer is located. This divides the system into two regions.
- In the upper region, the hindered settling velocities are calculated by using expressions of the form presented in Equation (1). The velocities are used to calculate solids transport from one elevation to another using an upwind-differencing formulation. Because this term is explicit in time, the time step, $\Delta t$, is restricted by the Courant limit (4),

$$\Delta t = \frac{\Delta x}{u} \qquad (8)$$

- In the lower region, the network pressure is calculated using Equation (7). The network pressure at the top of the sediment layer is assumed to be zero. The network values allow the sediment velocity at each elevation to be calculated using Equation (6), which is also subsequently applied to the calculation of the solids transport for that time step.

The unknown parameters that must be defined to use this model for any particular suspension are the compressive yield stress, $P_y(\phi)$; the hydrodynamic interaction parameter, $r(\phi)$; and the dynamic compressibility, $\kappa(\phi)$. The compressive yield stress curve may be determined using equilibrium sedimentation data and centrifuge data for different total solids content, as described above. The hydrodynamic interaction parameter may be estimated by a fit of transient sedimentation data at different initial solids concentrations. The dynamic compressibility may also be estimated using these data. Experience has shown that the sedimentation behavior is relatively insensitive to the form used for the dynamic compressibility and becomes noticeable only in the final stages of sediment compression.

## EXPERIMENTAL STUDY

A hot cell was used for gravity settling of the single-shell tank sludge from Tank C-107. The major equipment consisted of three tanks plus associated piping and pumps, as shown in a schematic in Figure 1. A total of 800 grams of as-received sludge from Hanford Tank C-107 were studied. The vessel for sludge settling, 10 cm in diameter and 1 m tall, was constructed of polysulfone, which is a transparent polymer resistant to boiling caustic and radiation. A fluorescent light was placed behind the settling column to improve visualization of the solid/liquid interface. A ruler, visible from the cell window, was attached to the column to observe the slurry/liquid interface level. Temperature control was provided by circulating water from a hot-water bath through an annulus surrounding the sludge-settling column. The interfaces were observed by time-lapse photography. The free or hindered settling rate was calculated from the early linear part of the settling curve, and the solids fraction in the final compacted sludge was determined at the end of the run.

A separate 3-gallon tank was used for ESW. This tank was equipped with an agitator, heater, thermocouples, inlet and outlet lines, and a port through which the tank waste sample was transferred. Initially, the virgin sludge was placed inside the ESW tank. Either water or caustic was then added to the sludge to perform each ESW step: a simulated retrieval step, followed successively by two caustic leaching steps and two inhibited-water rinse steps. The volume of water or caustic used in each step was determined based on the desired solids concentration. The target conditions of these steps are shown in Table 1. The solution was heated and agitated for the required length of time and then pumped from the ESW tank into the column for the settling studies.

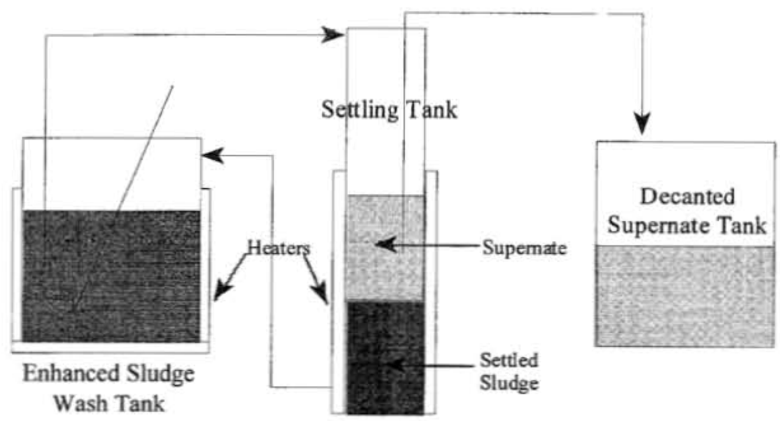


**FIGURE 1. Schematic of settling equipment used for actual waste testing.**

**TABLE 1. TARGET CONDITIONS OF THE ESW AND SETTLING**

| Step | Solids concentration (wt %) | Chemical concentration | Temperature and time of ESW | Temperature and solids conc. of settling |
|---|---|---|---|---|
| Retrieval | 10 | 0.01 *M* NaOH; 0.01 *M* $NaNO_2$ | 100°C 0.5 hour | 85°C 10% |
| Caustic leach 1 | 5 | 3 *M* NaOH | 100°C 5 hours | 85°C 5% |
| Caustic leach 2 | 5 | 3 *M* NaOH | 100°C 5 hours | 85°C 5% and 8% |
| Water wash 1 | 5 | 0.01 *M* NaOH; 0.01 *M* $NaNO_2$ | 50°C 0.5 hour | 50°C 5% |
| Water wash 2 | 5 | 0.01 *M* NaOH; 0.01 *M* $NaNO_2$ | 50°C 0.5 hour | 50°C 5% and 8% |

The solids concentration was based on the insoluble solids present in the slurry during each process step. The insoluble solids concentration was determined by removing a sample of the slurry mixture, centrifuging the sample, removing the supernatant, and drying the solids at 105°C to a constant weight. The solids concentration was combined with the particle and supernate densities to determine the overall volume fraction solids, $\phi$, used in the sedimentation model described above.

One or more settling tests followed each step in the ESW process. After the second caustic leach and second water wash, the settling tests were repeated at higher solids concentrations and at lower temperatures to provide the necessary data to fit the hydrodynamic interaction parameter described in Equation (3). This settling test at higher solids concentrations was performed by decanting a fraction of the supernatant, fluidizing the mixture, and repeating the settling test.

After the sludge had completely settled, the supernatant was decanted, and water or caustic solution was added to the mixture in preparation for the next ESW step. The slurry was then fluidized and transferred back into the ESW tank for agitation and heating.

Slurry samples taken after the second caustic leach and the second water wash were placed in a test tube and centrifuged over a range of speeds. The revolutions per minute were converted to gravitational acceleration. Sediment height was measured for increasing values of acceleration from 0 to 10,000 $m/s^2$. These experiments provided an understanding of sludge compaction at higher gravitational forces and were used to obtain the compressive yield stress data at higher network pressures as described in Equation (4).

## RESULTS AND DISCUSSION

### Results of the Sludge-Settling Experiments

As discussed above, samples of C-107 sludge were allowed to settle for each step in the ESW process. Settling tests were performed for the second caustic leach and second water wash for two solids concentrations. The settling conditions, rates, and compacted solids concentrations are provided in Table 2.

TABLE 2. C-107 SETTLING TEST RESULTS

| Step | Initial solids (wt%) | Temperature (°C) | Settling rate (cm/h) | Final compaction (vol %) | Final compaction (wt %) |
|---|---|---|---|---|---|
| Retrieval | 7.6 (9.4) [a] | 85 | 4.1 | 36 | 20 (24) [a] |
| Caustic 1 | 5.4 | 85 | 15.5 | 24 | 21 |
| Caustic 2 | 5.8 | 85 | 9.1 | 21 | 21 |
| Caustic 2 | 8.7 | 27 | 1.0 | 43 | 18 |
| Caustic 2 | 8.8 | 85 | 4.6 | 43 | 20 |
| Wash 1 | 5.7 | 50 | 11.7 | 20 | 25 |
| Wash 2 | 5.7 | 50 | 12.9 | 19 | 25 |
| Wash 2 | 5.5 | 27 | 7.6 | 18 | 23 |
| Wash 2 | 9.3 | 50 | 7.4 | 33 | 25 |

[a] All wt % solids are based on caustic leach-insoluble solids except those in parentheses, which represent water-insoluble solids.



In all the Sludge Pretreatment Demonstration experiments, the sludges settled with single, very distinct interfaces between the solids and the supernates. The light supernatant was clearly distinguishable from the dark sludges in each case. This interface generally developed within the first 20 minutes of settling and within the first 5 cm from the top of the settling column. In each experiment, the hindered settling was completed, and compaction began within the first 3 h after settling commenced.

In each case, the sludge settled in the column. This is in contrast to test-tube-scale settling tests performed by Lumetta et al. (5) with C-107 sludge. In these experiments, the sludge did not settle at all for the two caustic leaches and three water washes that were performed with material containing between 7 and 9 wt% solids. The difference could be attributed to the difference in the diameters of the settling containers. The sludge in the small container may have clung to the sides to form a colloidal bridge network across the sludge container, which would prevent the solids from settling (i.e., wall effects).

Typical settling curves for the test runs are shown in Figure 2. Generally, the curves are very similar. The initial hindered settling is linear, followed by the asymptotic compaction regime. The linearity of the hindered settling regime allowed accurate regression of the hindered settling rates shown in Table 2.

The initial hindered settling rate appears to depend strongly on concentration. Increases in solids concentration resulted in a greater-than-proportionate decrease in the settling rate. The settling rate generally appeared to increase with each subsequent ESW step. In spite of the decreased settling temperature of the washes (50 °C rather than 85 °C), the settling rates are fairly comparable.



## Results of Theoretical Settling Model

The results obtained from both the settling tests and the laboratory analysis were used to develop the expressions used in the following theoretical settling model. The results presented in this section are restricted to systems with the same particle size, the same component distribution, and the same solution chemistry as provided in the Tank C-107 sample.

## Hindered Settling

Expressions predicting the hindered settling rate as a function of solids loading were developed for both the caustic leach and the wash steps, based on the forms presented previously. The coefficients were determined by performing a least-squares fit on the measured settling rates. The hindered settling rates, in cm/h, for all caustic leach tests are given by the expression

$$u = \frac{42.4\,(1-\phi)}{\left(1-\frac{\phi}{0.11}\right)^{-4.2}} \qquad \phi < \phi_g \tag{9}$$

where the suspension temperature is 85°C. Values for the solid volume fractions, $\phi$, were determined using the measured supernate density and an assumed solid particle density of

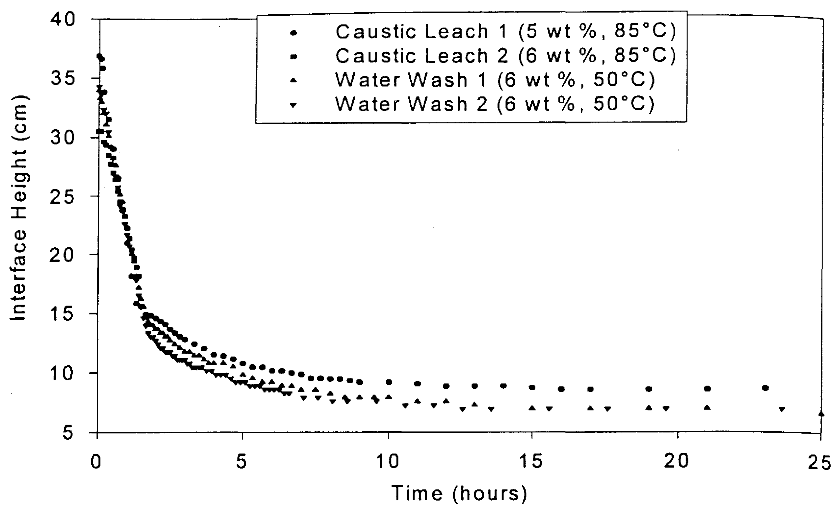


**FIGURE 2. Settling results with C-107 sludge for each step of the ESW process.**



2.5 $gm/cm^3$.[a] The hindered settling rate in cm/h for all wash tests is given by the expression

$$u = \frac{29.5\,(1-\phi)}{\left(1-\dfrac{\phi}{0.13}\right)^{-3.6}} \quad \phi<\phi_g \tag{10}$$

where the suspension temperature is 50°C. It is not surprising that the hindered settling expressions for the caustic leach step would be different from that of the water wash. The higher ionic strength of the caustic solution reduces the size of the electrical double layer, resulting in a change in the size and density of the particle aggregates.

The magnitude of these velocities provides some information on the extent to which the particles are aggregating in solution. Particles size measurements were performed

[a] Solid density measurements were attempted by helium pycnometry, but the results were inconclusive. The 2.5 $g/cm^3$ is an estimate based on slurry densities and wt % solids.

using Fraunhofer diffraction and photon correlation spectroscopy. From these measurements, we know that a significant portion of the insoluble solids exists in the form of submicron particles, down to at least 0.1 μm in diameter. Using typical solid (2.5 $g/cm^3$) and solution densities (1.1 $g/cm^3$), the settling velocity using Stokes' equation for individual 1-μm particles is on the order of 0.3 cm/h. The settling velocity for 0.1-μm particles is 0.003 cm/h, as compared with 15 cm/h for the liter-scale settling tests. The measured settling velocities can only be reached if these small (micron sized) particles aggregate to form flocs, which behave like large porous particles. It is estimated that flocs of at least 500 such particles are required to achieve the measured settling velocities. With a fractal dimension of 2.5, the floc diameter would be between 10 and 30 μm.



## Compressive Yield Stress

Expressions predicting the compression yield stress as a function of solids loading were developed for both the caustic leach and wash steps based on the forms in Equation (4). The coefficients were determined by performing a least-squares fit on both the measured equilibrium sediment heights resulting from the settling tests and those from centrifuge data. The compressive yield stress, in $g/cm^2$ for all caustic leach tests, is given by the expression

$$P_y(\phi) = 41.937\left[\left(\frac{\phi}{0.079}\right) - 1\right]^{4.0} \qquad \phi > 0.079 \tag{11}$$

The compressive yield stress in $g/cm^2$ for all wash tests is given by the expression

$$P_y(\phi) = 5.356e3\left[\left(\frac{\phi}{0.0425}\right) - 1\right]^{6.88} \qquad \phi > 0.0425 \tag{12}$$

These expressions were validated by calculating the sediment height for each sedimentation test or centrifuge measurement and comparing these values with data obtained in the experiments. The calculations for the sedimentation tests were based on the estimated solids loading provided by the mass balance calculations. In almost all cases, the predicted sediment heights were within 10% of the measured values. Such results give confidence that these expressions will give accurate sediment-height predictions for both high and low solids-loading situations.

### Transient Sedimentation Model

The hindered settling-rate expressions and the compressive yield-stress expressions are combined with the transient sedimentation model described previously to provide a computational model for predicting the sedimentation behavior of Tank C-107 pretreatment settle-decant systems.

The model was validated by comparing model results with the recorded settling data for the actual C-107 sludge. The model was used to calculate the entire density and network-stress profile at each moment in time. The location of the top of the sediment was interpolated from the density profile. Examples of these comparisons are shown in Figure 3 for the first caustic-leach and first water-wash cases. In each case, the initial settling velocity and the final sediment height appear to be relatively well predicted. The region of greatest discrepancy is the transition area between the settling of particles before contact with the sediment layer and the slow compression of the sediment layer. The computational model consistently underpredicts the sediment height in this region. The reason for this underprediction may be the formation of metastable network structures or some other dynamic effect not accounted for in a computational approach that uses equilibrium compressive-yield-stress information. Further study adjusting the value of the dynamic compressibility ($\kappa$) may allow an improved fit of these curves.



### Extrapolation to Full-Scale Tank Settling

The usefulness of this model is demonstrated by predicting the settling behavior of pretreatment settle-decant operations performed in a full-scale Hanford HLW tank. The height of the suspension is assumed to be 10 m, which is roughly equivalent to the height of existing waste in many of the double-shell tanks. Predictions were made for both the caustic leach and the wash steps (see Figures 4 and 5). It should be reiterated that these results apply only to C-107 and similar sludges over the conditions investigated here.

The caustic-leach simulation was performed assuming both 5 and 10 wt % solids loading and a temperature of 85°C. It took approximately 2 and 3.5 days for all the freely falling solids to contact the sediment layer at 5 wt % and 10 wt % solids, respectively.

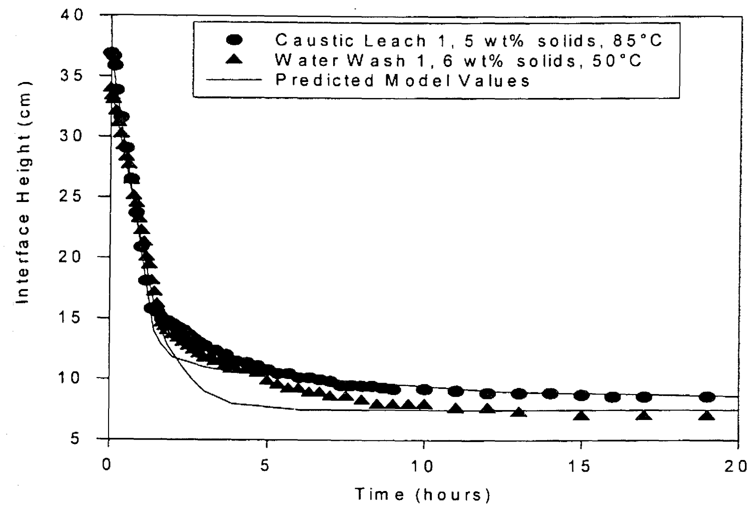


**FIGURE 3. Comparison of predicted and measured sludge interface heights for the first caustic leach and the first water wash.**

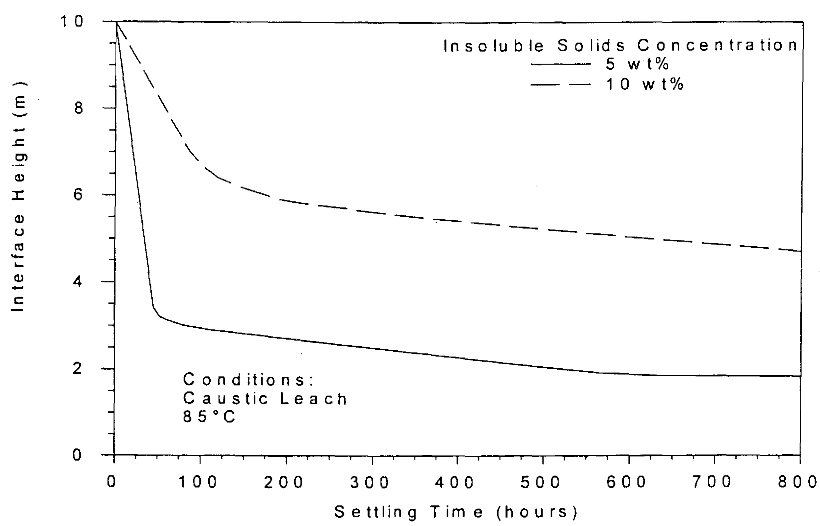


**FIGURE 4. Prediction of sludge interface height vs time for C-107 caustic leach in a full-scale tank.**

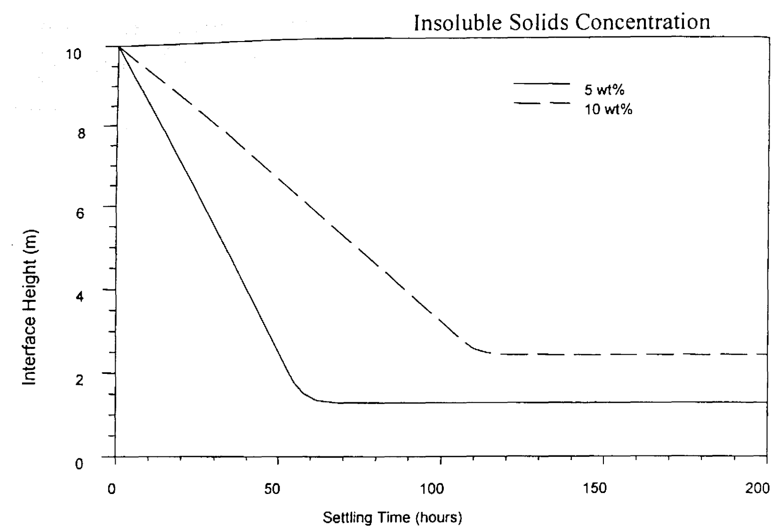


**FIGURE 5. Prediction of sludge interface height vs time for C-107 water wash in a full-scale tank.**



The sediment layer compressed slowly with time. In the case of 5 wt % solids loading, average densities of 20 wt % and 25 wt % were reached after 400 and 800 h, respectively. In the case of 10 wt % solids loading, compaction took considerably longer, reaching an average density of 20 wt % after 680 h and 27 wt % after approximately 1000 h.

The wash simulation was performed assuming two solids loadings (5 wt % and 10 wt %) and a temperature of 50°C. It took approximately 2.5 days for all the freely falling solids to contact the sediment layer. In each case, the sediment layer quickly (within 2 and 4 days, respectively) reached its final equilibrium sediment density of approximately 32.7 and 34.9 wt % for the 5 and 10 wt % solids loading cases. The sediment compression behavior for the two cases is different because the caustic-leach sediment was more resistant to compression and required more time to reach equilibrium.

For both the caustic-leach and the water-wash cases, the final sediment concentration was higher for the million-gallon tank than for the liter-scale process. This was to be expected because the greater sludge depth would allow increased compaction of the sludge.

## CONCLUSIONS

The gravity settle/decant process was performed with Hanford sludge from Tank C-107 during each stage of the ESW process. The sludge settled in a single, distinct interface in each case. This study focused on scaling up the liter-scale settle/decant process to a million-gallon tank. Sludge compaction was measured both in the liter-scale column under standard gravitational conditions and in centrifuge tubes using high gravitational forces. All of the resulting data were utilized in a semi-empirical settling model to develop expressions for settling rates and compressive yield stress for greater sludge depths.

The data obtained with this model appear reasonable and are consistent with expected results. The solids in the compacted sludge for a 10-meter tank were in the upper 20 wt % range for the caustic leach steps and the lower 30 wt % range for the wash steps. The time for the sediment to reach 20 wt % solids varied from 2.5 days for the water wash at 5 wt % solids initial loading to 30 days for the caustic leach at 10 wt % solids initial loading.

These results help us understand the settling behavior of the C-107 tank sludge and other similar sludge types; however, they cannot be applied across the board for all Hanford sludges or for sludges at other DOE sites. However, this same scale-up approach should be used to study other types of sludge, thereby providing an accurate picture of the feasibility of the gravity settle/decant process in the DOE complex.



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