Physical Behavior of Uranium-Metal-Bearing Hanford K East Basin Sludge Materials

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Abstract

Uranium-metal-bearing sludge from the Hanford's K-East (KE) Basin is to be retrieved, loaded into large-diameter containers, and moved to interim storage in a dry cell at T Plant on the Hanford site. Physical behavior of this sludge during loading and subsequent storage in large-diameter containers is of interest to design and safety because oxidation of its uranium generates power and hydrogen gas, with resulting implications for flammability of the container and cell headspaces, potential retention of gas in the settled sludge and subsequent expansion of the sludge material in the large-diameter container, and the potential for local temperature escalation. Key aspects of experimental work and model development necessary to support a robust technical basis for design and safety analyses are reported here: (1) Experimental data supporting the distribution of uranium metal in the large-diameter container, (2) Experimental data defining sludge thermal conductivity and shear strength (or yield stress), and (3) Experimental data and models demonstrating sludge plug movement, breakup, and limited atomization caused by internal gas generation.

Introduction

Metallic uranium Spent Nuclear Fuel (SNF) is currently stored in two water-filled concrete pools, 105-KE Basin (KE Basin) and 105-KW Basin (KW Basin), at the United States Department of Energy (U. S. DOE) Hanford Site, in southeastern Washington State. The Spent Nuclear Fuel Project (SNF Project) is responsible to DOE for operation of these fuel storage pools and for the 2100 metric tons of SNF materials that they contain. These fuel storage pools also contain hazardous substances that primarily result from the degradation of the SNF. The hazardous substances consist of the SNF, sludge, debris, and water.

Both the KE and the KW Basins contain contaminated [i.e., radioactive, polychlorinated biphenyl (PCB), etc.] sludge (~52 m^3). Sludge on the floor and in the pits of the KE Basin is a mix of fuel corrosion products (including metallic uranium, and fission and activation products), small fuel fragments, iron and aluminum oxide, concrete grit, sand, dirt, operational debris and biological debris [1, 2, 3, 4]. The large quantity of fuel corrosion products in the KE Basin floor and pit sludge is a result of the open tops, and in many cases open-screened bottoms, of the fuel storage canisters. Because the SNF stored in the KW Basin was placed in closed containers before storage, most of the corrosion products were retained within the canisters and the sludge buildup in the KW Basin is of much smaller volume than that in KE Basin. The small quantity of sludge on the floor of the KW Basin is assumed to consist primarily of dust and sediment; the floor sludge is not expected to contain significant amounts of fuel corrosion products because the KW Basin canisters have closed tops and bottoms. Sludge in the KE and KW Basin fuel storage canisters (i.e, canister sludge) consists primarily of fuel corrosion products. For the purposes of differentiating SNF and debris from sludge, any material that will pass through a screen with 0.64 cm (0.25 in.) openings is defined as sludge.

The Hanford K Basin sludge will be managed as two general waste streams: K East (KE) floor, pit, and canister sludge containing relatively low concentrations of fuel particles; and K West (KW) sludge, some of which will contain relatively high concentrations of fuel particles and graphoil. The KE sludge (~43 m^3), which comprises the majority of the total K Basin sludge inventory, will be loaded into Large-Diameter Containers (LDCs) (approximately 5 ft in diameter and 10 ft high) and stored in process cells at T Plant in the Hanford 200 Area. The KW sludge will also be stored at T Plant; however, the storage container configuration has not been specified.

The presence of metallic uranium fuel particles in the K Basin sludge creates the primary technical challenge to the design of the storage systems. The metallic uranium and uranium oxides within the sludge will corrode, hydrate, and, consequently, generate heat and hydrogen gas during storage. Additionally, heat is also generated within the sludge by radiolytic decay. To maintain thermal stability, the sludge must be retrieved, staged, transported, and stored in systems designed to provide a rate of heat removal that prevents the temperature in the sludge from rising above established limits (currently defined as below the sludge boiling point).

This paper describes experimental testing and modeling to determine and predict (1) the uranium metal distribution in KE sludge after loading into a container, (2) physical properties of the sludge (shear strength and thermal conductivity), and (3) behavior of the sludge caused by gas buildup during containerization and storage. Work reported here was performed by Pacific Northwest National Laboraties (PNNL) and Fauske & Associates, Inc. under contract to the Fluor Hanford Spent Nuclear Fuel (SNF) Project.

Settling/Uranium Distribution Tests

Large-scale settling test was conducted by PNNL with a K Basin sludge simulant that included metallic tungsten/cobalt (W/Co) fragments (density ~14.5 g/cm^3) as a surrogate for uranium metal (density 19 g/cm^3) [5]. The objective of the testing was to gain insight into how uranium metal is likely to be distributed within the K Basin sludge loaded into the large-diameter containers (LDCs) that will be used for storage at T Plant. In the LDCs, uranium metal will react with water and generate heat and hydrogen gas. During loading, transportation, and storage operations, the uranium metal distribution in the LDCs will have an impact on the thermal stability.

A five-component simulant was formulated for the settling test to approximate the average particle size distribution and particle density of a 40/60 volume basis mixture of K East Basin canister and floor sludge (Figure 1). For the test, 20 jars (batches) were prepared, containing 1 liter of sludge simulant each. A clear acrylic column, 1 ft in diameter (OD) and 5 ft tall, was used as the settling vessel (Figure 2). To simulate flow to the LDC, supernatant in the upper part of the settling column was recirculated using a peristaltic pump. Before the test was started, three closed-bottom glass cylinders (2.5 in. OD x 12 in. high] were placed on the bottom of the column to collect core samples of the settled sludge.

For the test, a batch of sludge (1 L) was added to the top of the column (uniformly over the cross section of the column) once every 20 minutes. The sludge settled through 4 to 5 ft of water/slurry before accumulating on the bottom of the column. Twenty minutes after the last batch addition, the recirculation loop was turned off, and the suspended particulate was allowed to settle. The final settled sludge depth in the column was about 11.5 in.

About 40 hours after the last sludge addition, clarified supernatant was removed and the lower portion of the column (containing the settled sludge) was transported (about 3 miles) via a pickup truck for X-ray nondestructive evaluation (NDE) analysis. Because X-rays could not penetrate the dense W/Co, the distribution of W/Co could be examined within the settled sludge matrix. However, the X-rays also could not penetrate the entire column thickness; consequently, images were captured by focusing the X-ray about 2 to 3 in. in from the edge of column. The column was rotated on a turntable to provide 360 degree imaging. Once the imaging of the column was completed, the closed-bottom sample cylinders were removed from the sludge column, and X-rays taken.

Results from Settling/Uranium Distribution Tests

Twenty well-demarcated sludge layers were formed in the settled sludge (Figure 3). Each layer corresponded to the addition of one batch of sludge simulant. Within these primary layers, sub-layers, enriched in one or more of the simulant components were visible. The primary layers were about ½ in. thick, with the exception of the top layer. The top layer was about 1.1 in. thick as a result of the additional particulate that settled after the recirculation pump was shut off. These settling results are in are in accord with the overall character predicted by a settling model [6]. This model predicts that when sludge is added to the LDC in serial additions, each batch forms a layer segregated into sub-layers enriched in and lacking in uranium metal.

Based on observations of the layers, the overall settled sludge depth, and the manner in which the sludge additions were made, the upper 10% to 15% of the settled sludge bed was expected to contain little or no W/Co fragments. The absence of W/Co in the top portion of the sludge bed results from the final layer that formed when the fine particulate settled after the pump shut off, and because W/Co in the twentieth (final) batch was mostly captured in the sludge layer created by the sludge from the nineteenth batch. The settling model also predicts the existence of a top layer, formed by longer-term settling, that contains no metal.

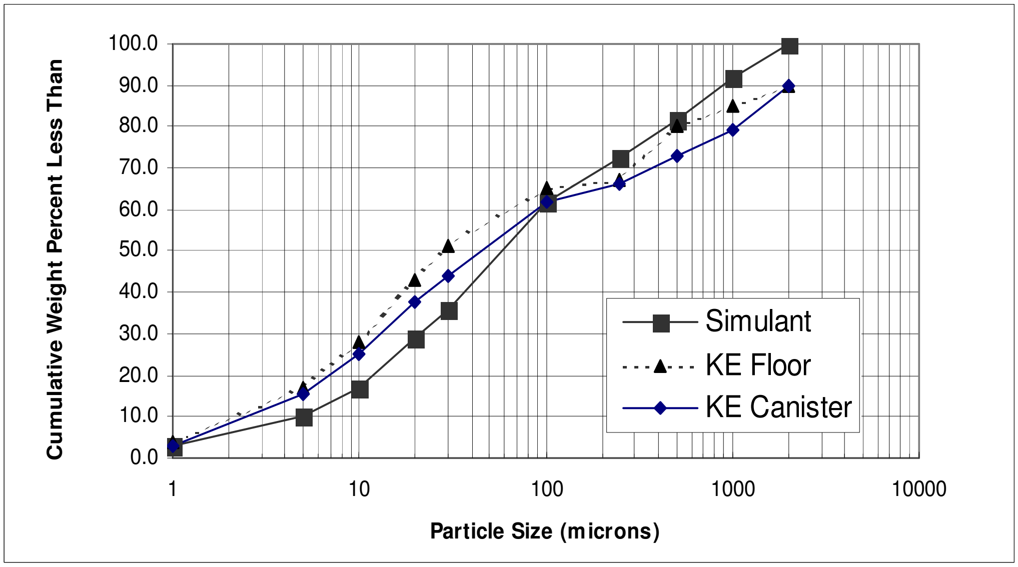


Figure 1. Comparison of the Particle Size Distributions of the Settling Simulant to Average of KE Floor and Canister Sludge Samples.

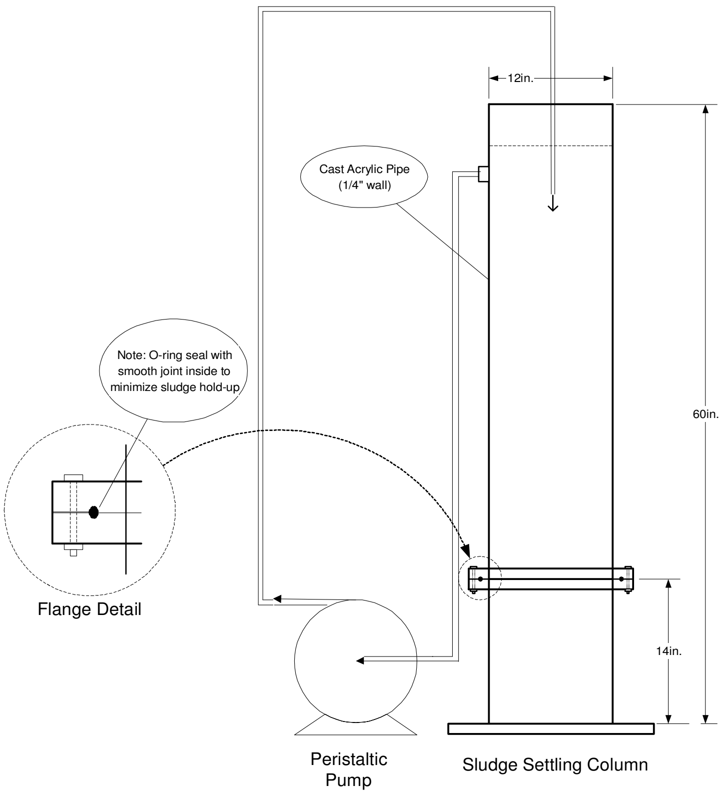


Figure 2. Diagram of Sludge Settling Equipment.

Figure 3. Settled Sludge Simulant, with 20 Distinct Layers Before Being Transported.

During the careful movement of the column to the transport vehicle and the subsequent transport to the NDE facility, accelerations/vibrations caused the settled sludge to liquefy, and the visually well-defined layers partially collapsed (Figure 4). Results from X-ray analysis of the settling column (after transportation/liquefaction) showed depletion of W/Co in the top ~35% of the column vs. the expected 10% to 15% depletion. In the bottom 65% of the column, uniform distribution of W/Co was observed. No evidence of W/Co segregation to the very bottom of the column was found.

After NDE was completed on the column, the three 2.5-in.-diameter sample cylinders were removed. Twenty reasonably well-defined layers of sludge were observed in each sample cylinder (i.e., no evidence of liquefaction, but some compaction).

X-rays showed the vertical distribution of the W/Co fragments in the 2.5-in.-diameter sample cylinders was very uniform, and no evidence of gross W/Co segregation was found. The concentration of W/Co fragments in the upper 5% to 15% of the sample cylinders appeared to be reduced. From these results, it can be inferred that before transportation (and liquefaction) the distribution of W/Co in the settling column was essentially uniform in the lower 85% to 90% of the column.

As a result of sludge reconsolidation after liquefaction during transport, the settled sludge depth in the settling column decreased about 15%, from ~11.5 in. to 9.9 in. Before transport, only a thin layer of water remained above the settled sludge. After transport, 1.3 in. of clarified supernatant was observed above the sludge. No changes were observed in the sludge depth in the 2.5-in.-diameter sample cylinders.

Figure 4. Settled Sludge After Transport. Only the lower four layers remain distinct; other layers have rotated or collapsed.

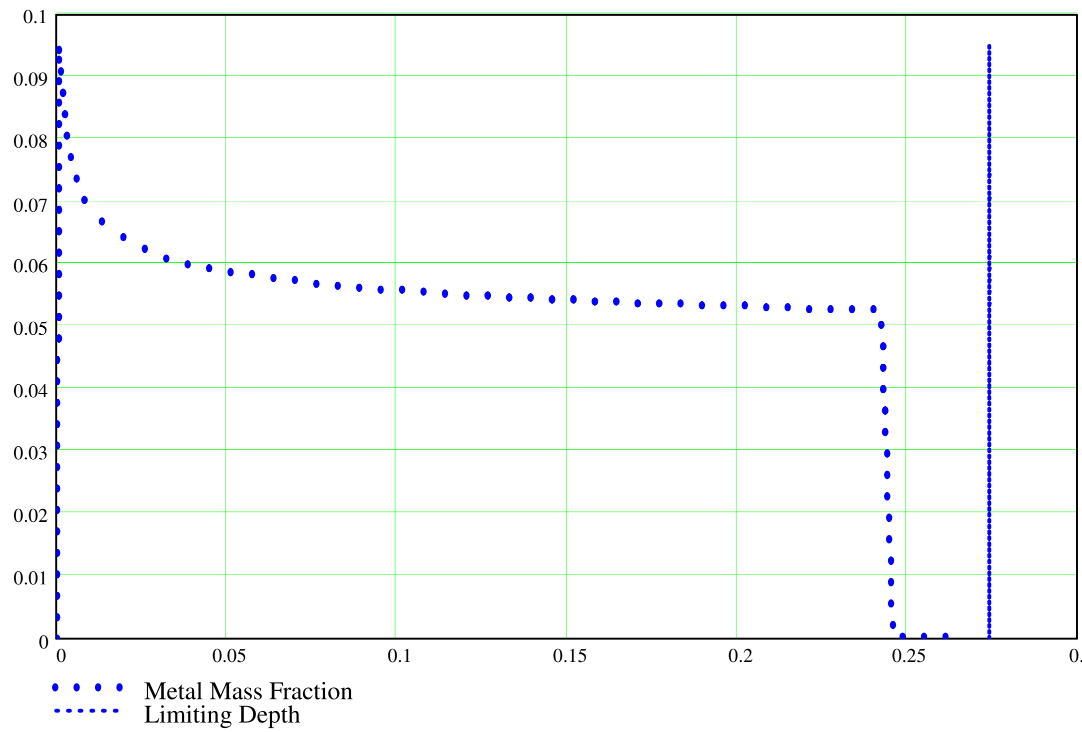
After the 2.5-in.-diameter sample cylinders were removed, X-rays were again taken of the settling column. Even though the settled sludge was significantly disturbed by withdrawing the cores, the distribution of the W/Co was largely unaffected (i.e., no significant segregation as the sludge and water moved to fill in the voids left behind when the cylinders were pulled).

Settling Model

As mentioned above, a settling model was created to simulate the LDC sludge filling operation [6] to quantify the extent to which metal-rich zones might form because of their possible impact on reaction heat release and gas generation. Key features of the settling model are (a) Stokes law determines the settling velocity with no particle aggomeration, (b) Settled particles pack haphazardly to a specified volume fraction, and (c) Separate particle size distributions and densities are used to characterize U metal, U oxide, and non-U oxide materials. The settling model consists of a set of differential equations for a mass balance of each material in each particle size bin, and an equation for the rate of change of the settled material interface. This interface velocity appears in each mass balance equation, so all the equations are coupled.

An example calculation with the settling model for addition of a 0.25 m^3 batch of settled sludge over one shift appears in Figure 5. Here metal mass fraction is plotted as a function of depth within the settled sludge. A peak value appears for low elevation, followed by a period somewhat steady but declining metal content, and ending with a region of zero metal content; the vertical line indicates maximum layer height. Here a batch sludge with 5% metal mass fraction originally has segregated into two sub-layers, the lower enriched in uranium metal, and the upper

Figure 5. Metal weight fraction vs elevation (m) for settling of 0.5 m^3 sludge batch added over 3.6 hours.



lacking in uranium metal. At the end of the simulation, some non-metal particles (chiefly uranium oxide due to the particle size distribution) remain suspended and require more time to settle. Thus, when multiple batches are added in series, each batch settles to a layer that is segregated into metal-enriched and metal-free sublayers, and a small portion of fine oxide remains suspended by the time the next batch is added. After the last batch is added, the remaining suspended material forms an metal-free layer thicker than previous metal-free layers.

The settling model proposed was clearly confirmed by the settling test. Because of the methods used and consistent behavior observed, no further settling model confirmation testing is proposed.

Shear Strength of K Basin Sludge

Shear strength is defined as the maximum stress force that can be applied to a material before it deforms. Materials that exhibit shear strength are typically solid/liquid multiphase systems and display solid-like behavior at low stresses and fluid-like behavior at high stresses. During the solid-like behavior, the material behaves elastically, where it will strain to a point at a given stress. When the stress is removed, the material will return to its initial state. The shear strength is regarded as the transition between elastic behavior and viscous flow. Examples of materials that display shear strength include cements, soils, paints, pastes, and various food products [7], as well as the sludge material in the Hanford K Basins.

The shear strength of the K Basin sludge is an important factor for predicting the mobilization behavior during retrieval. Additionally, the shear strength will affect how uranium metal fragments are distributed while the storage container is being filled. If sludge deposited in a container exhibits little or no cohesion or strength, incoming uranium metal fuel particles will pass through and accumulate on the bottom of the vessel.

Shear strength is the key physical sludge property that affects the fate of hydrogen gas bubbles generated during the corrosion of metallic uranium. These gas bubbles can escape the sludge matrix, accumulate in pockets, or be retained in the sludge as discrete bubbles. Terrones and Gauglitz [8] evaluated the potential formation of vessel spanning bubbles in K Basin sludge stored in large-diameter containers. Gauglitz and Terrones [9] examined gas retention within containerized K Basin sludge under the conditions of uniform gas generation.

Results from shear strength measurements conducted on K Basin sludge samples from 1995 - 2002 are provided in Tables 1 and 2. Most of the shear strength values were determined using a shear vane and a Haake M5 measuring head. The vane was rotated at a constant angular velocity (0.3 rpm or 0.6 rpm), and the force required to maintain this angular velocity was measured as a function of time. Material that exhibits shear strength shows a maximum torque value in a relatively small degree of rotation. Shear strengths were calculated from the measured maximum torque values. Before being used for measuring K Basin sludge samples, the Haake M5 measuring head was checked with standards. More extensive system performance checks were made to validate the results provided in Table 2 [10]. The test container and vane were set up to maintain the recommended clearance between the vane and container wall (e.g., space between the vane and walls was significantly greater than ¼ in., the maximum potential particle size possible in the sludge sample material).

Table 1 provides the earlier shear strength measurements, all of which were made on individual segregated layers (formed during settling tests) or size-fractionated subsamples of whole K Basin sludge samples [1, 11, 12, 13]. Most of the values on Table 1 are below 500 Pa. However, size-fractionated subsamples, KC-4 P-250 and KC-5 P-250 (both KE floor sludge), containing only particles greater than 250 µm, gave shear strengths values of 2700 Pa and 2800 Pa, respectively [13]. In comparison, the counterparts of these two samples, KC-4 M250 and KC-5 M250 containing only particles less than 250 µm, exhibited shear strengths of 300 Pa and 270 Pa, respectively.

Table 2 presents the results from recent (2002) measurements made on six “whole” KE Basin sludge samples [10], collected from the North Loadout Pit, the Weasel Pit, the basin floor (several locations), and the canisters. Another sample, “SNF + Sludge” (SNF Comp), a mixture of fuel fragments and KE Basin floor and canister sludge, was also measured. The sludge samples used for the measurements in Table 2 were collected in 1999 [14]. In comparison, the values in Table 1 were obtained from measurements performed the same year the samples were collected.

Table 1. Historical K Basin Sludge Shear Strength Data (1995 to 1999).

Sample ID	Sample Description	Shear Strength, Pa	Settled Density, g/cm3
1995 KE Floor and Pits Sludge, Makenas et al. [1] - measured using Bolin controlled stress rheometer.			
KES-M-13 Top	Top strata of the KE Floor Sludge, described as “liquid like”	2.2	1.11
KES-T-20 Top	Top strata of the KE Weasel Pit Sludge, described as “liquid like”	0.9	1.6 (estimate)
1996 KE Canister Sludge, Makenas et al. [11] - measured after 2 weeks of settling using a Haake M5 with 8-mm-diam. shear vane.			
96-04 U/L	Sample 96-04 settled into 2 layers, with 70% (vol) in the upper layer and 30% (vol) in lower layer. 96-04 U/L was collected at the interface between the upper and lower layer. 96-04 was collected from a canister containing very corroded fuel.	<100	1.09
96-06 U/M	Sample 96-06 settled into 3 distinct layers, with 5% (vol) in the upper layer, 53% (vol) in middle layer, and 42% (vol) in the lower layer. Sample 96-06 U/M was collected at the interface of the upper and middle layer. 96-06 was collected from a canister containing significantly corroded fuel.	200 ± 30	~1.7
96-06 M	Middle layer of sample 96-06. 83 wt% U (dry basis)	150 ± 20	1.92
96-06 M/L	Collected from the interface of the middle and lower layer of sample 96-06.	460 ± 40	~2.5
96-06 L	Lower layer of sample 96-06. 84 wt% U (dry basis)	470	2.99
96-11 U/L	Sample 96-11 settled into 2 distinct layers, with 7% (vol) in the upper layer and 93% (vol) in the lower layer. 96-04 U/L was collected at the interface between the upper and lower layer. 96-11 was collected from an unfueled canister.	130	~1.1
1996 KW Canister Sludge, Makenas et al. [12] - measured after 3 days of settling using Bohlin controlled stress rheometer.			
96-21 Rec	Size fractionated subsample of 96-21 (97 vol% of original sample) containing only particles less than 710 µm	30 to 40	3.30
96-24 Rec	Size fractionated subsample of 96-24 (84 vol% of original sample) containing only particles less than 710 µm	20 to 30	2.64
1999 Consolidated Sludge Samples, Bredt et al. [13] - measured after 2 weeks of settling using Haake M5 with 8-mm-diam. shear vane.			
KC-2/3 M250	Size fractionated canister sludge composite (sludge from 11 canister barrels) containing only particles less than 250 µm.	280 ± 110	2.13
KC-4 P250	Size fractionated floor sludge (collected on floor between slotted barrels) containing only particles greater than 250 µm.	2800 ± 800	1.3
KC-4 M250	Size fractionated floor sludge (collected on floor between slotted barrels) containing only particles less than 250 µm.	300 ± 10	1.2
KC-5 P250	Size fractionated floor sludge (collected on floor away from corroded fuel) containing only particles greater than 250 µm.	2700 ± 400	1.5
KC-5 M250	Size fractionated floor sludge (collected on floor away from corroded fuel) containing only particles less than 250 µm.	270 ± 20	1.2

Table 2. K Basin Sludge Shear Strength vs. Settling Time (2002 Data [10]).

Sample	Sample Description	Settled Density,[a] g/cm³ current data (previous data)	Days of Settling[b]	Average Shear Strength, Pa	Standard Deviation[c] Pa	Number of Measurements
KE Basin Pit and Floor Sludge Samples						
FE-3 (whole)	**North Loadout Pit**. Prepared from 6 core samples collected in Jun. 1999. About 80 wt% (dry) is made up of particles between 250 µm and 1410 µm. U content: ~1.7 wt% (dry)	1.24 (1.23 – 1.56)	2	760	340	3
			5	920	10[c]	2
			20+	240	61	5
FE-5 (whole)	**Weasel Pit**. Prepared from 8 core samples collected in Jan. and Apr. 1999. About 70 wt% (dry) is made up of particles less than 250 µm. U content: ~5 wt% (dry).	No new data (1.43 – 1.5)	2	1100	NA	1
			5	1800	640	4
			20+	4000	750	5
KC-4 (whole)	**Floor, between slotted canisters**. Prepared from 3 sample locations; collected in Mar. 1999. About 85 wt% (dry) is made up of particles less than 250 µm. U content: ~17 wt% (dry).	1.26 (1.24)	2	480	NA	1
			5	330	80[c]	2
			20+	400	160	4
KC-5 (whole)	**Floor, away from canisters**. Prepared from 3 sample locations; collected in Mar. 1999. Only about 36 wt% (dry) is made up of particles less than 250 µm. U content: ~6 wt% (dry). Sample was found moist in Oct 2001, but not saturated.	1.28 (1.19)	2	1100	NA	1
			5	1600	100[c]	2
			20+	1000	150	3
KE Canister Sludge Samples						
KC-2/3 (whole)	**Canister Sludge Composite**. Prepared from 10 sample locations; collected in Mar. and Apr. 1999. About 72 wt% (dry) is made up of particles less than 250 µm. U content: ~ 60 wt% (dry). A portion of the sludge used was found moist in Oct 2001, but not saturated.	1.3 (2.1)	2	5700	1700	3
			5	4600	560	3
			20+	8200	4000	3
KE Basin Floor + Canister Sludge + Fuel Fragments						
SNF + Sludge (SNF Comp)	By volume, ~64% floor sludge, ~26% canister sludge, ~7% fuel corrosion product (U_4O_9), and ~3% fuel fragments. About 20% to 30% of the material added to composite was dry sludge and fuel fragments.	2.38 (not previously measured)	2	740	NA	1
			5	No valid measurements obtained		
			20+	1900	NA	1

(a) Measurements of the settled sludge density were performed about 1 month before shear strength measurements were made. Measurements identified as previously reported were generally made within several months of when sludge samples were collected from the KE Basin. A large difference between the current measurements and the previously reported measurements may indicate the samples had undergone significant changes during sample storage.
(b) Measurements obtained at settling time identified as "20+ days" were performed just after moving settled sludge samples through several hot cells. The settled sludge may have been disturbed during the sample transport.
(c) For measurements with only two observations, the value given is the range between the average and the measurements.

In Table 2 the results of the shear strength measurements are given for the sludge samples at several settling times. Measurements were performed on "as received sludge"; sludge that was disturbed and allowed 48 hours to re-settle/reconsolidate; and sludge that was disturbed and allowed 5 days to reconsolidate. The "as received sludge" had not been disturbed for more than 20 days; however, several days before the measurements were made, the samples were moved, using relatively large manipulators, through several cell areas to the cell containing the yield strength test apparatus. Moving the samples may have been partially disturbed the sludge.

The results in Table 2 show the shear strength increased significantly with settling time for the Weasel Pit (FE-5), canister (KC-2/3) and the SNF Comp sludge samples. For the North Loadout Pit (FE-3) and floor sludges (KC-4 and KC-5), the shear strength was either constant or varied randomly with settling time.

The shear strength values in Table 2 (240 to 8200 Pa) are generally higher than most values obtained during the previous K Basin sludge characterization efforts (Table 1). Two factors may have contributed to the higher values in Table 2: sample history (handling and aging) and the presence of large-diameter particles (i.e., particles greater than 250 μm). During the time the samples were stored, metallic uranium and other compounds likely experienced some oxidation and hydration, resulting in physical changes to the sludge. The samples have also been subjected to considerable handling during characterization and process testing.

While handling and aging likely affected the shear strength of the sludge samples measured in 2002, the actual K Basin sludge will also be subjected to handling during retrieval (i.e., it will be pumped through several hundred feet of hose at velocities around 10 ft/sec), and will be stored in T Plant for a number of years. It should be noted that the sludge samples tested in 2002 (though previously handled and stored) represent the best sludge sample material remaining in at the hot cells from characterization activities over the past 4 years. Obtaining new representative samples from the K Basins was not an option because of the significant resources and time required.

All samples analyzed for shear strength in Table 2 were "whole" samples, and included some fraction of particles greater than 250 μm. While most measurements in Table 2 are higher, the results from KC-4 "whole" (15 wt%, dry basis, particles greater than 250 μm) in Table 2 (330 Pa to 480 Pa) are comparable to the value of 300 Pa obtained earlier for the KC-4 M250 sample (Table 2), which contained no large particles.

While most of the shear strength data obtained in earlier characterization activities was significantly lower than those measured in 2002 testing, a technical justification for discounting either set of data was not found. The technical reasons for the differences were substantiated based on functionality checks on the equipment and independent verification of the calculations. Calculations and analyses requiring K Basin shear strength data should include both the earlier determined values (Table 1) and the 2002 values given in Table 2. The result of combining the data sets is a wide range of yield strength values for the sludge: 1 to 8200 Pa. Consequently, the analyst performing calculations will have to consider either the total range or the source and handling of a specific sludge stream, making a case for using a subset of the observed values based on technical merits or design considerations.

Thermal Conductivity Measurements

The thermal conductivity for a material is defined as the ratio of steady-state heat flow by conduction across a surface (heat transfer per unit area per unit time) to the temperature gradient at the surface. The thermal conductivity of the K Basin sludge must be known in order to accurately predict the rate of heat removal and the temperature of the sludge within a storage container.

Twelve samples of KE Basin sludge, collected from the North Loadout Pit, the Weasel Pit, the basin floor (several locations), and the canisters, were used for the thermal conductivity measurements [10]. One sample, SNF + Sludge Composite (SNF Comp), was a mixture of fuel fragments and KE Basin floor and canister sludge. The settled density and weight fraction solids of the sludge samples were determined in parallel with the thermal conductivity measurements to allow better data interpretation and modeling.

The instrument used for the measurements was a Hukseflux TPSYS02 system with a needle probe, using a non-steady-state method in accordance with ASTM D 5334-92. Before the actual sludge samples were measured, the equipment and method were tested extensively at room temperatures using standards and simulants. This initial testing demonstrated the reproducibility and accuracy of the Hukseflux system for the type of samples analyzed (saturated sludges).

Thermal conductivity of each sample was measured at least three times, and the average value and the standard deviation reported (see Table 3). The measurements were performed at ambient hot cell temperature, which ranged from 29°C to 41°C. The percent standard deviations from multiple measurements ranged from 1.5% to about 6%, confirming good reproducibility. For most of the samples, essentially the same thermal conductivity value was measured. Excluding the sludge samples from the North Loadout Pit (1.03 W/mK), the Weasel Pit (0.86 W/mK), KC-5 P250 (0.80 W/mK – large-particle floor sludge, away from canisters), and KC-2/3 M250 (0.81 W/mK – small-particle canister sludge), the average thermal conductivity of the remaining eight sludge samples was 0.70 (standard deviation of 0.014) W/mK. In general, no direct trends were evident on the relationship between thermal conductivity and chemical composition, particle size, settled density, or mass fraction water. The relationship is more likely a complex combination of these properties along with oxidation states and crystalline structures. However, the volume fraction water (void fraction) for the all of the K Basin sludge samples was similar; 0.77 on average, with a standard deviation of 12%.

The thermal conductivities of the sludge samples were only a little higher than that of water (~0.60 W/mK). [As another comparison, the range of thermal conductivities of the sludges fell into the lower region of that expected for saturated sands and soils.] The high water content and the abundance of hydrated species in the sludges likely contributed to the relatively low values. The similar and relatively high volume fraction water in the K Basin sludge samples appears to be a dominating factor affecting the thermal conductivity. For nominal design modeling and calculations, a thermal conductivity on the low end of the measured range is desired. Therefore, a thermal conductivity of 0.70 W/mK is judged to provide both a reasonable and a defensible value for all KE Basin sludge types.

Table 3. Summary of Thermal Conductivity, Settled Solids Density, Wt% Water, and Void Fraction Results

Sludge Sample	Sample Description	Thermal Conductivity, W/mK (standard deviation)	Settled Sludge Density, g/cm^3	Wt% Water	Void Fraction (Volume Fraction Water)
	KE Basin Floor Sludge Samples				
FE-3 (whole)	North Loadout Pit sludge	1.03 (±0.05)	1.35	54	0.76
FE-5 (whole)	Weasel Pit sludge	0.86 (±0.04)	1.47	44	0.65
KC-4 (whole)	Floor sludge from between canisters	0.72 (±0.04)	1.26	64	0.81
KC-4 M250	Floor sludge from between canisters (minus 250-µm fraction)	0.69 (±0.04)	1.53	68	0.82
KC-4 P250	Floor sludge from between canisters (plus 250-µm fraction) – Reconstituted[a]	0.70 (±0.03)	1.49	54	0.81
KC-5 (whole)	Floor sludge from main basin	0.72 (±0.03)	1.28	66	0.84
KC-5 M250	Floor sludge from main basin (minus 250-µm fraction)	0.69 (±0.01)	1.29	73	0.94
KC-5 P250	Floor sludge from main basin (plus 250-µm fraction) – Reconstituted[a]	0.80 (±0.02)	1.43	48	0.69
	KE Basin Canister Sludge Samples				
KC-2/3 (whole)	Canister sludge composite	0.70 (±0.04)	1.30	54	0.70
KC-2/3 M250	Canister sludge (minus 250-µm fraction)	0.81 (±0.06)	2.15	44	0.88
96-13	Canister sludge collected in 1996 – Reconstituted[a]	0.68 (±0.04)	2.67	26	0.64
	KE Basin Floor + Canister Sludge + Fuel Fragments				
SNF + Sludge Composite	By volume, ~64% floor sludge, ~26% canister sludge, ~7% fuel corrosion product (e.g., U_4O_9), and ~3% uranium fuel fragments	0.70 (±0.03)	2.32	31	0.72

(a) Reconstituted – sludge sample dried out and was reconstituted to a settled sludge state through the addition of K Basin water and aggressive mixing.

Vessel-Spanning Bubble Formation in K-Basin Sludge Stored in LDCs Containers

The Hanford K-Basin sludge is to be retrieved and stored in the large-diameter containers (LDCs). This waste contains some fraction of uranium metal that generates hydrogen gas, which introduces potential upset conditions. One postulated upset condition is a rising plug of sludge supported by a hydrogen bubble that is driven into the vent filters at the top of the container. In laboratory testing with actual K-Basin sludge, vessel-spanning bubbles that lifted plugs of sludge were observed in 3-inch-diameter graduated cylinders.

Analytical assessments were performed to address the potential for the generation of a vessel-spanning bubble in the LDCs [8]. The assessments included the development and evaluation of static and dynamic bubble formation models over the projected range of K-Basin sludge mechanical properties. Additionally, the theory of circular plates was extrapolated to examine conditions under which a plug of sludge would collapse and release a spanning bubble.

Based on the conservative models developed in this report and on the latest mechanical property data of K-Basin sludge, the formation of vessel-spanning bubbles within LDCs is credible. This is due largely to the relatively high yield stress of K-Basin sludge, which could be as high as a few thousand Pascals. Vessel-spanning bubbles can form via two main mechanisms, from a single point source or from a uniformly distributed region of hydrogen-generating particles (uranium metal oxidation reaction). Effectively, a vessel-spanning bubble is a growing region occupied by gas that separates a lower hydrogen-generating sludge layer containing uranium metal from an overlying, mostly inert sludge layer.

Sludge plugs formed by spanning bubbles at yield stress measured for K-Basin sludges are predicted to be stable. Analysis based on thin circular plate theory showed that, if a sludge plug formed as a result of a vessel-spanning bubble, sludge batches with low yield stress are expected to collapse even for relatively thick plugs, thus preventing the sudden ejection of material from the container. However, as the yield stress increases beyond 100 Pa, sludge collapse is expected to occur only for thin plugs.

Based on the vessel-spanning-bubble analysis, it was concluded that the rheological behavior of K-Basin sludges, particularly the yield stress, is the dominant factor in determining bubble size. Assuming that the bubble is formed from a single-point source of gas generation and the rheological behavior of the sludge is described by the Herschel-Bulkley model for pseudoplastic materials, and using the conservative yield constant value of 0.061, we predict that vessel-spanning bubbles will not form in 5-ft-diameter vessels provided the yield stress is less than 900 Pa. For instance, the largest bubble size predicted for a sludge with a yield stress of 500 Pa is 3.2 ft. These conclusions do not exclude the formation of gas layers produced by a distributed gas-generating region within the sludge. If a gas-generating region spans the diameter of the vessel, a layer of gas will readily form and grow.

Sludge Plug Breakup and Source Term

As described above, a vessel-spanning bubble within the sludge cannot be definitively ruled out due to sludge rheological properties, and growth of such a bubble would lead to upward motion of the overlying sludge plug. It is therefore of interest to understand whether or not such a sludge plug could be broken up by vessel upper internals, so that sludge would not have the potential to leak from the vessel through an upper port. Also, in the event that sludge could begin to exit the vessel, the nature of the source term is of interest.

Experiments were performed to address sludge plug breakup and its behavior upon vessel exit [15]. A schematic diagram of the apparatus is shown in Figure 6. The main component of the apparatus is a Plexiglas™* column of 13.9 cm ID and 42.6 cm length. Actually two sections comprise the column: a lower 12.6-cm long section and an upper 30-cm long section. During an experiment the two sections are connected by a flange. The upper section contains the simulant sludge plug and the lower section receives air from a high pressure house line that is directed to a throttle valve just upstream of the test section. The steady and gradual introduction of air into the lower section results in an expanding, pressurized column of air and a steady upward displacement of the sludge plug. Mixtures of water and kaolin were used to represent the sludge plug in all the tests. The yield stress and density of this simulant as functions of concentration are known.

Five experiments (Tests 1 to 5) were performed to study sludge plug failure when the sludge is forced (extruded) through simple orifice geometry. These experiments also reveal the manner in which sludge is ejected from the storage container in the unlikely event that an actual sludge plug is displaced by the reaction product gas to the top of the storage container. For these purposes a 1.1-cm thick Plexiglas™* lid was attached to the top of the upper section. A centrally located 2.54-cm hole was drilled through the lid to represent the container vent. The thickness of the sludge plug varied from 5.1 cm to 13.0 cm. The simulant plug was slowly forced upward by opening the air valve and establishing a gas pressure sufficient to overcome the weight of the sludge plug. Once the upper surface of the plug made contact with the lid the gas pressure was increased to a predetermined value.

The following observations were made with the experimental system described in the foregoing. In all the tests the clay emerged from the vent in the form of a stable, long cylindrical extrusion. The diameter of the cylindrical extrusion was equal to the diameter of the vent (2.54 cm). A loud pop signaled the sudden end of the extrusion process. It was obvious that gas penetrated the vertical thickness of the clay plug which resulted in the rapid depressurization of the driver gas column. A significant fraction of the clay plug was left behind in the Plexiglas™* column pressed up against the inside surface of the column's lid. A top and side view of the clay morphology observed at the end of Test 3 is shown in the photographs in Fig. 7.

Interestingly enough, inspection of the residual clay plug revealed a centrally located, narrow vertical column or crack extending from the bottom surface of the plug to the vent. In all the tests the diameter of this crack was smaller than the 2.54-cm diameter of the vent. Moreover, during

* ™: Rohm & Haas Corp.

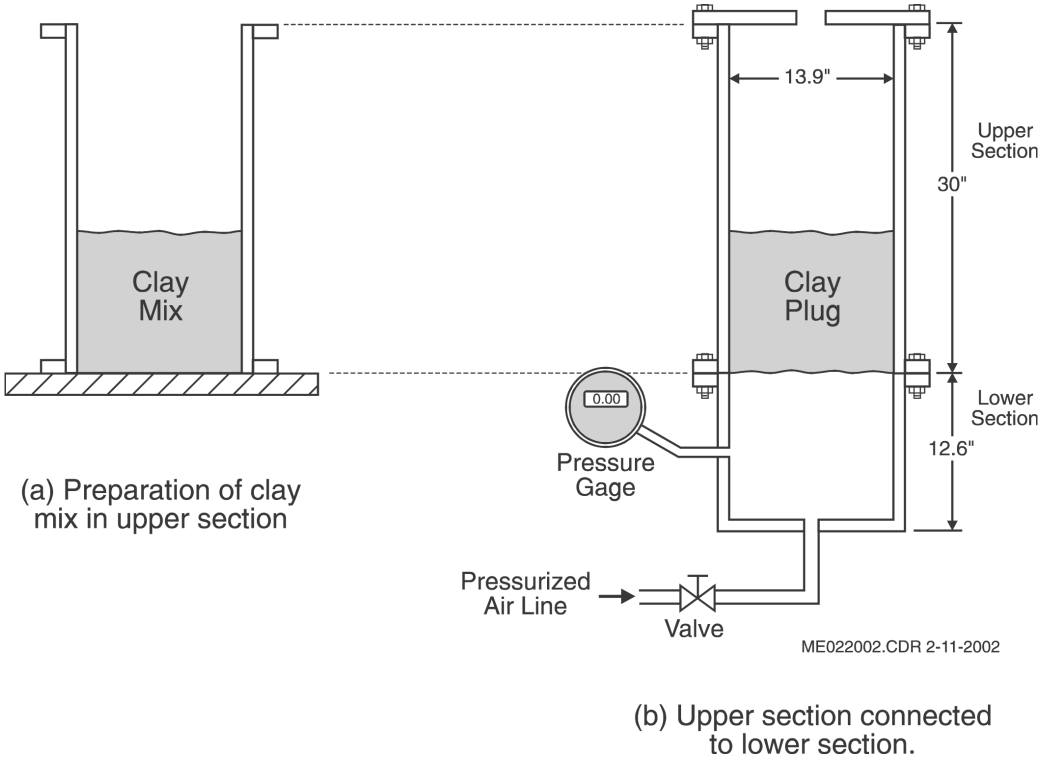


Figure 6. Schematic diagram of experimental apparatus (all dimensions in cm).

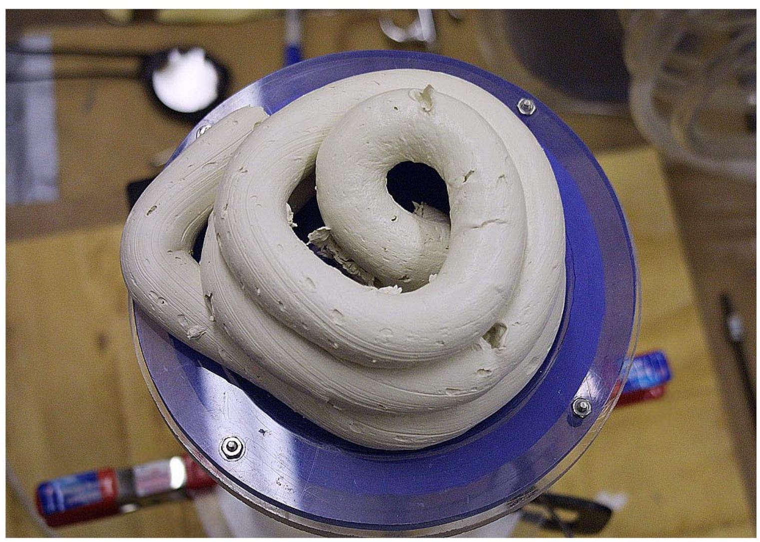


Figure 7. Extruded clay and residual clay left in test column after gas pull through.

the course of a test the central region of the bottom surface of the clay plug appeared to be crack-free to the visual observer. Apparently this vent crack grows very rapidly from the bottom surface of the clay to the vent and terminates the vent flow of clay via gas pull-through. This behavior appears to be similar to the gas column that forms when an inviscid Newtonian liquid is draining from a container. When the liquid (Newtonian) depth falls below a critical value the overlying gas is pulled through the liquid layer to the vent. The gas column may appear because of the presence of a vortex, which occurs, for example, when a bath tub is emptied. However, gas pull-through is also known to suddenly occur during vortex-free draining.

An analysis of gas pull-through within a Bingham plastic plug flowing upward to a vent was developed to predict the observed behavior. Model derivation and an explanation of the final model are too detailed for a proper explanation here, and interested readers are referred to [15]. The model relates a critical sludge plug height at the time of gas breakthrough to the sludge yield stress, sludge density, pressure difference across the plug, and the characteristic size of the opening. When a prediction of breakup internal to the vessel is desired, the opening size is defined by the reduced upward flow area due to obstacles, and the model predicts the maximum sludge plug height that can break up. When sludge release from the vessel is estimated, the difference between the actual plug height and the critical height determines the amount of material that will be extruded.

Results of the experiments are given in Table 4. Parameters given in the table are sludge density, yield stress, pressure difference at failure, ejected mass, actual plug height, and critical plug height based on the measured ejected mass. The predicted critical plug height is in good agreement with observation and details are contained in the reference.

Table 4. Clay Flow Contraction Experiment Summary.

Test	**Geometry**	ρ_f **(kg m^{-3})**	τ_0 **(Pa)**	ΔP **(Pa)**	m_{ej} **(kg)**	H_0 **(m)**	H_{cr} **(m)**
1	Orifice (d = 2.54 cm)	1410	1000	2.1×10^4	0.04	0.051	≥ 0.051
2	Orifice (d = 2.54 cm)	1410	1000	2.1×10^4	0.68	0.13	0.098
3	Orifice (d = 2.54 cm)	1410	1000	3.1×10^4	1.04	0.13	0.081
4	Orifice (d = 2.54 cm)	1370	350	1.4×10^4	0.38	0.08	0.061
5	Orifice (d = 2.54 cm)	1370	350	1.4×10^4	0.39	0.079	0.060
6	Grating (b = 2.5 cm)	1410	1000	1.7×10^4	0.97	0.13	0.085

Conclusions for Plug Breakup and Source Term

Application of the model to the LDC indicated that a sludge layer nearly free of metal would need to be about 0.29 m thick in order not to be broken up by LDC upper internals. This is not realistically expected to be possible because the pattern of sludge loading would not allow for such extreme stratification. As shown in Figure 5, while metal does concentrate near the bottom of a given batch, the metal-free zones are relatively narrow.

It is also notable that experimental data show that sludge cannot be violently ejected from a vessel, but it is instead benignly extruded. Thus, there is no source term associated with the extrusion phase of sludge motion. However, it is possible that some sludge aerosol might be formed during the period of gas blowthrough. While no particulate was observed during the tests above, demonstrating that gas blowthrough does not cause significant disruption of the sludge, any aerosol particles of interest would have been too small to see with the naked eye. A model created for sludge aerosolization during gas blowthrough yields an estimated respirable fraction less than 10^{-5}, but actual measurements are needed to validate the model.

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