

## Operational Challenges in Mixing and Transfer of High Yield Stress Sludge Waste - 10263

P. N. Bhatt, T. B. Caldwell  
Savannah River Remediation, Aiken, South Carolina 29808

### ABSTRACT

The ability to mobilize and transport non-Newtonian waste is essential to advance the closure of highly radioactive storage tanks. Recent waste removal operations from Tank 12H at the Savannah River Site (SRS) encountered sludge mixtures with a yield stress too high to pump. The waste removal equipment for Tank 12H was designed to mobilize and transport a diluted slurry mixture through an underground 550m long (1800 ft) 0.075m diameter (3 inch) pipeline. The transfer pump was positioned in a well casing submerged in the sludge slurry. The design allowed for mobilized sludge to enter the pump suction while keeping out larger tank debris. Data from a similar tank with known rheological properties were used to size the equipment. However, after installation and startup, field data from Tank 12H confirmed the yield stress of the slurry to exceed 40 Pa, whereas the system is designed for 10 Pa. A revision to the removal strategy was required, which involved metered dilution, blending, and mixing to ensure effective and safe transfer performance. The strategy resulted in the removal of over 255,000 kgs of insoluble solids with four discrete transfer evolutions for a total transfer volume of 2400 m<sup>3</sup> (634,000 gallons) of sludge slurry.

### INTRODUCTION

The Savannah River Site (SRS) is an 800 square kilometer industrial facility (300 square miles) constructed in the early 1950s. SRS is located in western South Carolina and covers portions of Aiken, Barnwell, and Allendale counties. The site produces weapons-grade plutonium, uranium, and tritium as part of the U. S. Department of Energy's (DOE) national defense mission. Environmental remediation at the site includes cleaning and closing nuclear waste storage tanks.

Radioactive waste at SRS is generated from the chemical separations facilities and is present in the tank farms as insoluble solid compounds and water-soluble salts. Since the first waste receipt in Tank 1F in 1954, the site has generated over 530,000 m<sup>3</sup> (140 million gallons) of high-level nuclear waste. Evaporation operations reduced this volume to the present inventory of about 136,000 m<sup>3</sup> (36 million gallons). The waste is stored in 49 underground waste tanks in F and H area. Two of the original 51 tanks were operationally closed in 1997. While stored in the tanks, the insoluble solids settle and accumulate on the bottom of the tanks in the form of *sludge*. The liquid volume is reduced by evaporating excess water. The concentrated salts crystallize forming hard (but porous) *saltcake*. Tank farm facilities also pre-treat the accumulated sludge and salt solutions to facilitate further processing at other SRS treatment facilities (i.e., Defense Waste Processing Facility (DWPF) and Saltstone Disposal Facility (SDF)). These treatment facilities convert the sludge and supernate to more stable forms suitable for permanent disposal.

The waste storage tanks comprise of four design types. The Type I tanks are the oldest with a nominal capacity of 2840 m<sup>3</sup> (750,000 gallons) and a 1.5m high (5 feet) secondary steel

containment pan within a concrete vault. The Type I tanks are approximately 23m (75 feet) in diameter. The next class of storage is the Type II tanks with a storage capacity of 3900 m<sup>3</sup> (1,030,000 gallons) and 9m (85 feet) in diameter. The most modern tanks are the 4900 m<sup>3</sup> capacity Type III tanks. They boast a full secondary containment, an integral cooling and ventilation system, heat treated steel liners, and numerous access openings. Each Type I, II, and III tank has an intertwining array of 50mm diameter cooling pipes (2 inch diameter). These pipes interfere with waste removal and tank closure activities. The 4900 m<sup>3</sup> *un-cooled* tanks make up the fourth design (often referred to as the Type IV tanks). The un-cooled tank variety has a self-supporting dome roof and no internal cooling coils. The un-cooled tanks were used primarily to store low activity waste.

The ability to mobilize and transport non-Newtonian materials is necessary to bring about closure of these tanks. The sludge waste in particular is difficult to remove and requires several high-energy mixer pumps to suspend the settled sludge into a slurry mixture. Prior to Tank 12H, SRS successfully suspended and transported almost 8800 m<sup>3</sup> of sludge slurry (2,300,000 gallons) from seventeen different waste tanks over a span of 40 years.<sup>1, 2</sup> However, Tank 12H proved unique in that the sludge slurry had an unusually high yield stress, and the waste did not conform to the expected properties of a tank of that class. This required re-evaluation of the removal strategy, which involved dilution and blending to form a transportable mixture.

## TANK 12 HISTORY

Tank 12H is a Type I tank targeted for closure and is located in the H-Tank Farm. It was the last of the original twelve tanks built in the early 1950s and first saw service in October 1956. The tank is fabricated from welded 12.7mm thick carbon steel plates. The tank top is supported internally by twelve 0.6m diameter columns. The primary tank sits in a secondary confinement pan 24.4m in diameter (80 feet) and 1.5m high (5 ft) with a capacity of 86 m<sup>3</sup> (22,800 gallons). The pan is also fabricated from 12.7mm thick carbon steel plate and provides a collection volume for the tank in case of leakage. The tank system is then encased in a steel reinforced concrete vault.

Tank 12H received a mixture of H-Area Purex High Heat Waste (HHW) and Low Heat Waste (LHW) through September 1963. This waste was rich in iron compounds and is characteristic of Purex waste at other DOE sites. From October 1963 through May 1973, the tank received large amounts H-Area modified (HM) HHW. HM HHW is an aluminum-based waste form. A few small additions of Thorex\* waste was received in the early 1960s.<sup>3</sup> Thorium nitrate [Th(NO<sub>3</sub>)<sub>4</sub>] compounds from the Thorex process are neutralized with sodium hydroxide to form thorium hydroxide [Th(OH)<sub>4</sub>].<sup>4</sup> This is the form sent to Tank 12H. After the discovery of leak sites in 1974, excess supernatant liquid was transferred to a nearby tank and Tank 12H was removed from service. From 1974 through 2004, the tank remained dormant. Residual water slowly evaporated exposing the settled solids. Over time, the settled solids layer desiccated and shrank from 2.4m to 1.9m (95 inches to 75 inches).<sup>5</sup> In preparation of waste removal activities, water

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\* *Thorex* – Thorium extraction process. A process where Uranium-233 is recovered and purified from neutron-irradiated thorium reactor fuels through tributyl phosphate extraction chemistry.

was added in November 2004 to re-hydrate the sludge. At the start of waste removal operations in August 2008, the tank level was approximately 2.8m (110 inches).<sup>6</sup>

## WASTE REMOVAL EQUIPMENT

Previous waste removal operations involved using equipment that imparted liquid jet action to scour the settled solids layer. The jets would ablate and slurry the solids into a transportable suspension. The resultant slurry possesses non-Newtonian fluid behavior following the Bingham plastic flow model. Therefore, equipment is designed and installed to produce the jetting action and pump the slurry to a destination tank. Sufficient liquid is added to the tank to fully suspend sludge solids. Usually, existing supernatant liquid from other tanks is used as the suspension medium to minimize introduction of new liquids into the tank farm system.

Tank 12H bulk waste removal operations required using four standard slurry pumps positioned nearly equidistant from each other. The pump body is submerged in the waste. A standard slurry pump generates 275 m<sup>3</sup>/hr (1200 gpm) of flow by taking suction from the pump bottom and discharging horizontally out of two diametrically opposed nozzles. The exit velocity out of each nozzle is 33.2 m/sec (109 ft/sec). The distance from the nozzle discharge to where sludge can be mobilized is called the *effective cleaning radius* (ECR). The ECR depends largely on the tenacity of the sludge. The pump is driven by a top-side 150 HP motor connected with a long shaft. The shaft is supported by intermediate bearings inside a pump column. The pump column is pressurized with sealing water to prevent migration of contamination to the tank top. The pump bodies are rotated on a slewing bearing at 1/5 rpm in a manner that causes the jet streams to sweep transversely over the vessel floor, thereby mobilizing waste across the effective radius of the jet streams. The pump assembly is supported from the tank top. Refer to Figure 1.

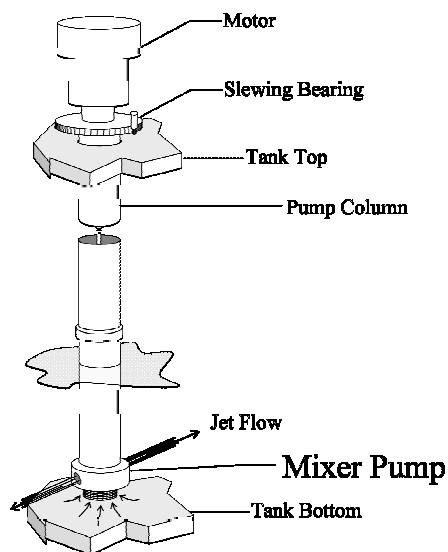


Fig. 1. The standard slurry pump at the Savannah River Site has been used in numerous waste removal campaigns

The slurry suspension is pumped out of Tank 12H to nearby Tank 51H with a 15 HP centrifugal submersible transfer pump (STP). The STP is inserted through one of the tank openings and is connected to an existing underground transfer line that ultimately goes to Tank 51H. The submergence of the STP can be adjusted in 0.3m increments (1 ft) to a position 3.65m above the tank floor (12 feet). The STP is placed in a well casing that extends from the tank floor to the tank ceiling. The well casing is designed to protect the STP from the vigorous mixing action caused by the standard slurry pumps. The well casing is perforated with 50mm diameter holes to permit the slurry suspension to enter the pump well.

## **PLANNED OPERATING SEQUENCE**

Early sludge removal campaigns involved inserting the slurry pumps at an elevation directly above the settled sludge layer. The pumps would agitate the contents for a few days, after which the slurry suspension would be pumped to a nearby tank. Fresh liquid would be added to the tank and the slurry pumps would be lowered to the new sludge elevation. Depending on the depth of the sludge, this evolution would require several iterations.

In 2000, with the Tank 8F sludge removal program, a new operating strategy was adopted. This involved positioning the slurry pumps above the sludge layer as in past operations, but instead of transferring the suspended slurry after each mixing cycle, the pumps would be lowered until the entire tank contents were suspended at which point a single slurry transfer would be made. This was used with success for Tanks 5F, 6F, 7F, and 11F. The new strategy reduced the number of operational evolutions, thereby reducing radiological risk. Tank 12H was the next tank in the queue for cleaning and remediation.

The sludge from Tank 12H would be used as part of Sludge Batch 6 as feed to the Defense Waste Processing Facility (DWPF). Because of the anticipated high aluminum content in the Tank 12H sludge, the batch would be treated later with high concentrations of sodium hydroxide to dissolve some of the inert aluminum compounds<sup>†</sup> as part of the Extended Sludge Processing (ESP) program. Only a portion of the Tank 12H sludge is needed for Sludge Batch 6; the rest would be used as part of later sludge batches. The original plan called for transferring approximately 50% of the Tank 12H sludge inventory, or 180,000 kg. As waste is suspended, the slurry pumps would be lowered in increments. Samples would be obtained to determine the weight percent of insoluble solids, and projections would then be made to determine the mass of solids needed for transfer. Following each transfer, sludge soundings and mapping would be used to confirm the volume of the sludge remaining.

## **ASSUMED SLUDGE RHEOLOGY**

Most of the SRS sludge is the result of the re-precipitation of inorganic compounds during the neutralization reactions. As such, the incipient particles are small (less than 1  $\mu\text{m}$ ), and when settled, form a gelatinous layer. However, age, heat (from radioactive decay) and a chemically

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<sup>†</sup> This process is called *Aluminum Dissolution*, which dissolves some solid aluminum compounds in the sludge (such as Boehmite and Gibbsite). This reduces the amount of solids that must be vitrified in borosilicate glass at DWPF, and allows these forms to be disposed of in the more economical grout form at the Saltstone Production Facility.

active matrix transform some of the compounds into larger particles and more complex waste forms. Because of this, laboratory surrogates are unsuitable to predict rheological properties of waste sludge.

During the development of the sludge processing facilities at SRS in the 1980s, the site embarked on a sludge characterization program. Large sludge samples were taken from several tanks, but notably Tanks 8F and 15H. These tanks represented sludge from the Purex and HM processes respectively. Each sludge class was found to closely follow the Bingham plastic flow model. A Bingham plastic is a non-Newtonian fluid that possesses a yield stress. This means that a prescribed amount of shear must be applied before the fluid begins to move. Once in motion, the material behaves like a Newtonian fluid. Fluids of this type include paint, river mud, and clay suspensions. In fact, the sludge at SRS is not unlike silt suspensions<sup>7</sup>, and kaolin clay slurries have been used as sludge surrogates.<sup>8</sup> Figure 2 represents a summary of the yield stress and solids concentration relationship for Purex and HM sludge.

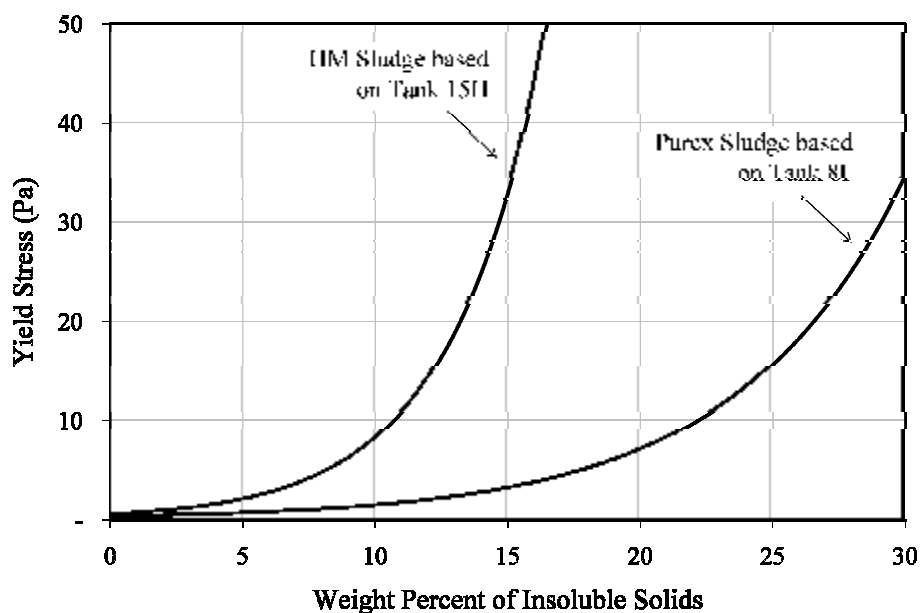


Fig. 2. The different processing areas at SRS produced waste forms that possessed different rheological properties. The data obtained from studies in the 1980s formed the basis for the subsequent waste removal programs.<sup>9</sup>

The transfer system from Tank 12H to Tank 51H had been evaluated to ensure the pump's capability and establish the drive motor setpoints.<sup>10</sup> The predicted flowrate rate was 26.1 m<sup>3</sup>/hr (115 gpm) at 67m (220 ft) of hydraulic head. This evaluation assumed a suspension of 11 weight % solids with a maximum yield stress of 10.8 Pa, which is consistent with the fluid properties of typical HM sludge. The assumed consistency (or plastic viscosity) was 9 cP.

## MIXING OPERATIONS AND TRANSFER ATTEMPTS

The four slurry pumps began operation in August 2008. As the sludge was ablated and suspended, the pumps were lowered in four increments. Total mixing time was 862 hours. During this time, periodic soundings were made to assist in lowering the slurry pumps. The tank level remained constant between 2.8 and 2.9m (110 and 116 inches). There was a small sealing water leak in one of the slurry pumps that caused the tank level to rise slightly during mixing operations. Based on previous experience, it was not expected that the sludge layer would ablate evenly, but large mounding or mining was not anticipated.

The first attempt at transferring the waste was made in December 2008 after 488 hours of mixing. Assuming a plugged well casing, 374 hours of additional mixing was performed before a second transfer attempt was made in January 2009.<sup>11</sup> During a post-mixing sludge sounding, operators reported that the steel weight used to obtain level measurements could float on the waste surface. Dip samples confirmed a thick and tenacious slurry suspension.

## SAMPLING AND RHEOLOGY

A non-diluted sample obtained on January 24, 2008 revealed the suspension to have a yield stress of 45 Pa and a plastic viscosity of 35 cP at an insoluble solids content of 10 weight % and a specific gravity of 1.38.<sup>12</sup> The yield stress was 4½ times the forecast, and the plastic viscosity was 3½ times over the predicted value. The specific gravity was also heavier (1.38 versus 1.23). A review of the transfer calculation confirmed the STP was undersized to transfer the slurry suspension. The Savannah River National Laboratory (SRNL) subsequently performed studies to predict the Tank 12H rheological properties at various weight percent solids. See Figure 3.

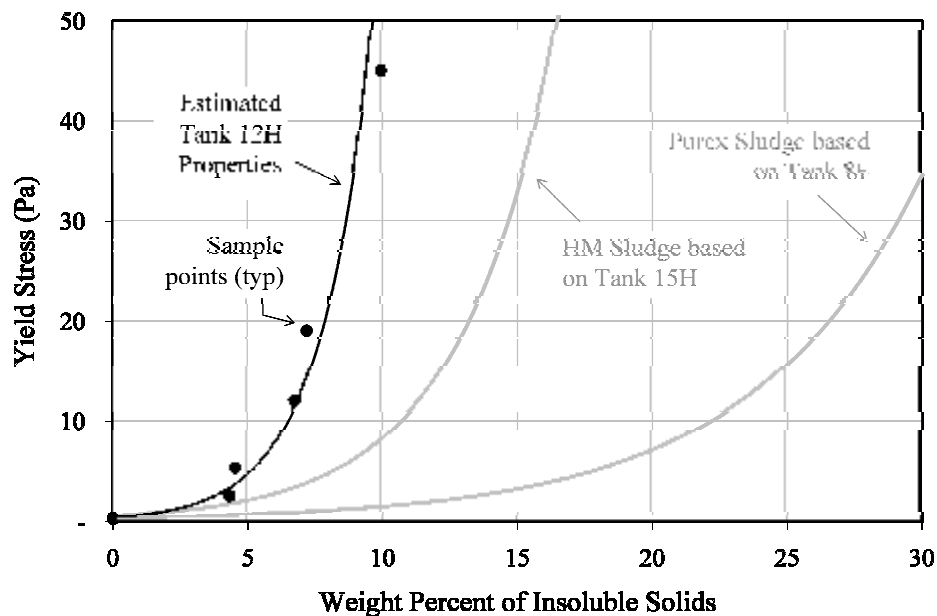


Fig. 3. Tank 12H sludge has drastically different rheological properties than other sludge tanks at the Savannah River Site.<sup>13</sup>

## RECOVERY AND PATH FORWARD

Studies at SRNL<sup>12, 13</sup> confirm that dilution of the sludge suspension will lower the yield stress and the plastic viscosity. The target yield stress for pumping was set at 10 Pa with a maximum allowance of 12 Pa. This equated to approximately 6-7 weight % solids. To underscore the dissimilarity among SRS sludge, a Purex slurry (as found in F-Area tanks) has a yield stress of 10 Pa with a corresponding solids content of 22 weight%. It is unclear why Tank 12H sludge has this type of fluid behavior. However, the history of the tank points to waste receipts containing thorium compounds. Suspensions of thorium compounds under caustic conditions have shown remarkable rigidity. Historical records indicate that significant dilution is needed to enable mobility of thorium hydroxide suspensions.<sup>14</sup> Goodlett reported such suspensions possess thixotropy as well as Bingham plastic properties, and dilution volumes of five times that of normal waste suspensions is needed to ensure flowability. There is also speculation that the fine particles of suspended aluminum compounds (Gibbsite and Boehmite) may also contribute to the high yield stress. Because Tank 12H has a varied receipt history (partly Purex, HM, and some Thorex), it is likely that vertical distribution of the waste is uneven. This supposition is consistent with earlier characterization reports.<sup>15</sup> Because of the suspicion of non-homogenous veins and outcroppings of sludge, the recovery plan did not involve unnecessary mixing and blending of the existing waste contents. Any unnecessary suspension of “new” sludge would exacerbate the high yield stress condition and may invalidate the dilution assumptions predicted by SRNL.

The recovery plan involved adding as much liquid to Tank 12 as allowed, and then blending to incorporate the fresh liquid.<sup>16</sup> Approximately 363 m<sup>3</sup> (96,000 gallons) of liquid was transferred from nearby Tank 24H (other sources of liquid included contaminated rain water previously collected in adjacent above-ground storage tanks). Only a few hours of blending with the slurry pumps were allowed before starting this first transfer. The recovery plan also included lifting and lowering the STP to ensure the well casing was clear of any high yield stress material. The first transfer commenced in March 2009 sending 644 m<sup>3</sup> (170,000 gallons) of slurry to Tank 51H. A second transfer of 704 m<sup>3</sup> (186,000 gallons) started in April 2009 after 265 m<sup>3</sup> (70,000 gallons) from Tank 24H was mixed for 34 hours. The slurry pumps were ultimately lowered to 0.25m (10 inches) off of the tank floor after adding more dilution liquid. The third transfer occurred in June 2009 and sent 632 m<sup>3</sup> (167,000 gallons) to Tank 51H. A fourth and final transfer of 382 m<sup>3</sup> (101,000 gallons) commenced later that month completing the Sludge Batch 6 feed specification.

Post transfer mapping studies estimated that 62% of the original sludge volume was removed from Tank 12H.<sup>17</sup> The mass of solids was estimated at 255,000 kgs. The final topography of the sludge layer showed drastic elevation differences. Figure 4 is a video screen capture of the Tank 12H interior one month after the last transfer.



Fig. 4. Post-transfer video in Tank 12H reveals uneven topography and significant mounding.<sup>11</sup>

## LESSONS AND CONCLUSION

Tank 12H was approached as a routine waste removal campaign. SRS had, to this point, performed seventeen sludge removal evolutions (starting in 1966) and provided feed to six discrete sludge batches without significant perturbations. Assumptions regarding sludge rheology were made based on historical (and repeatable) field data. Tank 12H had a slightly different waste receipt history than analogous tanks, but nothing remarkably dissimilar. Because of past successes and historical data, pre-transfer rheological studies were deemed superfluous and not performed. Several lessons resulted from the Tank 12 waste removal experience:

- Pre-transfer rheological studies are necessary; historical rheological data can only provide a rough estimate
- Sufficient tank space must be available to permit dilution liquid additions if needed
- Slurry pump performance varies significantly from tank to tank (because of differences in sludge properties); therefore, volumetric predictions of mobilized material can be erroneous
- Mixing the entire tank prior to transfer may not work for all waste removal campaigns

The Tank 12H waste removal program provided valuable insight to the varied and unpredictable nature of high-level waste sludge. Lessons involved with planning, analysis, and design have been incorporated into subsequent waste removal efforts.

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