RESOLUTION OF HIGH-HEAT ISSUE FOR HANFORD TANK C-106

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ABSTRACT

The high-heat safety issue for Hanford Tank C-106 was resolved in 1999 by sluicing the radioactive sludge from this 55-year old single-shell tank into a newer double-shell tank with better cooling capability. Approximately 704,000 liters of caustic sludge, containing 5 million Curies of Strontium-90 were transferred using an improved hydraulic sluicing technique. Recirculating radioactive supernate from the receiver tank was used to agitate and mobilize the sludge in Tank C-106 to minimize waste volumes. Special monitoring systems and procedures were employed to control slurry flow rates, waste temperatures, and flammable gases during the sluicing operation.

INTRODUCTION

In 1991 Tank C-106 was placed on the “watch list” of Hanford high-level waste tanks with a potential for significant release of radioactive material to the environment. The temperature in the center of the sludge in this tank was near boiling at 110-116°C. Approximately 23,000 liters of water were added to this tank per month to make up for evaporative losses and to keep the sludge covered with liquid. The heat generation rate by the radioactive contents of this tank was estimated to be approximately 30kW.

Without water additions, the sludge would have dried out, resulting in high temperatures that could have caused spallation of the concrete dome of the tank. This in turn could have led to collapse of the dome and a major radiological release.

In the event of a leak in this 55-year old tank, the water additions could not have been continued without flushing a large quantity of strontium, cesium, and plutonium into the ground.

The logical solution to the problem with Tank C-106 was to pump the waste into one of the newer double-shell tanks at Hanford. These double-shell tanks have two barriers to leakage, and none of the 28 double-shell tanks at Hanford have leaked to date. The double-shell tanks are also equipped with primary and secondary ventilation systems for cooling the waste without having to add water.

A hydraulic sluicing technique was used to pump the radioactive sludge out of Tank C-106 and into Tank AY-102 during 1998-1999. Similar methods were used previously at Hanford in the 1970’s, but during an earlier era of less stringent safety and radiological controls.

In addition to resolving the high-heat issue for Tank C-106, a secondary mission of this modern application of sluicing was to demonstrate that this technology can be used to retrieve sludge and salt cake waste from the other non-leaking single-shell tanks at Hanford, and to provide high-level sludge feed to demonstrate vitrification technologies.
TANK C-106

Tank C-106 was one of the first underground single-shell tanks built at Hanford to store high-level liquid radioactive waste. It was placed into service in 1944 and received various liquid wastes and sludges from the chemical processing of aluminum-clad, natural uranium plutonium production reactor fuel and subsequent uranium, cesium, and strontium recovery campaigns. This tank has a capacity of 2,000,000 liters and a diameter of 23 meters.

Tank C-106 is one of 149 single-shell tanks. It is made of a carbon steel lower liner approximately 1.3 cm thick, backed up by reinforced concrete, with an unlined dome of concrete, and covered by approximately 3 meters of soil. Approximately 67 of the other single-shell tanks at Hanford are suspected to have leaked; fortunately, Tank C-106 has not shown any leakage over its 55-year life.

WASTE CHARACTERIZATION

The waste in Tank C-106 consisted of 750,000 liters of thick sludge with a density of 1.55 g/ml, 1.8 meters deep, with less than a meter of supernate cover. The chemical composition and radiological content are shown in Table 1. The major chemical compounds were sodium, aluminum, and iron nitrates, nitrites, carbonates, hydroxides, phosphates, and silicates. The principal radioisotope was Sr-90 at about 5 million Curies, with lesser amounts of Cs-137 at about 300 thousand Curies, and a modest amount of about 100 kg of Pu-239.

<table>
<thead>
<tr>
<th>COMPOUND</th>
<th>AMOUNT</th>
</tr>
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<tbody>
<tr>
<td>A1</td>
<td>36,000 kg</td>
</tr>
<tr>
<td>CO₃</td>
<td>148,000 kg</td>
</tr>
<tr>
<td>Fe</td>
<td>47,000 kg</td>
</tr>
<tr>
<td>Na</td>
<td>145,000 kg</td>
</tr>
<tr>
<td>NO₂</td>
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<td>OH</td>
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</tr>
<tr>
<td>PO₂</td>
<td>14,000 kg</td>
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<tr>
<td>Si</td>
<td>18,000 kg</td>
</tr>
<tr>
<td>TOC</td>
<td>15,000 kg</td>
</tr>
<tr>
<td>Sr-90</td>
<td>5,000,000 Ci</td>
</tr>
<tr>
<td>Cs-137</td>
<td>300,000 Ci</td>
</tr>
<tr>
<td>Pu-239</td>
<td>3,000 Ci</td>
</tr>
</tbody>
</table>

The total organic compounds dispersed in the sludge was approximately 3%, resulting in one of the major problems that was encountered during sluicing. One of these organic compounds was bis(2–ethylhexyl)phosphate, which was employed 25 years previously for extracting Sr-90 from
the waste. This solvent decomposed into a series of volatile compounds which were retained in the sludge, but which escaped the laboratory analyses prior to the start of sluicing.

Based on process records, there was estimated to be six layers of different types of sludges in Tank C-106, each with varying chemical, physical, and radiological characteristics. Vertical core samples and grab (bottle) samples had been collected over the past 15 years for characterization purposes, and these were found to be very inhomogeneous. The upper layers of sludge were found to be loose and easily suspended, while the lower layers consisted of an impervious hard pan material. Lab analyses led to widely varying inventories and heat generation rates in the different layers. This in turn, complicated the prediction of peak waste temperatures and flammable gas generation rates (which depend on temperature, chemical composition, and isotope concentration).

Waste temperatures were predicted using thermal models at the peak unmonitored locations in both Tank C-106 and receiver Tank AY-102. Finite element modeling using the GOTHIC code, and estimated values of waste thermal conductivity were used. Process test data obtained with the ventilation system shutdown in C-106 was used to validate the models.

Flammable gas generation rates of 0.2-0.4 m$^3$/day of hydrogen were estimated from sensitive measurements of ventilation exhaust with inline instruments, and measurements by gas chromatography and mass spectrometry in the laboratory.

**RECEIVER TANK AY-102**

The waste from Tank C-106 was pumped into double-shell Tank AY-102. This is a newer tank built in the 1970’s to store the high-heat waste from PUREX reprocessing. Tank AY-102 is 23 meters in diameter and capable of holding 3,800,000 liters of waste. This tank consists of primary and secondary steel liners with a reinforced concrete shell and dome. It is equipped with a once-through annulus ventilation system that provides leak detection capability and cooling of the tank sides and bottom. It has a primary ventilation system for recirculating and exhausting the dome of the tank for both cooling and radiological emission control.

Tank AY-102 is equipped with 22 airlift circulators for mixing and convective cooling of the waste. However, the airlift circulators are ineffective when the bottoms are submerged in sludge, due to the wide spacing between the circulators. For this reason, the annulus ventilation and the dome ventilation systems for Tank AY-102 required major modifications to accommodate the hot waste from Tank C-106.

**SLUICING EQUIPMENT**

Two identical sets of pumping systems were installed in Tank C-106 and AY-102 (see Figure 1). A 30 kW centrifugal submersible pump was suspended in each tank by cable and winch to control the depth of submergence. The discharge from the submersible pump was directed through a flexible discharge hose into the suction of a 190 kW centrifugal booster pump, with mechanical nitrogen-supplied seals, that was mounted in a shielded pit on top of the tank. A variable flow of approximately 760-1,500 liter/min at a pressure up to approximately 700 kPa.
was discharged from the booster pump. The waste flowed through an above-ground, 10-cm diameter, double-encased piping system with 1-meter of soil shielding, along a distance of approximately 0.4 km between tanks. Slurry flowed into Tank AY-102 through a submerged header with four 2.5-cm outlets.

![Sluicing Equipment Schematic Drawing](image)

Supernate from high in the liquid layer of the AY-102 receiver tank was returned to Tank C-106 through a 2.5-cm diameter jet-nozzle sluicer assembly with a hydraulic aiming controller to agitate and mobilize the sludge in C-106. Both sets of pumps in both tanks operated simultaneously to set up a recirculating flow of slurry from C-106 to AY-102. This greatly minimized the creation of additional waste by eliminating water additions.

Tanks C-106 and AY-102 were equipped with special ventilation systems to reduce radiological emissions during sluicing and to provide waste cooling. Cooling was provided by recirculation through chillers. Filtration was provided with a combination of high efficiency mist eliminators (HEME’s), high efficiency metal filters (HEMF’s), and HEPA filters on the exhausts to keep the tanks at approximately 1-inch wg vacuum. Isokinetic stack monitoring systems were installed. In addition, Tank C-106 was provided with a chilled water cooler on the air inlet. The annulus ventilation system on Tank AY-102 was modified so that most of the cool air intake was directed to the bottom of the tank for waste cooling.

**INSTRUMENT APPLICATION**

Tanks C-106 and AY-102 were equipped with thermocouple trees to measure waste temperature profiles at approximately 0.6-meter vertical intervals (2 radial locations in C-106 and 25 radial locations in AY-102, plus concrete shell monitoring).

Both tanks were equipped with in-tank, color video systems with pan, tilt, and zoom, to observe pump and hydraulic sluicing operation, and to map the topography of the sludge during and after sluicing. Sludge buildup on the camera lens and lights was minimized by a built-in pressurized spray ring.
Liquid levels in both tanks were measured with ENRAF\textsuperscript{tm} displacement gauges accurate to within 0.13 cm. Sludge level in Tank AY-102 was measured with an ENRAF\textsuperscript{tm} densitometer that penetrated into the waste.

Slurry flow rates were monitored and controlled with in-line flowmeters (magnetic and Coriolis effect flowmeters were both used). A Coriolis flowmeter that measured both density and mass flow rate in the slurry line was very effective in reliably measuring slurry concentrations down to approximately 1-3 weight percent.

Grab samples collected at depth with cable-actuated bottles were periodically collected from Tank AY-102 after sluice batches for chemical, physical, and radiochemical hot cell analysis.

Standard Hydrogen Monitoring Systems employing electrochemical cells, gas chromatographs, and photo-acoustics were installed on the tank domes for C-106 and AY-102 to continuously measure concentrations of hydrogen, ammonia, nitrous oxide, and methane. The range of the instruments was from a few ppm up to the lower flammable limits.

Flame and photo-ionization detectors (FID’s and PID’s) were used to monitor emissions of volatile organic compounds from the ventilation stacks during sluicing.

A computer-based data acquisition system, relying on both hard-wired and telemetry inputs, was used to collect monitoring and pumping data during sluicing for process control and assessment.

**SAFETY ANALYSIS**

A comprehensive analysis of hazards and potential accidents was performed in a supplement to the tank farm Final Safety Analysis Report. The primary risks requiring engineering and process controls were:

- Transfer line leaks
- Ventilation releases
- Flammable gas deflagrations
- Steam bumps

Leakage accidents were mitigated by back flow preventors and by safety class pit and pipeline leak detectors and covers.

Ventilation releases were mitigated by safety-class stack continuous air monitors and fan interlocks.

Steam bumps and flammable gas deflagrations were prevented by temperature, flammable gas, and waste level monitoring, together with a special process control plan. There was a hypothetical concern that sluicing would cause “fluffing” of the sludge, which would increase the depth of the sludge that was transferred into receiver Tank AY-102. A thicker sludge layer could result in significantly higher sludge temperatures as well as increased gas retention in the sludge, which might be released periodically by buoyant displacement at flammable levels.
PROCESS CONTROL

Process controls were used to limit the slurry concentration to less than 30 weight percent; this was the assumed value for the accident analysis, and the maximum value to prevent pipeline plugging. Line velocity was kept above 1.8 m/sec to prevent settling of solids. The slurry concentration was controlled by adjusting the sluice and slurry pump flow rates, the depth of the slurry pump, the depth of the supernate, and the direction of the sluice jet.

Sluicing batches were limited to 12-hour duration each. Following the transfer of each 0.3-m sludge increment, temperatures were monitored for approximately five days to confirm thermal predictions. After campaigns of 0.6-meter each, grab samples of waste in the AY-102 receiver tank were analyzed to determine isotopic heat generation rates and sludge dissolution quantities. The amount of sludge transferred in each batch was determined by the inline mass flowmeter and by the waste densitometer in AY-102.

Because the sludge in tank C-106 was within approximately 5°C of the boiling point, the supernate head was controlled to prevent flashing. Also, 12-day hold periods were initially imposed to allow sludge cooling before lowering the supernate level for the next sluice batch.

During the hold periods between sludge campaigns, the release of flammable gas from both tanks was compared to baseline values to assure that gas was not being retained in the sludge at dangerous levels. Waste levels, corrected for evaporation rates, were also monitored to detect gas retention. Gas samples were analyzed to make sure that the composition did not change.

Midway through the sluicing, 15,000 liters of caustic (concentrated sodium hydroxide) had to be added to stay within corrosion control pH specifications, due to caustic consumption from solids.

OPERATING RESULTS

A total of 20 batches of sludge were transferred over a period of 12 months. See Figure 2 for cumulative sludge transfer based on the measured sludge depth in Tank AY-102. Approximately 704,000 liters or 97% of the sludge was removed from Tank C-106, including most of the hard pan solids on the bottom. The in-line mass flowmeter gave sludge removal quantities comparable to (i.e., within about 10%) the AY-102 densitometer after adjustment for solids dissolution.
The temperature trend near the center of Tank C-106 is shown in Figure 3. The initial temperature of 52°C was low due to a convection cell around the thermocouple tree and the layering of heat generating isotopes. Once sluicing started, the temperature peaked near the center at about 113°C. The temperature of C-106 dropped steadily as sludge was removed, ending at less than 21°C at completion.
The temperature trend in Tank AY-102 (Figure 4) increased steadily from about 21°C to about 60°C as a result of the sludge addition. From Figure 2, there was no evidence of waste fluffing in AY-102 which would cause higher temperatures or gas retention. The current waste temperature in tank AY-102 is approximately 22°C less than best estimate predictions, apparently due to lower Sr-90 in tank C-106 than initially estimated by thermal modeling. Flammable gas release has stayed near the lower baseline value, probably since equilibrium has not re-established between generation and release rates. Both temperatures and flammable gases will be monitored for a one-year period to detect any adverse trends.
The following lessons were learned for application to future waste retrieval operations:

1. High levels of volatile organic compounds from long-term accumulation of solvent radiolytic degradation products (mainly 3-heptanone) were released during initial sluicing operations. Extensive gas characterization studies and personnel respiratory protection measures had to be implemented and should be included in operational planning in the future. Air treatment systems were evaluated and found to be impracticable.

2. The flexible discharge hose on the slurry pump created a loop seal. This prevented priming and required the line to be blown out prior to each use. The hose also kinked when lowered, and was collapsed by the booster pump suction during high slurry loadings. A shorter hose would have solved this problem.

3. The slurry pump could not be lowered to the bottom of the tank, probably due to either hard sludge under the pump or foreign objects that flowed under the pump. A second sluice jet on the same side of the tank as the slurry pump would have helped.
4. The ventilation system flow rates were undersized. Extensive efforts had to be taken to seal tank penetrations to achieve acceptable vacuums for radiological filtration.

CONCLUSIONS

Sluicing of sludge from single-shell Tank C-106 into double-shell Tank AY-102 has resolved the high-heat issue, and demonstrated sluicing technology with modern equipment. Hydraulic sluicing is an effective technique for removal of both soft sludge and hard pan solids from non-leaking single-shell tanks. Industrial-grade pumps, pipelines, ventilation, and instrumentation systems are available for sluicing.

For this particular application, sluicing of caustic sludge mixtures did not cause fluffing of the sludge, which could have resulted in higher sludge temperatures or increased flammable gas retention.