INNOVATIVE TECHNOLOGY
Summary Report

DOE/EM-0520

# Sludge Washing

Tanks Focus Area

*Prepared for*

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INNOVATIVE TECHNOLOGY
Summary Report

# Sludge Washing

OST/TMS ID 233

Tanks Focus Area

*Demonstrated at*
Oak Ridge National Laboratory
Oak Ridge, Tennessee
Pacific Northwest National Laboratory
Richland, Washington

INNOVATIVE TECHNOLOGY
Summary Report

## *Purpose of this document*

Innovative Technology Summary Reports are designed to provide potential users with the information they need to quickly determine whether a technology would apply to a particular environmental management problem. They are also designed for readers who may recommend that a technology be considered by prospective users.

Each report describes a technology, system, or process that has been developed and tested with funding from DOE's Office of Science and Technology (OST). A report presents the full range of problems that a technology, system, or process will address and its advantages to the DOE cleanup in terms of system performance, cost, and cleanup effectiveness. Most reports include comparisons to baseline technologies as well as other competing technologies. Information about commercial availability and technology readiness for implementation is also included. Innovative Technology Summary Reports are intended to provide summary information. References for more detailed information are provided in an appendix.

Efforts have been made to provide key data describing the performance, cost, and regulatory acceptance of the technology. If this information was not available at the time of publication, the omission is noted.

All published Innovative Technology Summary Reports are available on the OST Web site at http://ost.em.doe.gov under "Publications."

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# SECTION 1
# SUMMARY

## Technology Summary

**Problem**
The U.S. Department of Energy (DOE) has approximately 340 million liters of waste left from weapons production activities stored in underground storage tanks. Much of this waste is stored at the Hanford Site, the Savannah River Site (SRS), Idaho National Engineering and Environmental Laboratory, and Oak Ridge National Laboratory (ORNL). These wastes have a wide range of chemical and physical properties and radiation levels as high as 10,000 rad/h. A large percentage of this waste is highly radioactive sludge.

DOE plans to immobilize the high-level radioactive sludge by vitrifying it into glass. Certain chemical constituents in the sludge (such as phosphorus, sulfur, and chromium) have limited solubility in molten glass, which allows only limited amounts of these chemicals to be added to each glass log. Washing sludge using a concentrated caustic solution is known as "enhanced sludge washing" (ESW). ESW removes some of these constituents from the sludge but leaves the radionuclides in the sludge for vitrification. ESW can also include other process enhancements or the addition of chemicals to further reduce the volume of high-level waste (HLW) glass or improve glass processability and durability.

**How It Works**
The term "enhanced sludge washing" refers to the process of extracting components from sludges with strong caustic solution, as opposed to simple sludge washing with only water. ESW removes nonradioactive components such as aluminum, chromium and phosphate salts from tank solids. (A small amount of the radioactive cesium is also removed with the nonradioactive components and is later removed.) The wash liquid forms part of the liquid low-activity waste (LAW), which is further treated to reduce the concentration of radionuclides, then is sent for LAW immobilization and on-site disposal. The solid HLW from solid/liquid separation contains the radionuclides remaining in sludges after ESW and is sent to HLW immobilization and off-site disposal.

ESW serves several purposes:

- It removes more of the nonradioactive components from radioactive sludges than simple sludge washing.
- It reduces the resulting volume of glass, reducing the ultimate cost of disposal. The total costs associated with immobilization and disposal of HLW were estimated to be $450/kg of waste oxide, compared to $15/kg for LAW (DeMuth and Shieh 1998).

## Demonstration Summary

ESW was implemented at SRS in 1993 in the Extended Sludge Processing facility. Figure 1 is a diagram of Hanford tank waste treatment. In 1993, ESW was selected as the baseline at Hanford; however, Hanford's Federal Facility Agreement and Consent Order (Tri-Party Agreement) required a complete evaluation of ESW to determine whether the process produced a reasonable volume of glass canisters (Jenson 1994). Sludge samples were washed in the laboratory, and the results were used to project the number of HLW canisters that would be produced from Hanford. In 1998, DOE determined that the ESW process was acceptable for pretreatment of Hanford tank sludges and that advanced separation processes would not be required.

Since 1993, Tanks Focus Area (TFA) sludge treatment studies included alkaline washes of Hanford and ORNL sludges, partitioning of sludge components by caustic leaching, and countercurrent decantation. In fiscal year 1998 (FY98), the focus of the research changed to parametric studies to test the effect on

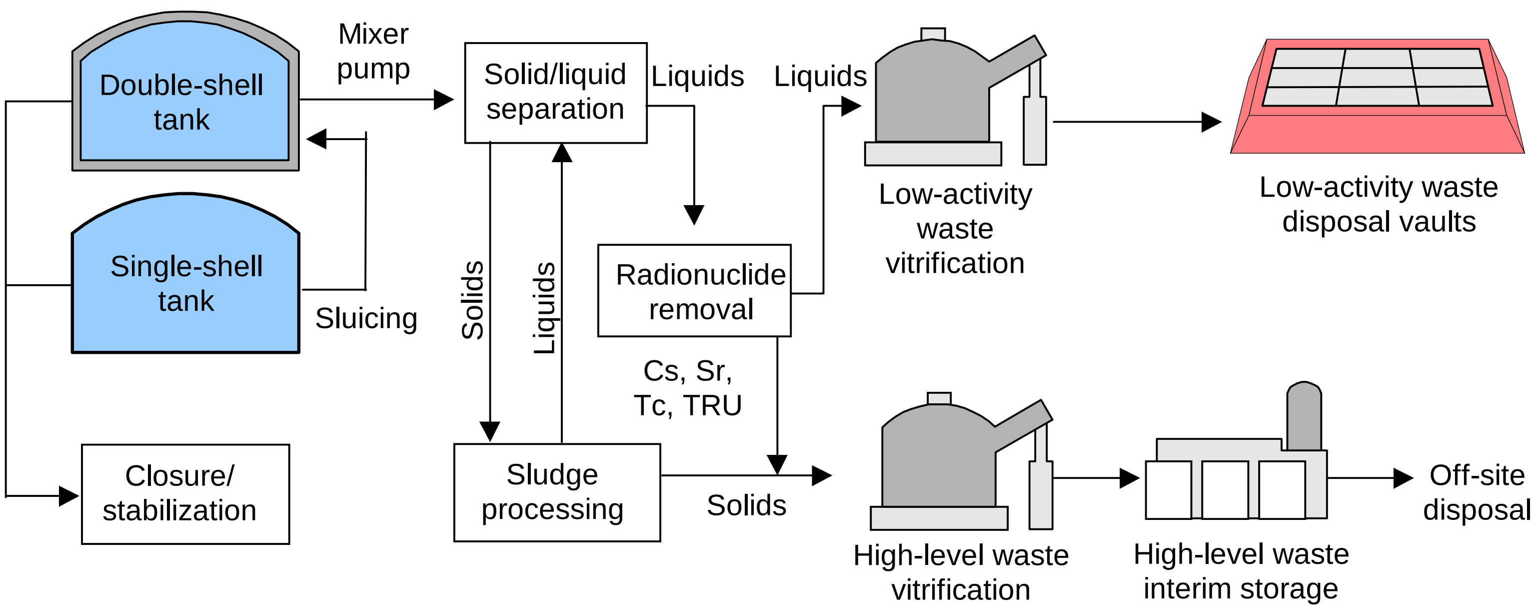


**Figure 1. Hanford tank waste treatment.**

overall performance of three main process parameters—leaching time, leaching temperature, and sodium hydroxide (NaOH) concentration. Selected accomplishments of the project include

- completing sludge treatment studies using Oak Ridge Melton Valley Storage Tank sludge samples (FY95),
- evaluating aluminum concentration in leachates for ESW (FY96),
- evaluating solids formation in filtered leachates and wash solutions from Hanford tank sludges (FY97),
- completing parametric tests of Hanford tank sludges to evaluate temperature and caustic concentration (FY98), and
- conducting chromium leaching tests on Hanford tank sludges (FY98).

**Potential Markets**

Results from these studies apply to pretreatment activities at DOE sites for removal of entrained radionuclides, salts, and minerals from highly radioactive sludge. At Hanford, these studies fulfilled a regulatory requirement (Tri-Party Agreement milestone M-50-03-T2B) to project the impact of ESW on glass production at the Hanford Site and provided a basis for the following benefits:

- reducing risks associated with implementation of the proposed Hanford baseline treatment system;
- reducing the amount of tank waste requiring processing and disposal as HLW, resulting in reduced costs; and
- providing a basis for a Request for Proposal for privatizing Hanford HLW treatment.

Drivers at all sites include improving the efficiency of full-scale vitrification processes and minimizing the volume of waste that must be treated for disposal at expensive off-site locations.

## Contacts

**Technical**

Rodney Hunt, Principal Investigator–Oak Ridge National Laboratory, (865) 574-5481, huntrd@ornl.gov
Gregg J. Lumetta, Principal Investigator–Hanford, (509) 376-6911, gregg.lumetta@pnl.gov
Donald J. Temer, Principal Investigator–Los Alamos National Laboratory, (505) 667-9636, dtemer@lanl.gov

**Management**

Kurt D. Gerdes, DOE–Headquarters Tanks Focus Area Lead, (301) 903-7289, kurt.gerdes@em.doe.gov
Ted P. Pietrok, DOE–Richland Tanks Focus Area Lead, (509) 372-4546, theodore_p_pietrok@rl.gov
C. Phil McGinnis, DOE–Oak Ridge Technology Integration Manager for Pretreatment, (865) 476-6845, cpz@ornl.gov

**Other**

All published Innovative Technology Summary Reports are available on the OST Web site at http://ost.em.doe.gov under “Publications.” The Technology Management System (TMS), also available through the OST Web site, provides information about OST programs, technologies, and problems. The OST/TMS ID for sludge washing is 233.

SECTION 2
# TECHNOLOGY DESCRIPTION

## Overall Process Definition

**Description of Technology**
TFA has conducted numerous laboratory tests of ESW. ESW testing consists of a series of steps in which sludge is mixed with wash solutions to separate the key elements (generally chromium, phosphorus, sodium, and aluminum). General process steps include the following:

- A weighed sample of sludge is mixed with a specific volume of inhibited water (0.01 molar [*M*] NaOH plus 0.01 *M* sodium nitrite solution). The mixture is stirred for a minimum of 30 minutes.
- Solid/liquid separation is done by centrifuge. The liquid is decanted off the solid, and a second volume of wash solution is added to the solid.
- The wash-separation cycle is repeated, with a composite of the wash solutions analyzed for key components and radionuclides. Ideally, the key components are removed, but the radionuclides—with the exception of cesium—remain in the sludge. During the repeated wash cycles, the early wash solutions are generally colored, and later wash solutions are progressively less colored.
- After the wash steps, the solids are leached with concentrated NaOH. The amount added to the sludge depends on the amounts of aluminum, phosphorus, and chromium present in the sludge, with a slight excess added over the stoichiometric requirements.
- The final product of the ESW test is a liquid fraction containing the removed chromium, phosphorus, sodium, and aluminum (and small amounts of cesium) and a solid fraction containing the majority of the radioactive components.

Figure 2 is a photograph of ESW bench-scale test equipment.

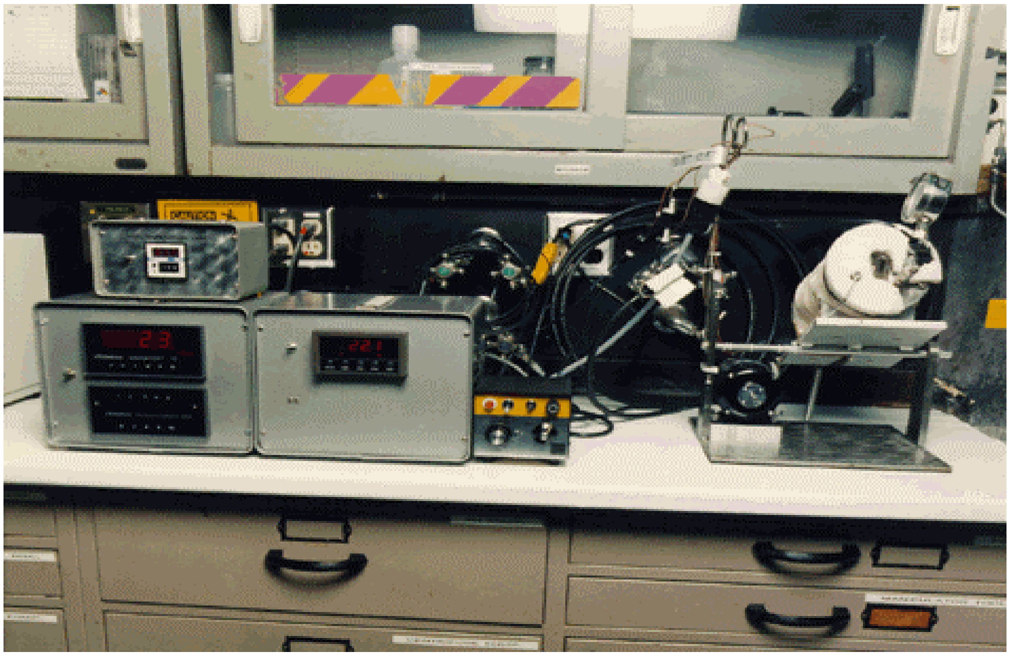

**Figure 2. Apparatus used for conducting enhanced sludge washing experiments in the hot cell at Oak Ridge National Laboratory.** (Photo taken on bench top before installing in hot cell.)
Source: Hunt et al. 1998

**Full-Scale DOE Applications**

In the full-scale process at Hanford, sludges will be mobilized with sluicers and mixer pumps inside the original underground storage tanks. Once retrieved, the tank contents will be sent to solid/liquid separation processes. The liquid fraction from solid/liquid separation will be further treated to remove radionuclides, then sent for LAW immobilization and on-site disposal. The solid fraction will be sent to sludge processing (included ESW) and then to HLW immobilization and off-site disposal.

At SRS, the ESW process is described in the HLW system plan (WSRC 1998). Sludges are transferred from HLW storage tanks to tanks at the Extended Sludge Processing Facility. The system consists of three tanks where the sludge is mixed with caustic and washed to reduce the aluminum concentration. After decanting, the washed sludge becomes feed to the Defense Waste Processing Facility.

**Basic Principle of ESW**

Understanding the process chemistry remains an important area of ESW research. Several mechanisms are likely to contribute to the dissolution of insoluble species. In some cases, insoluble metal salts are converted to soluble hydroxide salts that dissolve in the wash solutions. In the case of phosphorus and sulfur, a metathesis reaction forms soluble sodium salts from insoluble phosphates and sulfates. Chromium occurs in tank waste in two different oxidation states, Cr(III) and Cr(VI). The chromium found in the washing and leaching solutions is primarily Cr(VI), indicating that one part of the reaction mechanism must be chromium oxidation. One possible oxidation reaction is shown as the final reaction below. The major pathways used to estimate the amount of hydroxide that will be consumed during the process are listed below. One mole of aluminum will react with one mole of hydroxide, each mole of phosphate consumes three moles of hydroxide, and one mole of chromium may consume one or two moles of hydroxide, according to the following formulas:

Boehmite $AlOOH\ (s) + NaOH\ (aq) \rightarrow NaAlO_2\ (aq) + H_20$

Gibbsite $Al(OH)_3\ (s) + NaOH\ (aq) \rightarrow NaAlO_2\ (aq) + 2H_20$

$FePO_4\ (s) + 3NaOH\ (aq) \rightarrow Fe(OH)_3\ (s) + Na_3PO_4\ (aq)$

$Cr(OH)_3\ (s) + NaOH\ (aq) \rightarrow Na[Cr(OH)_4]\ (aq)$

$4Cr(OH)_3\ (s) + 3O_2 + 8OH \rightarrow 4CrO_4^{-2}\ (aq) + 10H_2O$

The chemistry of these elements during tank waste treatment is complex, and these may not be the only reactions they undergo during ESW.

**Demonstration Goals and Objectives**

The objectives of the various studies at Los Alamos National Laboratory (LANL), ORNL, Pacific Northwest National Laboratory (PNNL), and SRS are summarized in Table 1.

Before FY98, laboratory-scale testing was conducted using small (approximately 5-g) samples of actual wastes. Issues addressed included

- solubility of certain key components such as aluminum,
- the amount of bulk sludge dissolved at elevated temperatures,
- testing on difficult-to-dissolve sludges, and
- solids settling times and solids mixing.

In FY98, the focus of testing shifted to performing parametric tests on selected sludge samples. The purpose of the parametric tests is to vary individual process parameters one at a time so that process engineers can optimize full-scale process flow sheets for specific waste types.

Another focus of ESW process development is to understand the conditions under which unwanted solids form during ESW. Under certain circumstances that are not well understood, some sludges form precipitates and gel-like materials during reaction with NaOH. This could be a significant problem in a

**Table 1. Demonstration goals and objectives for enhanced sludge washing studies**

| Study | Performer | Years | Objectives |
|---|---|---|---|
| Sludge treatment studies | ORNL | 1994–1997 | • Conduct bench-scale tests on Melton Valley Storage Tank sludges to develop comprehensive process flow sheet<br>• Evaluate distribution of chemical species between liquids and solids |
| Hanford sludge wash and leach factors | LANL<br>ORNL<br>PNNL | 1994–1997 | • Conduct small-scale laboratory tests (5–20 g) to determine the effectiveness of enhanced sludge washing for Hanford Phase II sludges |
| Sludge partitioning chemistry | PNNL | 1996–1997 | • Maximize removal of troublesome components that affect waste vitrification rate or melter lifetime<br>• Understand leaching of actinides from Hanford sludges |
| Dissolution of chromium | PNNL | 1996–1998 | • Enhance chromium dissolution of Hanford sludges |
| Countercurrent decanting | SRS | 1996–1997 | • Evaluate out-of-tank process for enhanced sludge washing |
| Chemical modeling of sludge washing | ORNL<br>PNNL | 1995–1996 | • Develop thermodynamic models to predict enhanced sludge washing performance |
| Parametric studies of Hanford sludge washing | LANL<br>ORNL<br>PNNL | 1998 | • Evaluate caustic dissolution behavior of a broad range of components on actual sludge samples using up to 6 *M* sodium hydroxide and temperatures up to 95°C<br>• Understand the volume of HLW that would be produced at Hanford |
| Prevention of solids formation | ORNL | 1998–1999 | • Achieve 70–80% removal of aluminum, chromium, and phosphate without unwanted solids formation |

large-scale process, leading to clogged pipes and necessitating repairs and delays. The goal is to be able control their formation through process controls (e.g. adjustment of pH, temperature, or leaching time) or chemical addition. Addition of lime (calcium oxide) has shown some promise; samples with lime added did form a gel immediately, but after six months the gel had disappeared. Computer simulations of the solubility of compounds of concern are also being developed and tested (Beahm et al. 1998).

## System Operation

Bench-scale testing of ESW has been under way to identify process parameters to adequately remove unwanted chemicals from the sludge without solids formation. Testing is carefully controlled, occurs in the hot cell, and uses trained laboratory technicians. Requirements for implementing the full-scale process are under development. Some of the basic system operation requirements for ESW that must be considered for large-scale operation are summarized below.

### Special Operational Parameters

- Due to the highly radioactive nature of HLW sludge, ESW must be performed in an underground storage tank or a shielded facility.

- Operating conditions must be maintained to prevent unwanted precipitation of aluminum salts and other unwanted solids in the wash water.

- A series of washing steps at varying pH may be required to adequately transfer unwanted sludge components into supernatant.

- Wash steps should not dissolve the transuranic components to be immobilized in the HLW. It is understood that some cesium will be dissolved by this process; it will later be removed from the LAW stream.

- The washed sludge must be compatible with the vitrification process.

- Under certain circumstances that are not well understood, some sludges form precipitates and gel-like materials during reaction with NaOH. This event could be a significant problem in a large-scale process, clogging pipes, necessitating repairs, and causing delays.

**Materials, Energy, Other Expendable Items**

- ESW increases the volume of radioactive tank liquids that must undergo radionuclide separation and LAW treatment.

- ESW reduces the volume of HLW glass produced. Costs associated with HLW vitrification and disposal are thereby reduced.

**Personnel Required**
In-tank ESW is a batch process that requires operators to monitor process parameters and determine when washing is complete.

**Secondary Waste Stream**
The spent wash waters from ESW form a liquid waste stream that contains small amounts of cesium. This liquid waste stream is treated to remove radionuclides, then can be vitrified or grouted as LAW and disposed of on site. The solids remaining after ESW contain the bulk of the radionuclides and are sent for HLW vitrification and disposal off site.

**Potential Operational Concerns and Risks**

- Remote handling is required due to the highly radioactive nature of the waste.

- When sludges are being retrieved from multiple tanks, the sludges should be retrieved and blended such that the concentration of unwanted chemicals in the sludge does not exceed feed specifications for vitrification. Care must also be taken not to blend incompatible wastes.

# SECTION 3
# **PERFORMANCE**

## Demonstration Plan

Sludge treatment has been investigated since 1993 by the Hanford Tank Waste Remediation System (now the Office of River Protection), LANL, ORNL, and PNNL. Sludge treatment studies have been funded under TFA, the Efficient Separations and Processing Crosscutting Program, and end user programs. Table 2 summarizes the various studies.

Potential future development efforts include the following:

- Evaluating the need for and availability of process monitoring and control technology. To date, researchers have suggested using conductivity probes (similar to those used by the aluminum industry to monitor high-temperature caustic leaching of bauxite) and gamma spectrometry to measure $^{137}Cs$ activity. In-tank sensors that can monitor the concentration of soluble constituents in real time are under development.

- Evaluating methods to increase the solubility of chromium in alkaline processes. Much of the chromium may be present as the Cr(III) ion, which is much less soluble than the Cr(VI) ion. Oxidizers such as ozone and permanganate were being tested to increase the solubility of chromium.

- Testing processes to increase sludge separation, such as performing solid/liquid separation at elevated temperatures.

- Testing process parameters or chemical additions that decrease the formation of aluminosilicate gels and precipitates.

## Results

This section summarizes key results of FY98 TFA-funded parametric studies at PNNL, ORNL, and LANL to show the efficacy of ESW under the different processing conditions. The goal of this testing was to collect parametric data for sludge samples representing 90% of the sludge volume in Hanford tanks. The parameters tested were

- length of time of the leach step, ranging 4–168 h;
- temperature of the leach step, ranging 60–100°C; and
- concentration of caustic solution, ranging 0.1–6 *M* NaOH.

Figure 3 shows the aluminum concentration as a function of time during the caustic leaching of Hanford tanks BX-110 and BX-112 at three different temperatures and two different NaOH concentrations. Together these graphs illustrate several results commonly seen in ESW:

- Even though both tanks received waste primarily from the first decontamination cycle of the bismuth phosphate process, they respond very differently to ESW. The initial concentration of aluminum is approximately 2000 µg/mL, but with increasing leach time and temperature, the two sludges behave completely differently.

- The 1 *M* leach solution from BX-110 shows an increase in aluminum concentration with temperature. In every case, more than 95% of the aluminum was removed from the solid matrix after leaching for 168 h. When leaching with 3 *M*, aluminum dissolution was more rapid, with concentrations in the leach solution reaching more than 90% of their final concentration within the first 5 h. This is an important process design result: for aluminum removal from BX-110, the same removal can be achieved with a less concentrated solution at a lower temperature for a longer time as with a stronger solution at a higher temperature for a shorter time.

**Table 2. Review of major enhanced sludge washing studies**

| Study | Site | Process evaluated | Major conclusions | Publications |
|---|---|---|---|---|
| Sludge treatment studies | ORNL | ORNL solid/liquid separation | • Developed a sludge processing flow sheet for ORNL tank wastes | Collins et al. 1997 |
| Effectiveness of Hanford enhanced sludge washing | LANL<br>ORNL<br>PNNL | Sludge wash and leach factors | • Total Hanford high-level waste glass volume satisfies Hanford Tri-Party Agreement milestone M-50-03-T2B<br>• Chromium inventory in sludge could double the required volume of high-level waste glass | Colton 1995<br>Colton 1996<br>Colton 1997<br>Temer and Villarreal 1995<br>Temer and Villarreal 1996<br>Temer and Villarreal 1997 |
| Sludge partitioning chemistry | PNNL | Hanford enhanced sludge washing | • Leaching efficiencies less than predicted for aluminum, chromium(III), and phosphorus<br>• Cesium-137 will need to be removed from washing and leaching solutions<br>• Mass of sludge solids can be reduced further using commercial leach agents<br>• Work needed to understand settling behavior after enhanced sludge washing | Brooks, Myers, and Rappe 1997<br>Lumetta and Rapko 1994<br>Lumetta, Rapko, and Wagner 1996<br>Lumetta et al. 1996a<br>Lumetta et al. 1996b<br>Lumetta 1997<br>Lumetta et al. 1997 |
| Dissolution of chromium | PNNL | Oxidative chromium leaching | • Higher hydroxide concentrations increase dissolution of chromium and plutonium (not desired)<br>• Alkaline oxidative leaching using permanganate increases the dissolution of chromium but not plutonium | Fedoseev et al. 1998<br>Peretrukhin et al. 1996<br>Rapko et al. 1996<br>Rapko, Delegard, and Wagner 1997<br>Rapko and Wagner 1997<br>Rapko 1998 |
| Counter-current decanting | SRS | Out-of-tank process for sludge washing | • Continuous industrial process offers high production rates and reduced wash water requirements<br>• Costs for remotely operated, shielded facility need to be better understood | Peterson, Hay, and Lee 1997 |
| Chemical modeling of sludge washing | ORNL<br>PNNL | Hanford and ORNL enhanced sludge washing | • TEMPEST model can be modified to predict sludge dissolution and precipitation<br>• SOLGASMIX code can predict solubilities in caustic solutions | Onishi, Reid, and Trent 1995<br>Weber and Beahm 1996 |
| Parametric studies of Hanford sludges | ORNL<br>LANL<br>PNNL | Enhanced sludge washing to supply feed to private vendor | • Aluminum and chromium need to be removed by caustic leaching<br>• Aluminum and chromium leach factors increase with elevated temperatures or longer leaching times<br>• Aluminum removal can decrease when leach time and temperature are increased because of the formation of aluminosilicate minerals<br>• Aluminosilicates precipitation during ESW can be monitored using conductance probes | Brooks et al. 1998<br>Hunt, Collins, and Chase 1998<br>Lumetta et al. 1998 |
| Prevention of solids formation | ORNL | Hanford enhanced sludge washing | • Chemical additives can reduce unwanted solids formation | Beahm et al. 1998 |

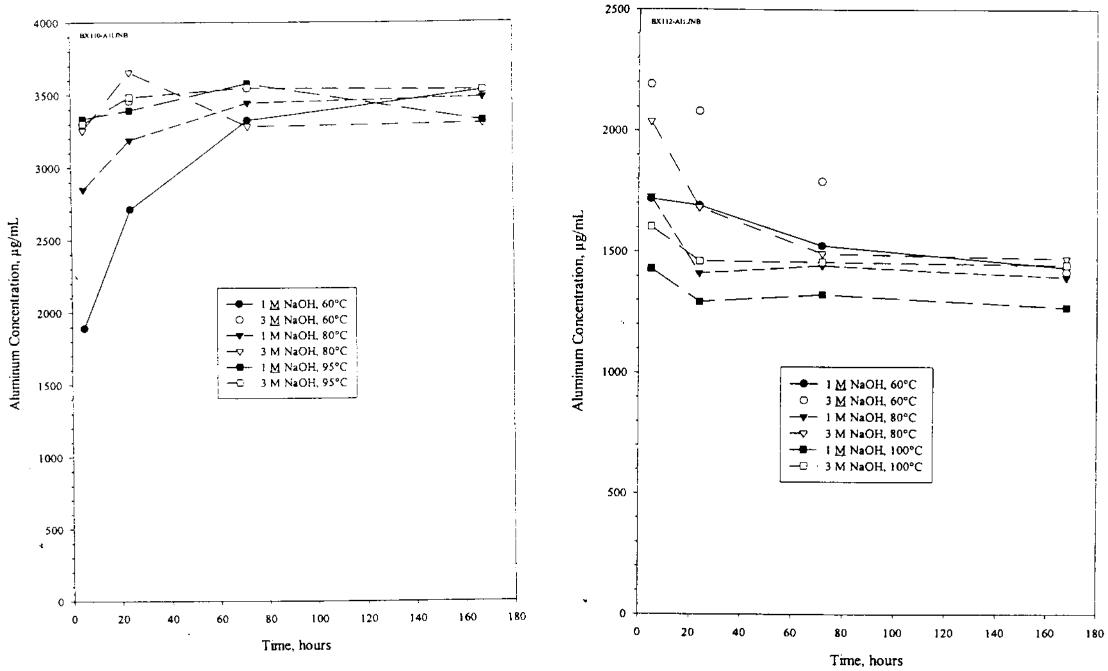


**Figure 3. Aluminum concentration in leach solution as a function of time during the caustic leaching of Hanford tanks BX-110 (left) and BX-112 (right).**
Note different *y*-axis scale on graphs. *Source:* Lumetta et al. 1998.

- The concentration of aluminum in the leach solutions from BX-112 decreased with time. This decrease was more gradual for 1 *M* leach solution at 60°C than it was for concentrated caustic solutions and higher temperatures. Transmission electron microscopy showed that, after sludge washing, the solids contained aluminum phosphates. It was hypothesized that those were converted to sodium aluminosilicate solids during leaching.

In contrast, Figure 4 shows that the chromium removal response from the two sludges is more similar. Leach solutions from both tanks show an increase in chromium concentration with time, increased temperature, and increased NaOH concentration. For BX-112, there was a definite improvement in chromium removal with an increase in temperature from 60 to 80°C, but not from 80 to 100°C. As with aluminum removal from BX-110, these data show that leaching time, leach solution concentration, and leaching temperature could all be used as process control variables for the targeted level of chromium removal from BX-110 and BX-112.

The low solubility of chromium may generally be a problem for ESW. Chromium in Hanford tank waste has generally been found to exist in the Cr(III) oxidation state. Chromium removal may be enhanced by oxidation with permanganate or ozone treatment to oxidize the less soluble Cr(III) to the more soluble Cr(VI).

Table 3, a summary of overall ESW removal of aluminum, chromium, and phosphorus results for five Hanford tanks, shows that phosphorus is the most easily removed of the three elements. The results also show the variability between tanks and different leaching conditions. For example, though 100% of the phosphorus was removed from BX-110 sludge, only 60% was removed from C-102 sludge.

Before ESW testing started, it was known that sludges from different tanks could have very different chemical and physical compositions, based on the process that generated them, mixing different waste streams, further processing that had been conducted in the tank, and simply the radionuclide decay and chemical aging process that had occurred over time. A major result of all the ESW tests is the confirmation that the sludges can behave in very different ways that are difficult to predict.

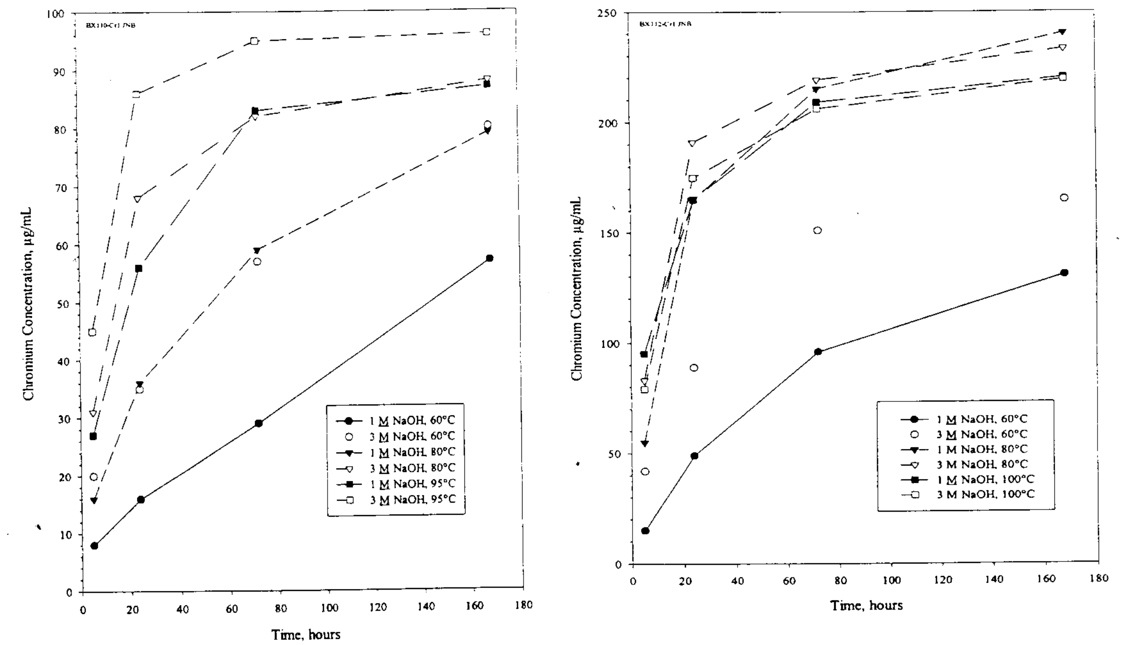


**Figure 4: Chromium concentration in leach solution as a function of time duri the caustic leaching of Hanford tanks BX-110 (left) and BX-112 (right).**
Note different *y*-axis scale on graphs. *Source:* Lumetta et al. 1998.

- Some sludges form insoluble compounds during caustic leaching and actually show increased sludge dry weight after leaching. (The main contributor is thought to be sodium aluminosilicate.) In ORNL studies of water-washing and caustic leaching of S-101 sludges, the aluminosilicates were able to redissolve with longer leaching times; in PNNL studies they were not.

- Some sludges have high concentrations of water-soluble materials; others do not. ORNL tests on C-103 sludges showed that the sludge volume did not change before and after water washing, nor did the wash water contain significant amounts of radioactivity.

Another conclusion from the repeated wash cycles is that a full-scale process may not need to remove as much of a key element as would technically be possible. The removal requirements will be set by the constraints on the vitrification process, and removal above those constraints would represent unnecessary added expense.

These observations demonstrate the benefit of careful process monitoring during the ESW process. Cesium-137 activity and total conductance measurements have been done on water washes and caustic leaches, and the conductivity probe has been found to be a reliable indicator of the change in the mass of solids. This instrument is also used by the aluminum industry to monitor hot caustic leaches of bauxite. Table 4 and Figure 5 show the correlation between conductivity measurements and $^{137}Cs$ removal from Hanford tank S-101 sludge in tests conducted at ORNL.

It is important to note that a process does not require a full-scale demonstration to provide useful information to DOE sites. Sites have often commented that in many instances they currently need technical data for the design and testing of baseline unit operations more urgently than they need deployment of entirely new technologies. The chemistry of many DOE wastes is complex and poorly understood. The data generated to date, both of ESW process performance and of tank waste characteristics, provide a technical benchmark for a portion of the pretreatment required for high-level radioactive waste at Hanford in Phase II privatization and reduce the technical and programmatic risks associated with the privatization procurement decisions. The performance testing data also increased confidence that Phase I sludge fed to the privatization contractor could be effectively washed.

**Table 3. Summary of aluminum, chromium, and phosphorus removal from five sludges after dilute hydroxide wash and caustic leach**

| Tank | Temperature (°C) | Sodium hydroxide concentration (*M*) | Percentage removed[a] | | |
|---|---|---|---|---|---|
| | | | Al | Cr | P |
| B-101 | 60 | 1.1 | 56 | 50 | 87 |
| | | 3.2 | 63 | 51 | 92 |
| | 100 | 1 | 59 | 52 | 85 |
| | | 3.1 | 62 | 59 | 95 |
| BX-110 | 60 | 1.2 | 95 | 61 | 100 |
| | | 3.2 | 99 | 82 | 100 |
| | 80 | 1.2 | 98 | 80 | 100 |
| | | 3.1 | 97 | 91 | 100 |
| | 95 | 1.2 | 99 | 90 | 100 |
| | | 3.2 | 99 | 95 | 100 |
| BX-112 | 60 | 1.1 | 64 | 62 | 99 |
| | | 2.9 | 69 | 76 | 99 |
| | 80 | 1.3 | 56 | 88 | 99 |
| | | 3.4 | 65 | 89 | 99 |
| | 100 | 1.1 | 53 | 86 | 99 |
| | | 3.4 | 61 | 86 | 100 |
| C-102 | 60 | 1.1 | 27 | *b* | 60 |
| | | 2.9 | 95 | *b* | 66 |
| | 100 | 1 | 20 | *b* | 56 |
| | | 2.9 | 95 | *b* | 61 |
| S-101 | 70 | 1 | 70 | 74 | *c* |
| | | 3 | 63 | 86 | *c* |
| | 95 | 1 | 87 | 71 | *c* |
| | | 3 | 89 | 76 | *c* |

[a]From the dilute hydroxide-washed solids after leaching for 168 h (72 h for C-102).
[b]Chromium was below analytical detection limit in this sludge.
[c]No value reported because of low mass recovery for phosphorus.
*Source:* Lumetta et al. 1998.

**Table 4. Inhibited water washes of sludge from Hanford Tank S-101**

| Wash | Total cesium removal (%) | Conductance (1/mΩ) |
|---|---|---|
| 1 | 62.2 | 625 |
| 2 | 87.1 | 274 |
| 3 | 92.6 | 122 |
| 4 | 95 | 55 |
| 5 | 96.5 | 26 |
| 6 | 97.6 | 17 |

*Source:* Hunt, Collins, and Chase 1998.

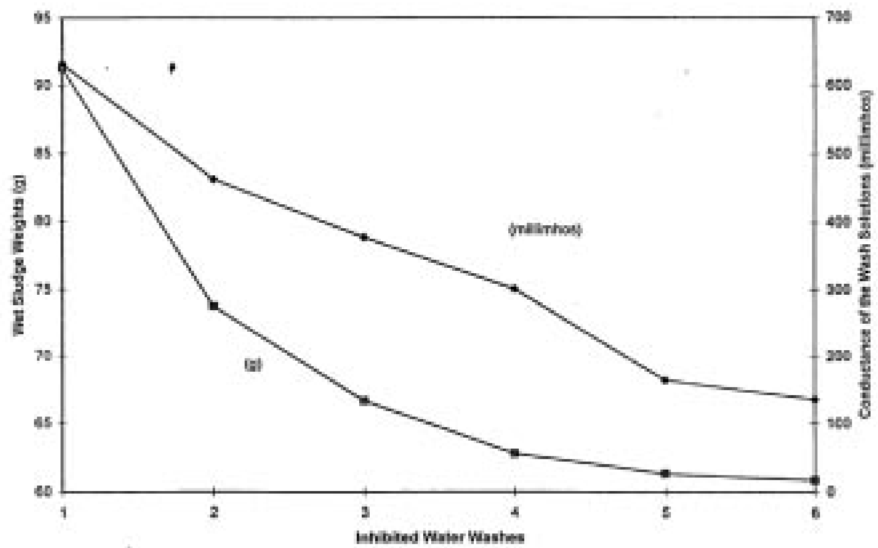


**Figure 5. Conductivity measurements and wet sludge weight with increasing number of inhibited water washes of Hanford Tank S-101 sludge.** *Source:* Hunt, Collins, and Chase 1998.

# SECTION 4
# TECHNOLOGY APPLICABILITY AND ALTERNATIVES

## Competing Technologies

At the Hanford Site, ESW is a baseline technology that allows unwanted, nonradioactive components to be removed from a solid HLW stream just before that waste stream is vitrified. Sites may opt for no sludge washing or simple sludge washing instead of enhanced sludge washing if the costs avoided from ESW are not sufficient.

There are several technologies that may be deployed in conjunction with ESW, including various process monitors and sensors, various chemical treatments to reduce gel formation or increase the solubility of chromium, and novel solid/liquid separation processes to increase the effectiveness of ESW. Advanced separations could be employed at the back end of the ESW process to further minimize the HLW volume.

## Technology Applicability

Of the 340 million liters of waste stored in underground tanks, approximately 75 vol % is the supernatant, salt cake, liquid portion of the slurry waste, sludge interstitial liquid, and calcine. The remaining volume consists of sludge and slurry solids. Table 5 shows the approximate volume of highly radioactive sludge stored at three DOE sites.

**Table 5. Estimated tank waste sludge volume at DOE sites**

| Site | Sludge (millions of liters) |
|---|---|
| Hanford | 45 |
| Savannah River Site | 45 |
| Oak Ridge | 0.8 |

Enhanced sludge washing may be applicable to HLW processed in the Federal Republic of Germany, France, Belgium, Canada, Finland, Japan, Spain, Sweden, Switzerland, and the United Kingdom. Much of this waste is spent fuel, which is currently stored at nuclear power plants or in storage pools. These storage pools can collect highly radioactive sludges on the bottom that might benefit from ESW prior to treatment and disposal.

The Hanford Spent Nuclear Fuels Program stores spent nuclear fuel in the 100-K Basins. Sludge has accumulated in the basins as a result of fuel oxidation and a slight amount of general debris being deposited by settling in the basin water. The sludge that collects at the bottom of these pools may be amenable to ESW.

The extensive bench-scale ESW testing to date has resulted in solid protocols for evaluating whether the process is applicable to these or other waste streams. However, before a full-scale process can be built to treat any of the above-mentioned waste streams, ESW will require further, larger-scale demonstration and testing. Some of the issues that will need to be addressed are how to most effectively mix sludges and wash waters on a large scale, how to most effectively separate the wash waters from the solids, and shielding requirements for a large-scale ESW process. However, these are issues that DOE and the international community have addressed many times for different projects and processes and should not be impediments to successful operation of a full-scale ESW process.

## Patents/Commercialization/Sponsor

This work has primarily been sponsored by the DOE Environmental Management Tanks Focus Area and by Hanford’s Office of River Protection (formerly Tank Waste Remediation System). No commercial partner is associated with this work, and no patents have resulted from it.

# SECTION 5
# COST

## Methodology

In 1993, ESW was selected as the baseline for sludge pretreatment at Hanford. Hanford's Tri-Party agreement milestone M-50-03-T2B required a complete evaluation of enhanced sludge washing to determine whether the process produced a reasonable volume of glass canisters. TFA studies helped Hanford meet Tri-Party Agreement milestone M-50-03-T2B and validate that the ESW process was an acceptable baseline for pretreatment of Hanford tank sludges.

Several studies estimate the potential cost avoidance included in the baseline from ESW for treating sludge from Hanford tanks. Cost estimates for ESW are continually updated as retrieval strategies change and better sludge washing data become available. This section is based on published Hanford waste volume estimates (Kirkbride et al. 1999) and available cost estimates from the following LANL documents:

DeMuth, S. R. 1996a. *Cost benefit analysis for enhanced sludge washing of underground storage tank high-level waste*. LA-UR-96-965. Los Alamos, N.M.: Los Alamos National Laboratory.

DeMuth, S. R., and D. Williams. 1997. *Cost-effectiveness of crystalline silicotitanate and resorcinol-formaldehyde ion exchange resins and enhanced sludge washing with and without chromium oxidation*. LA-UR-97-3903, Rev. 1. Los Alamos, N.M.: Los Alamos National Laboratory.

DeMuth, S. R., and A. Shieh. 1998. *Revised cost savings estimate for enhanced sludge washing of underground storage tank waste at Hanford*. LA-UR-98-3929, Rev. 1. Los Alamos, N.M.: Los Alamos National Laboratory.

DeMuth, S. R., and G. Thayer. 1999. *Updated cost savings estimate with uncertainty for enhanced sludge washing of underground storage tank waste at Hanford*. LA-UR-99-5464, Rev. 1. Los Alamos, N.M.: Los Alamos National Laboratory.

Each cost analysis has used a slightly different methodology, based on currently available needs and information available. A usual methodology for cost benefit analyses for OST-developed technologies is to compare the innovative technology to a baseline process. Since ESW has been part of the baseline since 1993, other comparisons have been needed. The DeMuth 1996a and DeMuth and Williams 1997 cost analyses compare ESW to simple water washing. This is essentially equivalent to retrieval with water only. DeMuth and Shieh 1998 and DeMuth and Thayer 1999 compare ESW to sludge washing on the basis of the advances enabled by the $30 million invested by OST. It was determined that 75–85% of the ESW performance currently achievable could have been accomplished without OST's development investment. Seventy-five to 85% ESW performance is approximately the same as the performance of simple sludge washing. Thus the two most recent studies compare the cost benefit from achieving 100% ESW performance, to 75% (1998 analysis) and 85% (1999 analysis) ESW performance.

The LANL studies cost information is derived from the Hanford Tank Waste Remediation System Environmental Impact Statement (Slaathaug 1995) and other documents. Table 6 shows assumptions for the unit operation remediation costs. While more recent cost estimates may have been developed, they have not been released to the public to avoid influencing privatization bids. Sludge wash factors for Hanford are continually being revised to update flow sheets for Phases I and II tank waste treatment (Kirkbride et al. 1999).

**Table 6. Unit operations costs from the Hanford Tank Waste Remediation System Environmental Impact Statement, in millions of dollars (1995 dollars)**

| Activity | Capital | Operating | Research and development | Total |
|---|---|---|---|---|
| Retrieval | 5,100 | 3,700 | | 8,800 |
| Liquid separations | 792 | 276 | 83 | 1,151 |
| Sludge wash | 69 | 129 | 9 | 207 |
| Low-activity waste immobilization | 2,228 | 624 | 264 | 3,116 |
| Low-activity waste disposal | 264 | 16 | 14 | 294 |
| High-level waste immobilization | 2,231 | 639 | 260 | 3,130 |
| High-level waste disposal | 5,858 | 31 | | 5,889 |
| Total | | | | 22,587 |

*Source:* DeMuth and Thayer 1999.

Other basic assumptions for the most recent cost analysis are as follows:

- The total costs associated with processing and disposal of high-level radioactive wastes using 100% ESW are compared with the total costs assuming a less efficient sludge washing (85%) is used instead. Since 100% ESW is the baseline for remediation at Hanford, the choice for level to which to compare the baseline is subjective.

- Approximately 57 million liters of Hanford tank waste was assumed to be amenable to volume reduction by ESW.

- The waste loading in HLW glass is 15% waste oxides.

- The remediation cost benefits occur over 30 years.

- The volume of HLW glass produced from sludge with ESW is consistent with the *Independent Review of Hanford HLW Volume* (Plodinec et al. 1996): 13,800–50,000 canisters with a median of 23,000 canisters.

The range of discrete remediation costs were converted to a normal distribution. An analysis of variance accounts for process and cost parameter uncertainties such as ESW separations factors, waste inventory, capital costs, and remediation costs.

## Cost Analysis

Figure 6 shows the flow of materials from Hanford tanks to a final disposal form with associated costs for the treatment process. The volume of HLW is significantly reduced by ESW. Because of the large cost for HLW vitrification and disposal, even a small reduction in the volume of HLW can have a major impact in total costs (DeMuth and Thayer 1999). Figure 7 shows the normal distribution curves for the 100% ESW and 85% ESW remediation cases. Total remediation costs in Figures 6 and 7 are different from Table 6 primarily because Table 6 assumes perfect, efficient waste blending scenarios. With perfect waste blending, the number of HLW canisters is estimated to be 12,000–13,000. However, imperfect blending is both more realistic and conservative and leads to the median estimate of 23,000 canisters. Revised Tank Waste Remediation System inventories, updated ESW performance data, and conversion from 1995 to 1999 dollars also contribute to the differences between these figures.

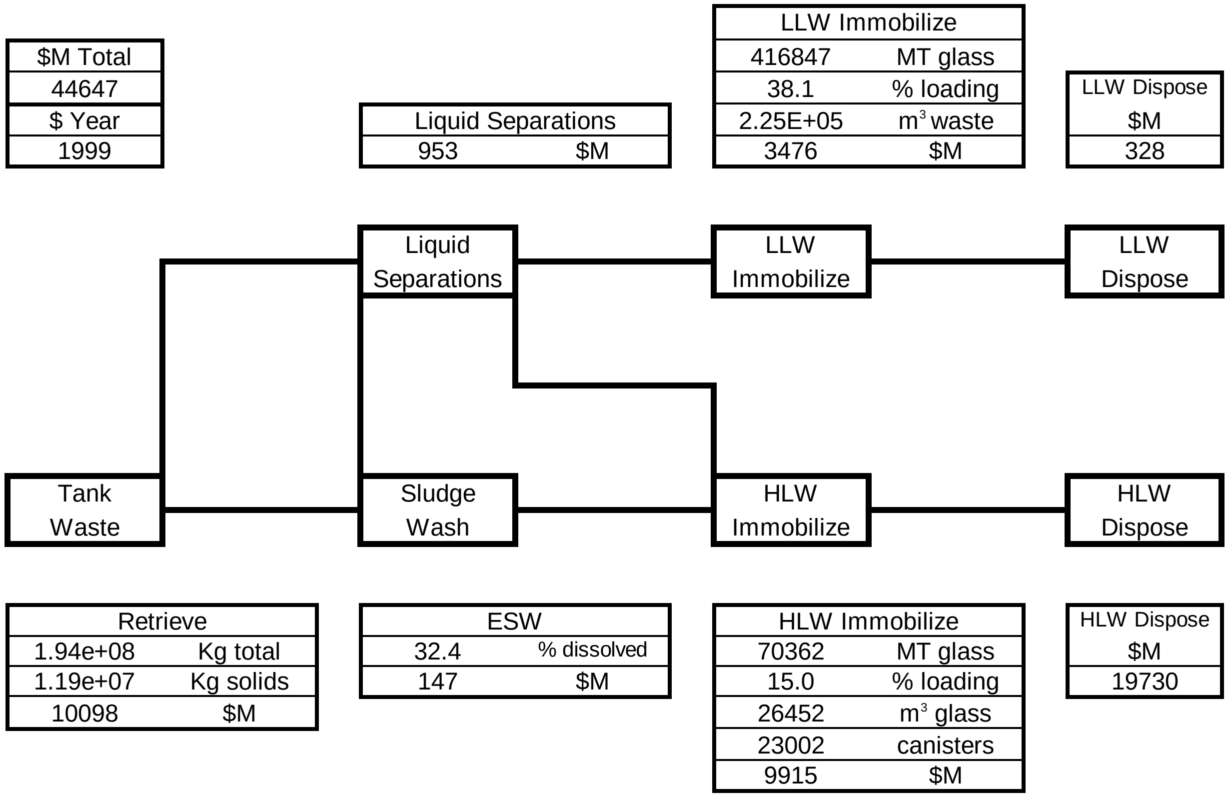


**Figure 6. Process model and costs for 100% ESW (1999 dollars).**
*Sources:* DeMuth and Thayer 1999; DeMuth, personal communication, 1999.

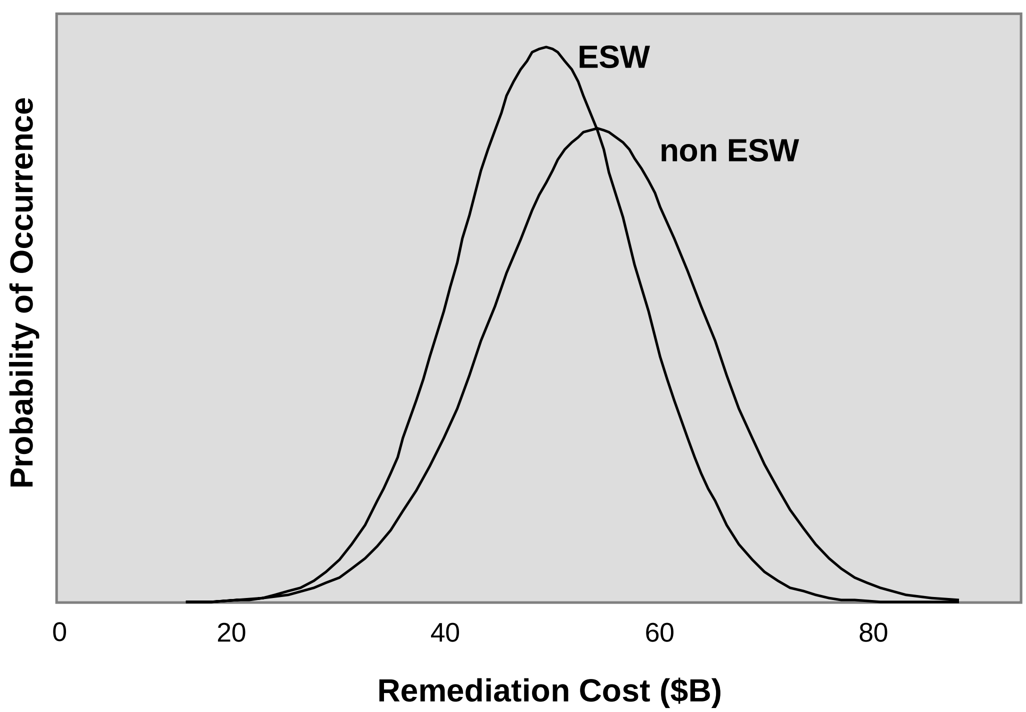


**Figure 7. Continuous distribution for remediation cost outcomes (1999 dollars).**
*Source:* DeMuth and Thayer 1999.

## Cost Conclusions

ESW impacts costs by reducing the final volume of HLW that is vitrified and sent for permanent disposal. DeMuth and Thayer 1999 provides a new cost estimate based upon recent waste inventory and ESW process performance revisions. The study includes an estimate of the associated cost uncertainty. An analysis of variance compares the two cost estimates in 1999 dollars. The revised study shows an approximate cost avoidance of $4.8 billion is included in the baseline treatment costs for all underground storage tank waste at Hanford, as shown in Figure 7.

# SECTION 6
# REGULATORY AND POLICY ISSUES

The use of any technology for environmental remediation and waste management is constrained by state, federal, and local regulations, which differ at each DOE site. State and local regulations can vary widely, despite some efforts by the U.S. Environmental Protection Agency (EPA) and states to encourage regulatory reciprocity (acceptance of testing from one state or region to another). The regulatory approval and permitting of ESW will likely be closely linked with the rest of the pretreatment process. As with all complex treatment processes, open lines of communication between regulators, stakeholders, and DOE sites facilitate efficient progress. No regulatory or permitting issues have been identified with ESW. It does not appear to be controversial in terms of public acceptance.

## Regulatory Considerations

Regulatory and permitting requirements for ESW are comparable to other waste processing technologies such as ion exchange and precipitation. It is anticipated that, because this process reduces the volume of HLW, it will meet with favorable regulatory consideration.

The data collected to date have already been used to satisfy a regulatory requirement at Hanford, meeting the Tri-Party Agreement milestone M-50-03-T2B to “project the impact of ESW on glass production at the Hanford Site” and to provide a basis for selecting processing and chemical conditions that reduce costs and reduce HLW volumes.

Currently, ESW data are needed to evaluate retrieval sequences and blending strategies to satisfy Tri-Party Agreement milestones M-45-02D through M-45-02I to minimize the volume of HLW to the extent that can be achieved by ESW alone. The retrieval sequence provides the foundation for preparation of the Phase II privatization contract. A significant cost avoidance is expected if DOE is armed with information that allows a more concise contract to be written and a realistic knowledge of Phase II costs with which to evaluate vendors’ proposals.

### Secondary Wastes

The secondary wastes generated from ESW will consist the wash solution. The wash solution increases the volume of radioactive liquids that must be treated to remove radionuclides and subsequently immobilized as LAW. Other wastes include personal protective equipment, contaminated equipment and hardware, plastic sheeting and sample containers, analytical solutions, piping materials, and miscellaneous hardware.

### CERCLA/RCRA Considerations

This technology is currently being considered for wastes regulated by the Resource Conservation and Recovery Act (RCRA). Hazardous and dangerous waste permit(s) will be required to operate treatment facilities. Treatment of wastes regulated by Comprehensive Environmental Response, Compensation, and Liability Act (CERCLA) may be considered at a later date. CERCLA considerations are discussed below.

### Human Health and Environment

Overall protection of human health and the environment is high. ESW minimizes the amount of HLW resulting from waste treatment.

### Compliance with Applicable or Relevant and Appropriate Requirements (ARARs)

Compliance with ARARs is required when the waste is disposed of on site. Vitrified HLW sludge will be sent to an off-site repository for disposal. If CERCLA waste has been immobilized, the off-site disposal facilities must be qualified to accept waste from a CERCLA site.

### Long-Term Effectiveness and Permanence

The vitrification process for HLW sludge produces a very durable, homogeneous waste glass. The waste glass is expected to be stable over a period of time beginning at a few tens of thousand years to several hundred thousand years. This is important since unwanted leaching and migration of radioactive waste from the waste glass could pose risk to future generations.

**Reduction of Volume, Mobility/Toxicity**
The HLW fractions produced from ESW processes are of a much smaller volume and are more stable than if no treatment were conducted. Therefore, reductions of toxicity, mobility, and volume of HLW are more effective compared to doing no pretreatment or an alternative type of treatment.

**Implementability**
Full-scale implementation is not complex. The remote-handling designs and procedures already exist, all equipment and reagents are commercially available, people are currently trained in this process, and regulatory permits can easily be obtained compared to other technologies.

**Costs**
Costs can be avoided by reducing the volume of HLW to be vitrified.

**State and Community Acceptance**
State and community acceptance is addressed as part of the total remedial action. The ESW technology improves acceptance for the remedial action since the technology reduces the volume of HLW to be vitrified.

## Safety, Risks, Benefits, and Community Reaction

Because of the decreased risk associated with decreased volumes of HLW, ESW complies with "as-low-as-reasonably-achievable" (ALARA) principles. The normal procedures for working with radioactive material are applicable, and the staff would be trained in operation of the system, whether that system is a bench-scale system in a hot cell or a full-scale system.

**Worker Safety**
Radiological exposure of personnel must be kept ALARA pursuant to DOE regulations. ESW does not expose workers to hazardous or radioactive materials because all laboratory-scale work is done in a fume hood or shielded glovebox. Larger-scale processes will have appropriate levels of automation, containment, and shielding on the equipment to reduce risks from exposure pathways such as dermal contact or inhalation.

**Community Safety**
There is no history of accidents with this technology. Future scale-up processes would be required to comply with safety policies and guidelines of DOE, EPA, and other applicable regulatory agencies.

**Environmental Impact**
Protection of human health and the environment is relatively high with ESW because it is one step in getting stored wastes out of underground storage tanks. The process results in reduced volumes of HLW requiring space in a repository, and the low-activity effluent stream from the process will be much less hazardous than the feed material. There is no routine release of contaminants with ESW.

**Socioeconomic Impacts and Community Reaction**
Community reaction to ESW is difficult to assess at this time; however, the public has firmly stated that it is interested in treating underground storage tank waste as quickly and efficiently as possible. Additional risks, safety concerns, community concerns, and socioeconomic impacts pertaining to the pretreatment process in general, and ESW specifically, will be addressed as the full-scale processes are designed and built.

**Benefits**
Waste disposals costs are reduced because of the smaller volume of HLW after ESW. Waste handling costs are reduced because the HLW fraction will be much smaller after ESW than it would be without ESW.

# SECTION 7
# LESSONS LEARNED

## Implementation Considerations

The most important consideration for implementation of ESW is that each sludge may behave differently; each may require a longer or shorter leaching period, a different NaOH concentration, or a different temperature for optimal component removal. The process will need to be developed carefully for each sludge, considering the requirements for the final glassmaking process downstream. The ESW process may require careful process monitoring.

The ESW process encompasses a trade-off: the total volume of LAW is increased, but the volume of HLW is decreased. This is a logical decision given the relative disposal costs for low-activity and high-level wastes. The final disposition of the waste drives many of the decisions for implementing ESW.

## Technology Limitations and Needs for Future Development

Process monitoring and control technology should be carefully evaluated as the ESW process is tested at larger scales. The conductivity probes used successfully in the bench-scale processes are similar to those used by the aluminum industry. Sensors for other key components may require further development.

If it is determined that chromium removal with the ESW process alone does not meet downstream process requirements, chromium oxidation technology to convert the less soluble Cr(III) to Cr(VI) will need further evaluation and testing on a larger scale.

## Technology Selection Considerations

Sites have already selected ESW as the baseline. Many options exist for its implementation. The main technology selection considerations are

- downstream process feed requirements,
- performance of the process on the element requiring removal, and
- the need for more processing to prevent gels or further reduce the volume of HLW.

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# APPENDIX B
# ACRONYMS AND ABBREVIATIONS

| | |
|---|---|
| ALARA | as low as reasonably achievable |
| ARAR | applicable or relevant and appropriate requirements |
| CERCLA | Comprehensive Environmental Response, Compensation, and Liability Act |
| DOE | U.S. Department of Energy |
| EIS | environmental impact statement |
| EPA | Environmental Protection Agency |
| ESW | enhanced sludge washing |
| FY | fiscal year |
| HLW | high-level waste |
| LANL | Los Alamos National Laboratory |
| LAW | low-activity waste |
| *M* | molar |
| NaOH | sodium hydroxide |
| ORNL | Oak Ridge National Laboratory |
| OST | Office of Science and Technology |
| PNNL | Pacific Northwest National Laboratory |
| RCRA | Resource Conservation and Recovery Act |
| SRS | Savannah River Site |
| TFA | Tanks Focus Area |
| TMS | Technology Management System |
| TWRS | Tank Waste Remediation System |