

Treatment and Disposal of High-Level Radioactive Waste at the Hanford Site: The Technical Challenge

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**TREATMENT AND DISPOSAL OF HIGH-LEVEL RADIOACTIVE WASTE
AT THE HANFORD SITE: THE TECHNICAL CHALLENGE**

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1.0 INTRODUCTION

The U.S. Department of Energy's (DOE) Hanford Site, located in southeastern Washington State, has the most diverse and largest amount of radioactive tank waste in the United States. A Tank Waste Remediation System (TWRS) Program was established in 1991 to safely store, treat, and dispose of those wastes. This paper describes the technical challenge in conducting the TWRS Program that will take more than 30 years and cost tens of billions of dollars to complete.

2.0 BACKGROUND

High-level waste (HLW) has been stored at the Hanford Site in large underground storage tanks since 1944. Approximately 225,000 m³ (59 Mgal) of waste have accumulated in 177 tanks. These caustic wastes consist of different chemicals and include liquids, slurries, saltcakes, and sludges.

The radioactive waste stored in these tanks came from different sources. These sources include (1) three plutonium and uranium recovery processes from approximately 100,000 Mtu of irradiated fuel, (2) three radionuclide recovery processes from waste, and (3) miscellaneous sources (e.g., laboratories and reactor decontamination solutions). These wastes were then concentrated and mixed to minimize the number of storage tanks required. The neutralized wastes include sodium nitrate/nitrite; sodium hydroxide; sodium aluminate; sodium phosphate; the hydrous oxides of iron, chrome, and other transition metals; large amounts of organics; and approximately 250 MCi of radionuclides.

The wastes are stored in 149 single-shell tanks (SST) and 28 double-shell tanks (DST). The SSTs are made of reinforced concrete with a carbon-steel liner and can hold 208 m³ (55,000 gal) to 3,800 m³ (1 Mgal) of radioactive waste. Of the older SSTs, 67 have leaked or are suspected to have leaked approximately 3,800 m³ (1 Mgal). No waste has been added to the SSTs since 1980. The pumpable liquids are being removed from the SSTs so that the remaining waste is mostly sludge and saltcake. The DSTs are made with a carbon-steel tank within a steel-lined concrete tank. The DSTs have a

nominal capacity of 3,800 m³ (1 Mgal), as shown in Figure 1. There is no evidence to suspect that any of the newer DSTs, the first placed in service in 1971, have leaked (Hanlon 1994).

In addition to the waste stored in the tanks, significant amounts of ⁹⁰Sr and ¹³⁷Cs were removed from the tank waste, converted to salts, doubly encapsulated in metal containers, and stored in water basins (Figure 2). There are approximately 1,900, 6.7 cm (2.6 in.) dia x 52 cm (20.5 in.) long capsules containing approximately 160 MCi.

3.0 TWRS TREATMENT AND DISPOSAL STRATEGY

A technical strategy for treating and disposing of the Hanford Site tank waste has been developed to achieve the objectives listed below. This technical strategy is shown in Figure 3.

1. Characterize the waste to provide the data necessary to retrieve, pretreat, immobilize, and dispose of the waste.
2. Retrieve the waste from all DSTs and SSTs to the extent necessary for closure.
3. Separate the waste into HLW/transuranic (TRU)^a waste and low-level waste (LLW) fractions, so that most of the radionuclides and only a small part of the other waste materials are in the HLW and the remainder is in the LLW.
4. Remove radionuclides from the waste stream destined to become LLW to the extent needed to:
 - Meet the U.S. Nuclear Regulatory Commission's (NRC) "incidental waste" classification (58 FR 12345)
 - Meet DOE's LLW disposal requirements, which include an as low as reasonably achievable (ALARA) policy and an acceptable disposal system performance assessment
 - Be less than the comparable limits for commercial LLW (10 CFR 61 class C waste)
 - Permit construction of a lightly shielded LLW vitrification facility, if practical.
5. Retrieve the SST waste by 2018, close all SSTs by 2024, and complete immobilization of all waste by 2028.

^aAs used in this document, HLW includes TRU waste.

Figure 1. Double-Shell Tank.

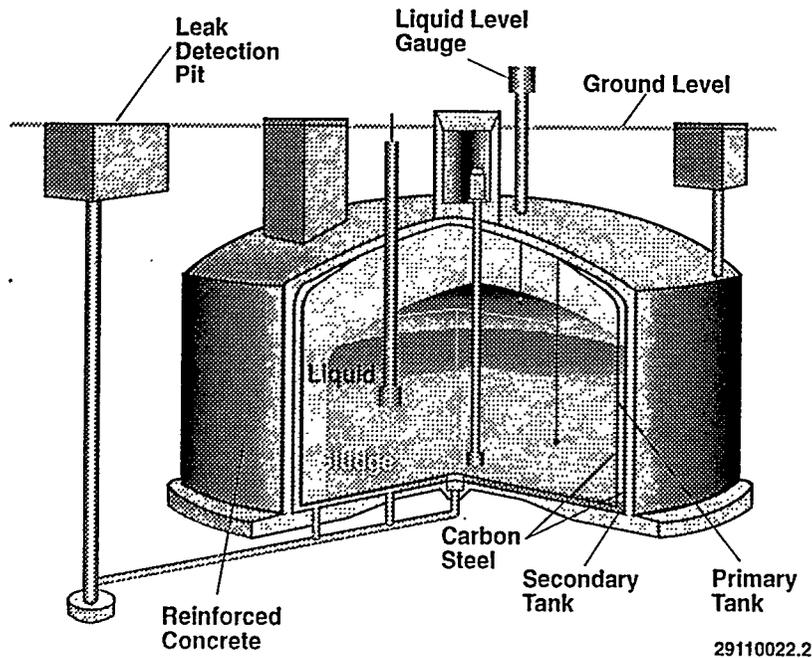


Figure 2. Strontium and Cesium Capsules.

- Cesium and Strontium Encapsulated 1974 - 1975
- Capsules are 6.6 cm dia. x 52.1 cm long (2.6 in. dia. x 20.5 in. long)
- Double Encapsulated
- 1338 Cesium Capsules Contain:
 - Cesium Chloride
 - ¹³⁷Cesium ^{137m}Barium 400×10^{16} Bq (110MCI)
- 610 Strontium Capsules Contain:
 - Strontium Fluoride
 - ⁹⁰Strontium ⁹⁰Yttrium 185×10^{16} Bq (50MCI)

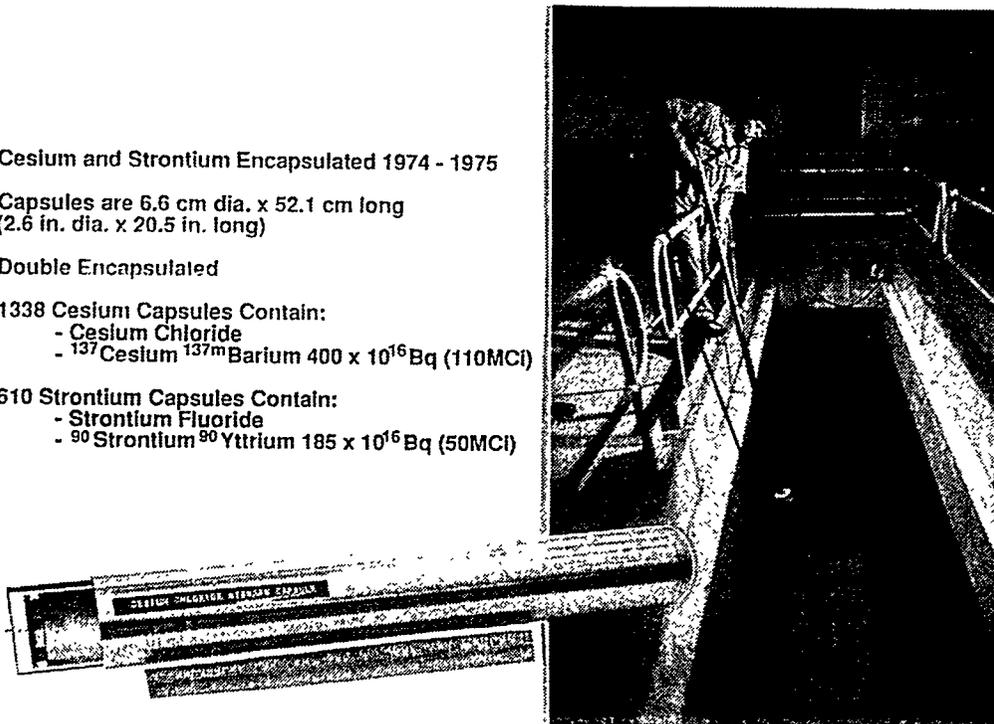
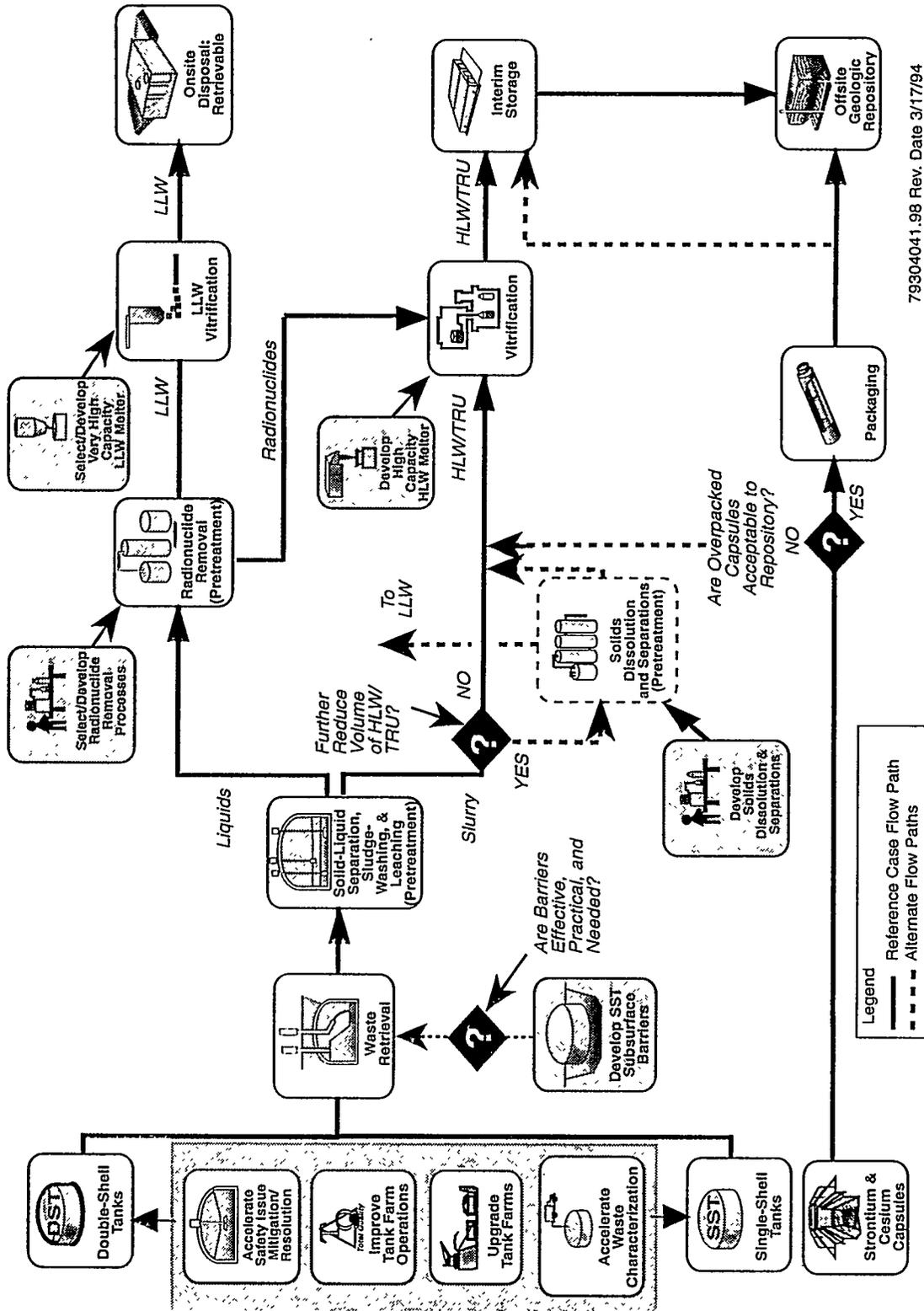


Figure 3. Hanford Tank Waste Remediation System Strategy.



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6. Reduce the HLW stream volume so that the immobilized volume is acceptable to the Geologic Repository Program and is cost effective.
7. Vitrify the LLW and dispose of it onsite, near-surface, and in a retrievable form that conforms to DOE disposal requirements.
8. Vitrify the HLW and place it in canisters that meet the Geologic Repository Program waste acceptance criteria; store the waste canisters until they can be shipped to a geologic repository for disposal.
9. Store the strontium and cesium capsules until they can be packaged and shipped to a geologic repository for disposal. Package (or process) the capsules in a manner that conforms to the Geologic Repository Program requirements.

3.1 TECHNICAL CHALLENGES IN IMPLEMENTING STRATEGY

The size and scope of the TWRS Program provides many technical challenges, and demands that significant effort be given to finding ways to reduce cost while doing the work safely. The major TWRS Program activities and technical challenges are described in this section.

3.1.1 Waste Characterization

There is inadequate information on the chemical, physical and radiological makeup of the tank waste to develop process flowsheets and design equipment to retrieve, pretreat, and immobilize the waste. Information on the process waste streams routed to the storage tanks was quite good, but over the past 50 years, the wastes have been further processed, mixed, and aged such that knowledge about the waste in each tank is limited. A Data Quality Objectives process is being used to identify waste characterization data needs. Additional sampling equipment and analytical facilities are being built. However, problems are being encountered in taking complete representative samples and sampling and analysis is very costly.

The technical challenge is to determine the minimum amount of waste characterization needed and then to do it better, faster, and at lower cost.

3.1.2 Waste Retrieval

Waste will be retrieved from the tanks for treatment, immobilization, and disposal. Hydraulic sluicing has been the method used to retrieve waste from underground radioactive waste tanks at the Hanford Site and elsewhere. While sluicing is the preferred method of waste retrieval, it may not be acceptable in SSTs that leak (or it may not remove some of the hard sludges). Most of the SST waste is saltcake or sludge and will require water dilution of approximately 3:1 to sluice it from the tanks. Mechanical waste retrieval

would be difficult because operations must be conducted with remotely operated equipment. Subsurface barriers and other methods are being investigated for confining tank leakage, and a robotic arm-based retrieval system is being developed.

The technical challenge is to efficiently remove this large volume of waste without further contaminating the surrounding soil from leaks or spills.

3.1.3 Waste Pretreatment

The waste retrieved from the tanks will be separated into two fractions so that most of the radionuclides, and only a small part of the waste volume, are in the HLW fraction. The HLW will be vitrified and shipped offsite for disposal in a geologic repository. Most chemicals, and only a few radionuclides, would be in the LLW stream which will be vitrified and disposed near the surface onsite. The strategy is to use proven separations technology (including solids-liquid separation, sludge washing, and ion exchange) to the extent possible. More advanced separations technologies such as sludge dissolution and solvent extraction of several radionuclides will be developed but only implemented, if needed, to achieve the required level of radionuclide removal or an acceptably small enough volume of HLW.

The amount of radioactive waste to be pretreated is very large. Approximately 95 percent is expected to be soluble in caustic solutions and, after removal of ^{137}Cs (and possibly other radionuclides such as ^{90}Sr), to become the LLW stream. The preliminary flow sheet shows 630,000 metric tons of waste solution will be fed to this ^{137}Cs ion exchange columns where an additional 230,000 metric tons of chemicals and 430,000 tons of water will be added. Large-scale evaporators will be used between unit operations to reduce the waste volume by evaporating the water for reuse.

If more advanced separations technologies are needed to reduce the volume of LLW, the process becomes more complex. The sludges must first be dissolved in nitric acid and other more aggressive chemicals. Then the acidic solution must be processed through solvent extraction and other unit operations to separate ^{137}Cs , ^{90}Sr , and TRUs (and possibly other radionuclides) so that the remaining chemical solution can be routed to the LLW stream. These processes are not yet fully developed, and the aggressive chemical solutions require highly corrosion-resistant materials, thereby increasing equipment costs and the potential for failure from corrosion.

The technical challenge is to adequately reduce the HLW stream volume with alkaline side sludge washing and leaching methods so that advanced separations technologies are not needed.

3.1.4 Low-Level Waste Immobilization

The LLW will be vitrified and disposed near-surface, onsite in a retrievable form. The waste form must accommodate the waste feed characteristics and the disposal system performance assessment requirements. The LLW Vitrification Facility will be lightly shielded if a practical balance of pretreatment and LLW vitrification costs can be achieved. Chemicals

vaporized during vitrification will be captured and/or treated to meet effluent release limits. The vitrified LLW will be placed in an onsite near-surface disposal system in a manner that will allow retrieval for placement elsewhere, should that become necessary. The Low-Level Waste Vitrification Facility capacity will be at least 100 tons per day of glass. It is currently estimated that 570,000 metric tons of LLW vitrified product will be disposed in onsite vaults.

Vitrifying high-sodium content radioactive waste at the high capacity needed has not been done. Commercial glass melter technology holds promise that it can be utilized. However, a testing program with the commercial glass industry is just being initiated to determine if their technology and equipment can be adapted to radioactive service. The off-gas system will have to handle large volumes of volatile chemicals and some volatile radionuclides.

The technical challenge is to provide a high-capacity vitrification system that can be operated and maintained in radioactive service.

3.1.5 High-Level Waste Immobilization

Vitrification as borosilicate glass is generally accepted as the method to immobilize HLW. The TWRS strategy is to provide a high-level vitrification capacity that will be able to vitrify all the HLW in 20 years. This will require a melter capacity of approximately 15 tons per day.

Several melter technologies are being considered. A joule-heated liquid-fed ceramic melter is the current state-of-the-art radioactive waste melter in the United States and in some foreign countries. However, the current capacity is approximately 2-1/2 tons per day -- far less than the 15 tons per day needed. Increasing the melter capacity by stirring the melt and increasing the temperature is being tested. Using multiple melters is also an option.

The technical challenge is to provide a highly reliable, high-capacity vitrification system that will consistently encapsulate the waste in high-quality glass.

The waste canister capacity and configuration will also be optimized considering such factors as the vitrification plant, interim storage and the geologic repository. A larger HLW package (up to a 10 m³ canister) may reduce the cost of disposal at the repository.

3.1.6 Interim HLW Storage

HLW containers will require onsite storage for many years until a geologic repository is ready to accept them. A storage facility with capacity to store all of the vitrified waste will be provided as needed.

The ^{137}Cs and ^{90}Sr will be stored in capsules that will remain onsite until they can be packaged and shipped to the geologic repository for disposal. Overpacking multiple capsules in a canister is the reference plan. If the overpacked capsules do not meet repository acceptance criteria, the ^{137}Cs and ^{90}Sr capsules will have to be processed and vitrified with other HLW.

The technical challenge is to develop an overpack that will contain the cesium chloride and strontium fluoride salts in a manner that will meet the Repository Waste Acceptance Criteria.

3.2 SCHEDULE

Treatment and disposal of the tank waste is included in the *Hanford Federal Facility Agreement and Consent Order* (Tri-Party Agreement). This agreement between the DOE, U.S. Environmental Protection Agency (EPA), and the Washington State Department of Ecology (Ecology), establishes legally enforceable milestones for environmental cleanup actions at the Hanford Site (Ecology et al. 1994). The current TWRS strategy, developed over the past three years with significant public involvement, led to an amendment to the Tri-Party Agreement that was approved in January 1994. Major Tri-Party Agreement milestones for the TWRS Program are listed in Table 1. Note that the milestones call for (1) completing retrieval of all SST waste by 2018, (2) closing all SST farms by 2024, and (3) vitrifying all waste by 2028. Near-term milestones include: (1) initiating retrieval of SST 241-C-106 waste by October 1997, (2) determining whether advanced separations processes are required by March 1998, (3) selecting a LLW reference melter by June 1996, and (4) selecting a HLW reference melter by September 1998.

4.0 CONCLUSION

The Hanford Site TWRS Program is a large, complex program that will require many years to complete. Acquiring the commitment and funding to conduct this program will require a national consensus that this work is necessary and is being done in a cost-effective manner. Therefore, it is imperative to work safely, protect the public, seek the best technology, reduce costs, and use national expertise in planning and conducting this TWRS Program.

Table 1. Tri-Party Agreement Major Milestones for TWRS Program.

Milestone title	Scheduled completion date
Mitigate/resolve tank safety issues	09/2001
Complete single-shell tank (SST) interim stabilization	09/2000
Provide additional double-shell waste tanks	12/1998
Complete double-shell tank (DST) space evaluation	09/1994 (Annually thereafter)
Complete tank farm upgrades	06/2005
Complete tank waste characterization <ul style="list-style-type: none"> • Issue tank characterization reports for all SSTs and DSTs (177) 	09/1999
Complete closure of all SST farms <ul style="list-style-type: none"> • Develop SST waste retrieval technology • Complete evaluation and testing of subsurface barriers • Initiate full-scale demonstration of waste retrieval • Initiate tank waste retrieval from one SST • Complete waste retrieval from all SSTs 	09/2024 09/1994 09/1997 10/1997 12/2003 09/2018
Complete pretreatment process of Hanford Site waste <ul style="list-style-type: none"> • Start construction of LLW pretreatment facility • Start hot operations of LLW pretreatment facility • Start hot operation of HLW pretreatment facility 	12/2028 11/1998 12/2004 06/2008
Complete vitrification of Hanford Site HLW <ul style="list-style-type: none"> • Initiate construction of HLW vitrification facility • Initiate hot operations of HLW vitrification facility 	12/2028 06/2002 12/2009
Complete vitrification of Hanford Site low-level tank waste <ul style="list-style-type: none"> • Select reference melter • Initiate construction of LLW vitrification facility • Initiate hot operations of LLW vitrification facility 	12/2028 06/1996 12/1997 06/2005

NOTE: See the Tri-Party Agreement Part 1, Amendment 4 for a description of these milestones and the additional subtier milestones.

5.0 REFERENCE

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