Disposal of Hanford Site Tank Waste

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DISPOSAL OF HANFORD SITE TANK WASTES

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INTRODUCTION

Between 1943 and 1986, 149 single-shell tanks (SSTs) and 28 double-shell tanks (DSTs) were built and used to store radioactive wastes generated during reprocessing of irradiated uranium metal fuel elements at the U.S. Department of Energy (DOE) Hanford Site in Southeastern Washington state. The 149 SSTs (Figure 1), located in 12 separate areas (tank farms) in the 200 East and 200 West areas, currently contain about $1.4 \times 10^5 \text{ m}^3$ of solid (predominantly) and liquid wastes. Wastes in the SSTs contain about $5.7 \times 10^{18} \text{ Bq}$ (170 MCi) of various radionuclides including ^{90}Sr , ^{99}Tc , ^{137}Cs , and transuranium (TRU) elements. The 28 DSTs (Figure 2) also located in the 200 East and West areas contain about $9 \times 10^4 \text{ m}^3$ of liquid (mainly) and solid wastes; approximately $4 \times 10^{18} \text{ Bq}$ (90 MCi) of radionuclides are stored in the DSTs.

Important characteristics and features of the various types of SST and DST wastes are described in this paper. However, the principal focus of this paper is on the evolving strategy for final disposal of both the SST and DST wastes. Also provided is a chronology which lists key events and dates in the development of strategies for disposal of Hanford Site tank wastes. One of these strategies involves pretreatment of retrieved tank wastes to separate them into a small volume of high-level radioactive waste requiring, after vitrification, disposal in a deep geologic repository and a large volume of low-level radioactive waste which can be safely disposed of in near-surface facilities at the Hanford Site. The last section of this paper lists and describes some of the pretreatment procedures and processes being considered for removal of important radionuclides from retrieved tank wastes.

WASTE TANK SYSTEMS AND CONTENTS

Double-Shell Tank Wastes

The DSTs (tank-within-a-tank) were constructed from 1970 to 1985; all of the DSTs are designed to contain 3,800 m^3 of waste.

All the DSTs are constructed of mild steel (high carbon content). The sides and

bottoms of all tanks are supported by concrete structures. Openings in the unsupported tank domes allow limited access for sampling wastes and for measuring waste temperatures and liquid levels. An extensive network of buried piping is provided for the transfer of liquid wastes and waste slurries within and between tank farms.

The total present and expected future inventory of wastes in the existing 28 DSTs is classified into five types:

- Neutralized current acid waste (NCAW)--5,300 m³
- Neutralized cladding removal waste (NCRW)--3,300 m³ of sludge
- Plutonium Finishing Plant (PFP) waste--970 m³ of sludge
- Complexant concentrate (CC) waste--18,200 m³
- Double-shell slurry (DSS), double-shell slurry feed (DSSF), and dilute noncomplexed (DN) wastes--75,700 m³ (including the future addition of 35,300 m³ DN waste to be evaporated to 5,300 m³ DSSF)

All the DST wastes consist of a liquid portion and a solid portion. The NCAW is the waste that was produced when concentrated acidic plutonium-uranium extraction (PUREX) process high-level radioactive waste generated between 1983 and 1988 was made alkaline and stored in DSTs. An ammonium fluoride-ammonium nitrate solution was used in the PUREX plant during the 1983 to 1988 period to dissolve Zircaloy cladding from N Reactor fuel. The NCRW resulted when the spent cladding waste was made alkaline and stored in DSTs. The PFP waste resulted when composite acidic waste from the PFP was made alkaline and stored in a DST. The CC waste has a very high concentration of organic chelating agents and their degradation products. The CC waste is the concentrated aqueous raffinate from ⁹⁰Sr liquid-liquid extraction operations performed in the 1960's and 1970's. The DSS and DSSF wastes are viscous, highly-alkaline liquid wastes containing high concentrations of sodium salts generated from evaporation of dilute low level waste solutions. The DSS waste differs from DSSF waste in that it has been evaporated past the aluminate phase boundary and does not normally separate into sludge and supernatant liquids.

The solid portion of the NCAW, NCRW, CC waste, and PFP waste all contain > 100 nCi/g of TRU elements. The NCAW solids also contain the majority of the ⁹⁰Sr present in the PUREX process high-level radioactive waste; other DST solid wastes do not contain large concentrations of ⁹⁰Sr. The NCAW and CC alkaline waste solutions contain relatively high concentrations of ¹³⁷Cs. Because of the large amounts of organic chelators, some alkaline CC liquid waste also contains > 100 nCi/g of TRU elements.

Single-Shell Tank Wastes

Wastes in the SSTs consist mainly of two types of solids: sludge and salt cake. A small amount (2,300 m³) of interstitial liquid is also present. Sludges consist principally of metal (e.g., iron, chromium, nickel, aluminum, etc.) oxides and hydroxides. These precipitated when the acidic liquid wastes from the bismuth phosphate (BiPO₄), reduction-oxidation (REDOX), and PUREX processes were made alkaline before routing to the SSTs. Salt cake is mainly composed of water-soluble sodium salts (e.g., NaNO₃, Na₂CO₃, NaNO₂, NaOH, etc.) that crystallized when the original highly alkaline liquid wastes were evaporated. Of the SST radionuclide inventory, over 99 percent of the uranium, plutonium, and other TRU elements, ⁹⁰Sr, and some of the ⁹⁹Tc are in the sludge, while

the salt cake contains over 90 percent of the ^{137}Cs and the rest of the ^{99}Tc .

Twenty four of the SSTs contain so-called ferrocyanide wastes. These wastes were generated in the late 1950s when aged BiPO_4 process wastes were hydraulically retrieved and processed to recover large amounts of valuable uranium. Sodium ferrocyanide and nickel sulfate solutions were added to the aqueous raffinate from the Metal (Uranium) Recovery Process to precipitate nickel ferrocyanide at pH 9-10. The nickel ferrocyanide precipitate carried down with it almost all the ^{137}Cs in the waste. The supernatant liquid remaining after settling of the nickel ferrocyanide and hydrated metal oxides met all then existing requirements for disposal to the ground. Nickel ferrocyanide solids in the 24 SSTs are of particular interest and concern because of their potential, under certain conditions, of reacting vigorously with dried nitrate and/or nitrite salts.

EVOLUTION OF STRATEGIES FOR DISPOSAL OF HANFORD TANK WASTES

The earliest studies to define and develop an acceptable strategy for disposal of Hanford Site tank wastes began in 1972 (Table 1). The early and preliminary studies were documented in 1977 with issuance of the first technical report (ERDA, 1977) on selection and evaluation of technical alternatives for long-term management of Hanford Site tank wastes. Additional engineering studies, supplemented by on-going bench-scale tests of waste treatment and disposal form technology, were performed in the period 1979-1985. The first phase (1972-1988) of the efforts to develop a tank waste disposal strategy concluded in 1988 with the issuance of a Record of Decision on the Final Environmental Impact Statement, Disposal of Hanford Defense High-Level, Transuranic, and Tank Waste (DOE, 1988).

Table 1. Significant Events In Evolving Strategy for Disposal of Hanford Site Tank Waste: 1972-1988.

Date	Event	Reference
1972 - 1977	Preliminary engineering studies and evaluations; preliminary bench-scale experiments with simulated and actual tank wastes	
1977	First technical report on alternatives for long-term management of Hanford Site high-level radioactive waste	ERDA, 1977
1980	Follow-on reports on alternatives for disposal of Hanford Site high-level wastes	RHO, 1980a RHO, 1980b RHO, 1980c
1983	Definitive engineering study on disposal of DST wastes	Schulz et al., 1983
1985	Hanford Defense Waste Disposal Alternatives Engineering Support Data for the Hanford Defense Waste Environmental Impact Statement	RHO, 1985
1987	Final Environmental Impact Statement, Disposal of Hanford Defense High-Level, Transuranic, and Tank Wastes, Hanford Site, Richland, Washington	DOE, 1987
1988	Record of decision on final environmental impact statement	DOE, 1988

DOE = U.S. Department of Energy

DST = Double-shell tank

The 1988 Record of Decision stated that:

- The DST wastes would be retrieved and separated into two fractions, with the high-level fraction vitrified and disposed of in a deep geologic repository and the low-level fraction grouted in near-surface facilities. It was generally accepted that any pretreatment of DST waste to separate it into high- and low-level waste fractions would be done in the Hanford B Plant, a facility constructed during Manhattan Project days in the early 1940s.
- A final decision on disposal of SST wastes would not be made until additional development and evaluation studies were completed.
- Capsules of $^{90}\text{SrF}_2$ and $^{137}\text{CsCl}$ produced in the 1960-1985 time frame from selected retrieved wastes would be disposed of in a deep geologic repository.

Figure 3 illustrates the 1988 version of the Hanford Site plan for disposal of tank wastes and stored capsules. This plan envisioned that some SST wastes, perhaps most, would be left in place for disposal and that some SST waste might have to be retrieved. The 1988 disposal plan called for disposal of cesium and strontium capsules directly in a deep geologic repository after, perhaps, repackaging them in canisters suitable for geologic disposal.

Events (Table 2) which occurred in 1989 immediately after the Record of Decision was issued supported the 1988 tank waste disposal strategy. Thus, in 1989 the U.S. Department of Energy, U.S. Environmental Protection Agency, and the Washington State Department of Ecology co-signed the Hanford Federal Facility Agreement and Consent Order (Tri-Party Agreement)(Ecology et al., 1992). This Tri-Party Agreement established a strategy, schedule, and milestones for disposal of all the tank wastes. Also, engineering studies in 1989 and 1990 provided updated assessments of proposed DST pretreatment technologies and facilities.

Several recent events which occurred in 1991 combined to drastically alter the 1988 plans for disposal of tank wastes. For example, the Washington State Department of Ecology indicated in 1991 that the 40-year old B Plant did not meet minimum licensing and permitting regulations for extended use as a central facility for pretreating DST wastes. Also, in 1991, 6 DSTs and 48 SSTs, including the 24 SSTs containing ferrocyanide wastes, were found to have unreviewed safety questions requiring high priority mitigation and remediation. These events prompted a comprehensive engineering study to redefine the baseline plan for disposal of tank wastes; results of this study were published in late 1991.

Table 2. Significant Events in Evolving Strategy for Disposal of Hanford Tank Wastes: 1989-1993.

Date	Event	Reference
1989	Hanford Federal Facility Agreement and Consent Order signed by DOE, EPA, and Ecology	Ecology et al., 1990
1989	Updated assessment of processes and facilities for pretreating DST waste	Kupfer et al., 1989
1989	Further updated assessment of DST waste pretreatment alternatives	WHC, 1990
1991	Washington state Department of Ecology advises that B Plant does not meet minimum licensing and permitting criteria	Wodrich, 1992
1991	6 DSTs and 48 SSTs found to have unreviewed safety questions requiring high priority mitigation and remediation	Wodrich, 1992
1991	Tank Waste Disposal Program Redefinition	Grygiel et al., 1991
1992	DOE announces decision to proceed with NEPA documentation for retrieval of all SST wastes for pretreatment and disposal	
1992	Secretary of Energy provides initial set of goals and objectives for the newly-established Tank Waste Remediation System	Wodrich, 1992
1993	Tank Waste Technical Options report discusses technical details and projected costs for various options for disposal of tank wastes	Boomer et al., 1993
1993	Tank Waste Decision Analysis Report establishes current strategy for disposal of Hanford Site tank wastes	WHC, 1993a

DOE = U.S. Department of Energy

Ecology = Washington State Department of Ecology

EPA = Environmental Protection Agency

NEPA = National Environmental Protection Agency

Continued development of a new and completely satisfactory strategy for disposal of Hanford Site tank wastes was further complicated by DOE's decision in 1992 to proceed with all the legal steps required to make a decision to retrieve all SST wastes for pretreatment and disposal. At the same time, the DOE established the Tank Waste Remediation System (TWRS) to address safe interim management of both SST and DST wastes; mitigation and remediation of all tanks with unreviewed safety questions; and retrieval, pretreatment, and disposal of all tank wastes.

In response to the 1992 DOE actions, Westinghouse Hanford Company with assistance from Battelle's Pacific Northwest Laboratory conducted extensive and exhaustive examination of several technical options for disposal of Hanford Site tank waste. Results of these studies, published in 1993, were used by the DOE to establish the current TWRS strategy (Figure 4). This strategy embraces three key activities:

- Manage Tank Waste
- Process Tank Waste
- Manage System-Generated Waste and Excess Facilities

Manage Tank Waste

The proposed new TWRS strategy for managing tank wastes focuses first upon mitigation and/or remediation of tank safety issues. It also includes establishment of safety

and environmental basis envelopes for continued tank farm operations. The tank farm infrastructure will be upgraded to compliance with today's standards.

Process Waste

The proposed new technical strategy for processing tank waste addresses changes that have occurred since the current baseline was developed. Early progress towards disposal will be initiated by retrieval of supernatant liquid and/or salt cake and subsequent processing to remove selected radionuclides. These radionuclide removal operations will be performed either in distributed compact processing units or in a to-be-constructed initial pretreatment module.

For processing sludge, two bounding technologies will be developed in parallel until sufficient technical data are available to permit decision makers to choose a disposal processing method. The bounding technologies are as follows:

- Extensive Separations. Technology, much of it still being developed, will be used to separate retrieved tank wastes into a small volume of high-level waste and a large volume of low-level radioactive waste. The goal is to produce low-level radioactive waste with greatly reduced chemical toxicity and whose radionuclide concentrations are within Nuclear Regulatory Commission limits for Class A waste. The small volume of high-level radioactive waste will be vitrified for eventual deep geologic disposal.
- High-Capacity Vitrification. Currently available and relatively simple technology will be used to separate retrieved tank wastes into high-level and low-level fractions. The high-level fraction will be vitrified, while the low-level radioactive waste fraction may be converted to a solid cementitious product for near-surface disposal. In this bounding technology, the low-level radioactive waste fraction would meet regulatory requirements for near-surface disposal but would contain higher radionuclide concentrations than the waste obtained in the extensive separations approach.

Manage System-Generated Waste and Excess Facilities

Low-level radioactive waste generated during processing of tank wastes will be disposed of as grout or some other compliant form. Other secondary wastes will be prepared for disposal to meet applicable regulatory requirements. Excess facilities will be transferred to the Environmental Restoration Program for decontamination and decommissioning.

PRETREATMENT TECHNOLOGIES

As noted in the previous section, pretreatment of Hanford Site tank wastes prior to their disposal has long been and continues to be a highly important element of all disposal strategies. Pretreatment processes are essentially partitioning processes to divide the tank wastes into a high-level radioactive waste fraction and a low-level radioactive waste fraction. Envisioned pretreatment processes range from simple operations to very complex chemical separations processes. Thus, pretreatment processes currently under consideration for application to Hanford Site tank wastes include the following:

- Water dissolution of salt cake
- Solid-liquid separations, e.g., salt cake solution from sludges, etc.
- Acid dissolution of sludges
- Removal of ^{137}Cs from salt cake and/or sludge solutions
- Removal of ^{99}Tc from salt cake and/or sludge solutions
- Removal of ^{90}Sr from sludge solutions and, as needed, salt cake solutions
- Removal of transuranium (TRU) elements from sludge solutions and, as needed, salt cake solutions
- Destruction of organic chelators in both alkaline and acidic waste solutions

The simplest pretreatment scheme involves treatment of the retrieved salt cake-sludge mixture with water to solubilize sodium salts and most of certain radionuclides, e.g., ^{137}Cs and ^{99}Tc . Appropriate solid-liquid separation techniques, e.g., decantation, filtration, etc., are used to separate the salt cake solution from the undissolved sludge. After washing with water, the separated sludge is vitrified. Selected radionuclides, e.g., ^{137}Cs , ^{99}Tc , and, perhaps, others are removed from the salt cake solution and the sludge washes before their incorporation and solidification in a grout form for onsite disposal. This simple pretreatment approach, first studied by Schulz (1980), would be applied if the High Capacity Vitrification bounding technology approach of the TWRS strategy is implemented.

Pretreatment of the washed sludge to reduce the mass of material, mainly nonradioactive constituents, which must be vitrified involves additional processes to dissolve it and remove selected radionuclides, e.g., TRU elements, ^{90}Sr , ^{99}Tc , and, possibly, ^{137}Cs . Potential reagents and procedures which may be applied to aqueous dissolution of various types of Hanford Site SST sludges have been discussed by Schulz and Kupfer (1991).

Various aqueous-based processes are available (Table 3) for removing selected individual radionuclides from acidic and, in some cases, alkaline media. A bounding process, the so-called CLEAN process (Straalsund 1992), has been proposed for removal of TRU elements, ^{90}Sr , ^{99}Tc , and ^{137}Cs from acid solutions of Hanford SST sludges. [The CLEAN process is an adaptation of the previously known CURE (Clean Use of Energy) concept (WHC, 1990)]. Two major objectives of the CLEAN process are (a) to generate a large volume of low-level radioactive waste meeting or exceeding Nuclear Regulatory Commission specifications for Class A waste and (b) to produce not more than 1,000 canisters (61 cm diameter x 3 m long) of vitrified high-level radioactive waste. To achieve this latter goal, the CLEAN process also entails processes for separation of trivalent actinide elements from trivalent lanthanides and of ^{90}Sr from any associated nonradioactive barium.

It is not yet clear whether the CLEAN or some less ambitious radionuclide removal scheme will be implemented on a plant-scale at the Hanford Site. What is clear is that some form of pretreatment of Hanford Site tank wastes continues to show great advantages in schemes for final disposal of such wastes. It is also clear that, for some time in the future, scientists and engineers in various DOE laboratories and elsewhere will spend considerable energy in modifying, developing, and demonstrating pretreatment technology for use in disposal of Hanford Site SST and DST wastes.

Table 3. Listing of Representative Candidate Technologies for Pretreatment of Hanford Tank Wastes.

Candidate Technology		Reference
1.	<u>Sludge Dissolution</u> Aqueous Dissolution Procedures Fusion Dissolution Procedures	Schulz and Kupfer, 1991 Schulz and Kupfer, 1991
2.	<u>TRU Element Removal^(a)</u> TRUEX SX ^(b) Process CMP SX Process Diamide SX Process	Horwitz and Schulz, 1985 WHC, 1993b Cuillerdier et al., 1993
3.	<u>⁹⁹Tc Removal</u> TRUEX SX Process ^(a) CMP SX Process ^(a) Anion Exchange Resins ^(c)	WHC, 1993b WHC, 1993b Roberts et al., 1962
4.	<u>⁹⁰Sr Removal</u> SREX SX Process ^(a) Inorganic Sorbents ^(a,d) Cobalt Dicarbolide SX Process ^(a,c)	Horwitz et al., 1991 Schulz and Bray, 1987 Reilly et al., 1992
5.	<u>¹³⁷Cs Removal</u> Cobalt Dicarbolide SX Process ^(a,c) Inorganic Sorbents ^(a,e) Organic Sorbents ^(c,f) Precipitation ^(a,c,g)	Reilly et al., 1992 Schulz and Bray, 1987 Bibler and Wallace, 1987 Schulz and Bray, 1987
6.	<u>Destruction of Organic Chelates</u> Ozonation ^(c) Calcination	Bollyky and Beary, 1981 Colby, 1993

(a) From strongly acidic solutions

(b) SX = Solvent Extraction

(c) From alkaline solutions

(d) For example, antimonie acid

(e) For example, zeolites, crystalline silicotitanates, etc.

(f) For example, formaldehyde- or resorcinol-formaldehyde-based cation exchange resins

(g) For example, cesium phosphotungstate, cesium tetraphenylborate, nickel ferrocyanide, etc.

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Figure 1. Single-Shell Tanks.

Single-Shell Tanks

- 149 tanks constructed 1943-1964
- Bottom of tanks at least 50 m (150 ft) above groundwater
- No waste added to tanks since 1980
- Tanks currently contain
 - ~9,800 m³ (37 Mgal) of saltcake, sludge, and liquid
 - ~570 x 10¹⁵ Bq (155 MCi)

Figure 2. Double-Shell Tanks.

Double-Shell Tanks

- 28 tanks constructed between 1968-1986
- 3,785 m³ to 4,315 m³ (1 to 1.14 Mgal) capacity
- Tanks currently contain
 - $\sim 9 \times 10^4$ m³ (24 Mgal) of mostly liquids (also sludges and salts)
 - $\sim 400 \times 10^{15}$ Bq (110 MCi)
- None have leaked

Figure 3. Hanford Site Radioactive Waste Tanks and Capsule Disposal Plan 1988-1991

Hanford Site Radioactive Waste Tanks and Capsule Disposal Plan 1988-1991

Figure 4. Tank Waste Remediation System Proposed New Technical Strategy.

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