HANFORD TANK WASTE REMEDIATION SYSTEM FAMILIARIZATION REPORT

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QUALITY OF DATA: Sources of data are referenced in each chapter. The respective sources of non-CNWRA data should be consulted for determining their levels of quality assurance.

ANALYSES AND CODES: Computer software used in analyses contained in this report are: ORIGEN2, Version 2.1; MCNP, Version 4A; ArcView, Version 2.0B; and ARC/INFO, Version 6.1. All these codes are commercial codes and only the object codes are available to the CNWRA. ORIGEN2 and MCNP are defined in Technical Operating Procedure (TOP)-018, Development and Control of Scientific and Engineering Software, as acquired/existing software required to be under CNWRA control. ORIGEN2 and MCNP are currently being placed under TOP-018 control. ArcView and ARC/INFO are graphical management software which are used to store, manipulate, and display data and are not controlled under TOP-018.

EXECUTIVE SUMMARY

The U.S. Department of Energy (DOE) established the Tank Waste Remediation System (TWRS) program at the Hanford site to manage the retrieval and cleanup of radioactive waste contained in 177 aging underground storage tanks. The DOE plans to privatize the waste solidification operations under a two-phase program. In Phase I, a feasibility study, scheduled for completion in January 1998, would be conducted by two contract teams, and one or more demonstration facilities would be constructed for solidifying about 10 percent of the waste by June 2011. In Phase II, which will be a full-scale operation phase, all the wastes are planned to be processed. Under a Memorandum of Understanding, reached between the Nuclear Regulatory Commission (NRC) and DOE for Phase I activities, the NRC will develop sufficient knowledge of the physical and operational situation of the Hanford waste tanks and Phase I activities to enable the NRC to (i) assist the DOE in performing reviews in a manner consistent with the NRC regulatory approach, and (ii) be prepared to develop an effective regulatory program for the possible licensing of DOE contractor-owned and -operated facilities during Phase II. The review of information pertaining to the Hanford site, tanks, and TWRS reported in this document is a first step in this process.

Chapter 2 summarizes the information available regarding the Hanford site geography and geology, status of knowledge regarding site contamination, processes leading to waste generation, tank farms, transfer systems, and ongoing activities pertaining to TWRS. This chapter also describes the status of characterization of site contamination. While groundwater contamination has been rather extensively characterized, there is less information available on the vadose zone.

Identification and quantification of Hanford tank waste contents are subjects of extensive study. Chapter 3 of this report includes a general description of double-shell tank and single-shell tank waste characteristics, and a discussion of tank inventories of chemicals and radionuclides. The wastes have been produced over a long period of time by a variety of processes. Characterization of tank contents chemically and radiologically is, therefore, a challenging task. Two approaches to this question are being employed, each complementing the other: direct sample assay and estimation based on facility records. The former is limited by the extreme physical and chemical heterogeneity of the tank contents, while the latter may be unreliable due to incomplete or inaccurate documentation of process and waste transfer transactions. The DOE effort is centered on determination of a "best-basis" value for each constituent in each tank, based on a combination of the assay and historical data. Until that evaluation process is completed, the historically-based Hanford Defined Waste (HDW) model being developed at Los Alamos National Laboratory (LANL) is the most complete and thorough dataset of tank inventory estimates. The Center for Nuclear Waste Regulatory Analyses (CNWRA) has prepared a database (based on inventories from the HDW model) allowing access of tank information utilizing ARC/INFO geographical information system software. The geographic and geologic map presented in appendix B can be combined in ArcView with the location of the tank farms, if their coordinates can be accurately described, to represent the complete spatial description of the tank farms for eventual hazard analysis.

Chapter 4 reviews the various hazards posed by tank wastes and associated with the retrieval and mixing of wastes prior to solidification. The safety issues associated with solidification will be discussed in another report as part of subtask 1.2. The hazards posed by tank wastes and the TWRS activities are classified under four Watch-list categories: (i) flammable gas, (ii) organic oxidation, (iii) ferrocyanide oxidation, and (iv) high-heat. Other hazards identified in this review include: crust burn associated with secondary ignition of organic-nitrate/nitrite mixtures in the crust layer, High-Efficiency Particulate Air

filter blow out associated with flammable gas ignition, environmental hazards and flammability due to organic solvents, known and unknown leaking tanks, criticality, and lightning strikes. This report also evaluates potential safety concerns associated with retrieval, mixing, and transfer of tank wastes. Two important concerns in this regard are: (i) safety problems arising from commingling wastes under interim storage, and (ii) operability of waste transfer systems that may be impeded by plugging, trapped flammable gas, exothermic reactions, and corroded lines. The physical and chemical data needs for compatibility assessments are provided in chapter 4.

The time constraints did not permit a comprehensive or critical review of the information on the Hanford site and TWRS activities. However, based on the reviews performed to date, the status of knowledge and additional information requirements, as outlined in chapter 5, may be summarized as follows.

- Knowledge of vadose zone contamination in the potential areas of TWRS activity and waste disposal is sparse, although several investigations are under way to characterize contamination migration in the vadose zone under some of the tanks (e.g., SX tanks), and in the 600 Area associated with a commercial Low-Level Waste disposal site. On the other hand, considerable effort has been made in characterizing the groundwater contamination in the unconfined aquifer. Understanding vadose zone contamination is necessary for performance assessments associated with Low Activity Waste disposal, as well as hazard analyses related to waste retrieval and solidification.
- Knowledge of tank waste contents and chemistry is evolving, due to ongoing waste characterization programs and estimations based on facility records. However, better understanding of the chemical interactions of various waste constituents is necessary to assess the safety aspects of retrieval and mixing of wastes from different tanks, and to determine the operating envelopes for solidification. Knowledge of the distribution of chemical species within the tanks is also necessary for evaluation of contaminant migration under the tanks.
- There is a paucity of easily accessible documentation on the location, design, and problems associated with waste transfer lines within the 200 Areas and the cross-transfer line. It is known that some of these transfer lines have suffered from plugging and leakage. Knowledge of processes leading to plugging and extent of leakage is necessary to better understand safety hazards associated with waste mixing and transfer to the privatization facilities.
- The evaporator/crystallizer in the 200-East area, which has been monitored closely over the past few years, has shown a service life before materials replacement of about 10 years. However, it is not known how the service life will be affected by the mixture of wastes expected during the Phase II operations.
- The information on the Hanford site and various components of the TWRS is scattered and, in some cases, not easily accessible. It is recommended that this report be augmented as further information is gained, to assist in future safety analyses.

1 INTRODUCTION

The U.S. Department of Energy (DOE) established the Tank Waste Remediation System (TWRS) program at the Hanford site in 1991 to manage the maintenance and cleanup of radioactive waste contained in 177 aging underground storage tanks. The DOE is legally bound to remediate the waste tanks under the Hanford Federal Facilities Agreement and Consent Order of 1989 (Ecology, 1994), also known as the Tri-Party Agreement (TPA). To accomplish the TWRS requirements, the DOE plans to privatize the waste treatment and immobilization operations. The TWRS privatization is divided into two phases, a proof-of-concept or demonstration phase (Phase I) and a full-scale operations phase (Phase II). The Phase I program, scheduled for completion in 2012, is divided into Part A (feasibility study), which is scheduled for completion in January 1998, and Part B (demonstration pilot plant study), which is scheduled for completion in June 2011. A Memorandum of Understanding has been reached between the DOE and the Nuclear Regulatory Commission (NRC)1 for Phase I activities, which provides for the NRC to acquire sufficient knowledge of the physical and operational situation at the Hanford waste tanks and processes involved in Phase I activities to enable the NRC to (i) assist the DOE in performing reviews in a manner consistent with the NRC regulatory approach and (ii) be prepared to develop an effective regulatory program for the possible licensing of DOE contractor-owned and contractor-operated facilities during Phase II. A program to assist the NRC in developing technical and regulatory tools for the TWRS privatization activities was established at the Center for Nuclear Waste Regulatory Analyses (CNWRA). The program consists of four tasks, of which only task 1 (Familiarization and Regulatory Development and Safety Review) is currently active. The objective of the CNWRA activities in subtask 1.1 is to gather detailed, current information related to the Hanford site in general, and the 200 Area tank farms in particular, that will be useful to support execution of other subtasks.

The volume of information on the Hanford site and activities related to environmental cleanup is quite large (over 6,500 documents in the DOE bibliographic database that can be accessed via the Internet). Because of the limited time available to acquire the relevant documents and review the information for inclusion in the present report, this report is necessarily neither critical nor complete in addressing the information. This report is prepared in a modular format so that, as further information is acquired, corrections or augmentation of the present report can be made in the future. Chapter 2 of the report provides a description of the site and facilities. Included in this chapter are the descriptions of site geology and geohydrology and the present understanding of sitewide contamination of both radioactive and hazardous species. Histories and brief descriptions of the processes that produced the wastes are also provided. Finally, descriptions of various operational areas, especially of the tank farms and transfer facilities, and ongoing activities relevant to TWRS are included. Chapter 3 of the report and appendix A provide a tank-by-tank description of waste content. The list of tank waste contents is derived from the Tank Waste Inventory Network System (TWINS) database. As part of the familiarization activities in subtask 1.1, a pictorial database of tanks was constructed using a Geographic Information System. A summary of the information available in this database is provided in appendix B. Chapter 4 reviews the hazards posed by tank wastes and TWRS operations, with the exclusion of waste solidification operations, which have not been initiated at the Hanford site as part of the TWRS. Detailed information of tank waste contents is provided in the appendix. A glossary of frequently used terms is also included with the report.

¹ Memorandum of Understanding Between the Nuclear Regulatory Commission and The U.S. Department of Energy, January 29, 1997, Federal Register, V. 62, No. 52, 12861. March 18, 1997.

regulatory compliance based on Phase I and II data. Phase I and proposed Phase II facilities are shown in figure 2-4.

Existing vadose zone monitoring systems consist of three soil vapor monitoring wells, three solar stills, and six trench cap areas. Soil gas samples collected from the soil vapor monitoring wells are analyzed for methane, radon, tritium, fixed gases, and benzene, toluene, ethyl benzene, and xylene. Various trench caps from trenches 5 and 7 are regularly sampled and analyzed for radon. The solar stills are used for the collection of tritium samples (Landau Associates, Inc., 1996).

Site contamination values presented in the following paragraphs are based on 1992, 1993, and 1994 US Ecology monitoring data. Low concentrations of some volatile constituents (maximum concentrations of ethyl benzene and xylene at 1 mg/m³ and 3 mg/m³, respectively) primarily in the gas phase, have been detected at the trench edges indicating limited migration. Samples collected from the soil vaporing monitoring wells show slight elevations above background concentrations of carbon dioxide and methane, and reduced oxygen concentrations, which may be indicative of organic wastes decomposition (Landau Associates, Inc., 1996).

Background concentrations for tritium were exceeded in both vapor monitoring wells and the solar stills with recorded maximum concentrations of about 400,000 pCi/L and 4100 pCi/L, respectively. Based on these findings, tritium migration from the trenches is indicated. Radon concentrations were elevated in well VW-101, measuring up to > 1627 pCi/L, and close to background concentrations in well VW-102. Although trench cap radon concentrations were measured at levels below background, these low concentrations are believed to be the result of dilution from the surrounding atmosphere (Landau Associates, Inc., 1996).

In 1991, soil samples also were collected from about the 15-28.3 m (45-85 ft) range below land surface during the vadose zone monitoring well installations and analyzed for volatile organic constituents. Analytical results indicated no volatile constituents were present. No indication of radionuclides analysis was given for these samples in the US Ecology reference report.

Graphics presenting detailed areas of soil contamination were not readily available at the time of this writing. However, through continued DOE work to better determine the extent and magnitude of soil contamination, maps delineating site surface and subsurface soil contaminants should become available.

2.3.3 Surface Water and Sediment Contamination

Surface water and sediment samples are collected from riverbank springs and the Columbia River, with additional surface water monitoring of onsite ponds and offsite water sources. Water samples collected from the surface water disposal units and springs for the 1995 monitoring program were analyzed for Pu-238, Pu-239, Pu-240, tritium (H-3), Sr-90, U, and gamma-emitting radionuclides, as well as pH, temperature, and nitrate. Surface water disposal units in the 200 Areas consist of holding ponds, such as the B and U Ponds, and drainage ditches. The results indicated that radionuclide concentrations in the surface water in the 200 Areas were mostly at or below detection limits, and below DOE Derived Concentration Guides of 100 millirem per year (mrem/yr). Nitrate concentrations were below applicable Drinking Water Standards and pH measurements were below liquid effluent RCRA standards.

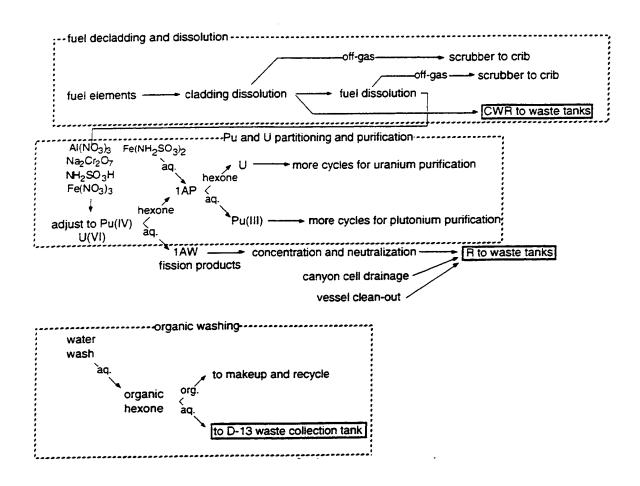


Figure 2-11. A synopsis of the REDOX process and associated waste streams (Agnew, 1997)

organic phase. The wastes, which were in the aqueous phase, went primarily to the S and SX farms. The organic phase was further washed with aqueous Fe(II) solution to reduce the Pu to Pu(III) which then partitioned to the aqueous phase. The Pu can be oxidized again to the tetravalent state to separate it from other actinides such as americium (Am). After removal of U from the organic phase, the organic medium was washed with water to remove the residual radionuclides and then recycled. The aqueous waste was sent to the tanks.

2.5.5 PUREX Process

The PUREX process started as a pilot plant in the Hot Semiworks (C plant) and became a production process in January 1956 in the PUREX or A plant. Al and Al-Si cladding was dissolved using caustic solution which does not affect the fuel. From 1968 to 1972, Zr clad was treated to dissolve the Zr cladding using a process called Zirflex process, presumably using hydrofluoric acid. Following cladding dissolution, the fuel was dissolved in nitric acid. After cladding and fuel dissolution, the aqueous Pu and U were complexed in a nitric acid solution and extracted into a TBP/NPH (kerosene) organic phase. The separation process chemistry is similar to that in the REDOX process, as shown by the process synopsis in figure 2-12. The redox condition prior to solvent extraction of U and Pu is adjusted using ferric nitrate instead of a mixture of nitrates, bisulfites, and dichromates as in the case of the

recovery. The acid PUREX wastes were sent directly to the B plant. The waste from the B plant after Cs and Sr recovery was then directed to the B and BL tank farms.

The Cs and Sr recovery process synopsis is shown in figure 2-13. For the caustic Cs wastes, zeolites were used to extract Cs. For the acidic wastes, phosphotungstic acid was added to precipitate Cs. The supernatant from this process was treated with nitric acid and the Sr was recovered using solvent extraction into an organic phase containing TBP. The organic phase was washed as described before for the REDOX process to recover the organic solvent and recycle to the solvent extraction process. The Cs was converted to a Cs chloride (CsCl), melted, and stored in double-walled capsules made of Type 316L stainless steel. The Sr was converted to a strontium fluoride (SrF2) in a powder form, compacted, and stored in capsules of similar design to the Cs capsules. However, for the Sr capsules, the inner wall was made of a Ni-base alloy, Alloy C-276, and the outer wall was made of Type 316L stainless steel. A schematic of the capsule design is shown in figure 2-14. The centerline and surface temperatures calculated for air (figure 2-14) are presumably relevant to open, still-air conditions, although the assumptions in this calculation are not shown in the Environmental Impact Statement (EIS). There are at present 601 Sr and 1,328 Cs capsules with a total radioactivity of 76.3 MCi as of 1994. The capsules are stored under water in the Waste Encapsulation and Storage Facility (WESF) located close to the B plant. Monitoring and maintenance activities of the capsules involve calculating annual inventory, physically verifying that the inner capsule has not bulged (for Cs capsules only), and monitoring pool cell water contamination. The Cs capsules are "clunk tested" on a quarterly basis to determine if bulging of the inner canister has occurred. This is done in the pool itself by grasping one end of the capsule with the pool tong and moving it rapidly vertically by about 15 cm. This allows the inner canister to slide down within the outer capsule, making a clunking sound. If the capsule fails the clunk sound, it is removed to the hot cell for additional evaluation. Various alternatives for the long-term storage and disposal of these capsules are being pursued (U.S. Department of Energy, 1996b). The WESF is scheduled to be decontaminated and decommissioned within the next 10 years.

2.5.7 Other Processes

Various other processes were initiated in the production of Pu in the Z plant [also known as the Plutonium Finishing Plant, (PFP)], located in 200-West. The plant started by concentrating the Pu nitrate from the B and T plants into a paste, which took place from 1945 to 1949. In 1949, production of Pu metal was started and continued with some hiatus until 1988. This process used carbon tetrachloride, nitric and hydrofluoric acids, and various oils and degreasers. The wastes from these operations were sent to tanks TX-118 and SY-102 (Agnew, 1997). The RECUPLEX process started in 1955 to recover Pu from the Z plant waste stream and, during the operation, generated various organic wastes. Due to a criticality event, the RECUPLEX facility had to be shut down in 1962 (U.S. Department of Energy, 1992a). This was replaced by the Plutonium Reclamation Facility (PRF) which operated along the same principles until 1987. Another operation that took place in the Z plant was the recovery of Am from the PFP waste stream using an ion-exchange process. This process was stopped in 1976 after an explosion in one of the recovery units (U.S. Department of Energy, 1992a). A review of reports regarding the analyses of the causes of the explosion has not been performed.

2.5.8 Solidification Technologies

The history of various solidification processes at the Hanford site and other DOE sites has been described by McElroy and Platt (1996). Prior to 1965, solidification experiments were carried out in the

2.6 DESCRIPTION OF OPERATING FACILITIES

2.6.1 Overview of TWRS Facilities and Components

The TWRS facilities consist of waste tanks, evaporators, transfer piping, and processing vaults. Additionally, the privatization contractors are expected to construct the solidification facilities and waste handling systems. In evaluating the hazards associated with TWRS operations, it is important to obtain information on the materials of construction of various components. An overview of the materials of construction is provided in table 2-2 (Edgemon and Anantatmula, 1995). It must be noted that discrepancies can be found in the description of materials of construction and corrosion protection methods between various Hanford site reports. Where possible, these differences are pointed out. As further information becomes available, a resolution of these discrepancies may be attained.

2.6.2 Waste Tank Operations

Approximately 99 percent of the total radioactive waste volume at the Hanford site is stored in underground tanks. The tanks are grouped into tank farms and buried approximately 6 to 8 feet below ground in the 200-East and 200-West Areas (figure 2-15). In addition to the tank farms, there are Miscellaneous Underground Storage Tanks (MUSTs), most of which are inactive.

The first 149 tanks constructed at the Hanford site, starting in 1943, were made of a single carbon steel wall and floor in the form of an open container encased in an outer shell and dome of reinforced concrete. There are four basic designs of these single-shell tanks (SST), as shown in figure 2-16, with capacities ranging from 208 to 3,785 m³ (55,000 to 1,000,000 gal). Construction of these SSTs was discontinued after 1964.

The tanks have two primary functions: (i) confinement—the tank shells and liners provide confinement barriers for liquid and solid wastes, and (ii) structural stability—the reinforced concrete structure and the steel liners of tanks provide acceptable safety margins for continued operation under normal and abnormal loads (Ohl et al., 1994). Table 2-3 provides details of the construction of the 149 SSTs including information on year of construction, location, farm, number of tanks in farm, capacity, steel grade, and condition of the steel plate. The SSTs were built by welding steel plates using flux- and later gas-covered electrodes, but none of the tanks were subjected to the stress relief post-weld heat treatment (PWHT) that is used in most recent designs. Changes in the specification of the steel were due to the introduction of new steel grades reflecting improvements in steel manufacturing practices and the development of tighter specifications by the American Society for Testing and Materials (ASTM) and American Society of Mechanical Engineers (ASME). Nominal design lives are not available in the SST design archives but an intended use of 20 to 40 yr temporary storage is generally accepted by the current Hanford site engineering staff (Ohl et al., 1994). All the SSTs have exceeded their intended life and 67 of them are known or assumed to have leaked radioactive waste to the surrounding soil (see chapter 4).

It is considered (Anantatmula et al., 1995) that the leakages experienced by the SSTs are the result of the initiation and propagation of cracks in the proximity of the welds due to stress corrosion cracking (SCC). The residual stresses arising from the welding operations contribute to the SCC. This failure mechanism was confirmed for non-stress-relieved waste tanks at the Savannah River Site (Poe, 1974) and it has been observed in laboratory tests simulating the chemistry of the waste (Ondrejcin, 1978; Kirch, 1984). SCC of carbon steels in hot alkaline nitrate solutions within the range of concentrations typical of high-level radioactive wastes resulting from the reprocessing of spent fuel is a well recognized

phenomenon (Donovan, 1977; Ondrejcin et al. 1979; Cragnolino, 1993). The occurrence of SCC can be inhibited, however, at high $[NO_2^-]/[NO_3^-]$ and $[OH^-]/[NO_3^-]$ ratios (Ondrejcin et al., 1979). Therefore, it may be possible to avoid SCC, even in tanks with relatively high residual stresses along the welds, by controlling the concentration of these anions in the waste streams entering the tanks or by pumping out of the tanks the supernatant and the interstitial liquids. These approaches have been adopted at the Hanford site in the process of stabilizing the waste in specific SSTs, an operation designated as tank stabilization.

As part of the resolution of waste tank safety issues at the Hanford site, other failure mechanisms and corrosion control options have been identified to minimize further degradation of the SSTs (Ohl et al., 1994). Localized (pitting and/or crevice) corrosion is another potential failure mode for SSTs (Anantatmula et al., 1995). A corrosivity factor (CF) has been defined as the ratio of the molar concentration of NO₃⁻ to the combined molar concentration of NO₂⁻ and OH⁻ to evaluate the propensity to localized corrosion of the waste contained in a tank. The critical CF above which the waste promotes localized corrosion is estimated to be 2.5. Different actions are recommended and eventually adopted for each tank, including pumping of liquid, addition of NaOH, and corrosion monitoring depending upon the estimated value of CF. Other failure modes, such as uniform corrosion, microbially influenced corrosion (MIC), concentration cell corrosion, erosion corrosion, hydrogen embrittlement, thermal embrittlement, radiation damage, fatigue, creep/stress relaxation, mechanical wear, and environmental degradation of the reinforced concrete are not expected as generic problems under the conditions prevailing in the tanks, although isolated instances of failure due to some of these processes may be plausible (Ohl et al., 1994, Anantatmula et al., 1995; Edgemon and Anantatmula, 1995).

Over the years, the design of the SSTs changed to better accommodate the waste being stored and to reduce the occurrence of corrosion. Alterations include adding equipment to handle self boiling waste, increasing size and changing the bottom to a flat surface instead of a bowl shape. Another change was the addition of a grid of drain slots beneath the steel liner. The grids were designed to collect leakage and divert it to a leak detection well. Another design difference is that several SST were built in cascades of three or four tanks connected with piping at different levels. Thus, when a tank filled to the level of the pipe, waste would flow through the pipe to the next tank. This construction allows the contents of the tank to settle to the bottom and therefore, the waste that went to the next tank had less solid and less radioactivity (mostly in the form of Cs, since Sr had settled out in the solids). This design also allowed the waste to be pumped into one location until all the tanks were full, reducing the amount of waste rerouting to fill the tanks in a particular cascade group.

Figure 2-17 shows the configuration of the instrumentation currently available in SSTs. All SSTs have measuring devices to monitor the surface level of the waste, including manual tape, automatic FIC, which is a device manufactured by the Food Instrument Company, and/or ENRAF, which is a gauge fabricated by ENRAF Incorporated. These tanks have thermocouples and a camera observation port for taking in-tank photographs and videos. Drywells are located around the SSTs to allow monitoring by gamma radiation or neutron-moisture sensor of any tank leakage. However, only two SSTs are currently monitored monthly by gamma radiation sensor. The remaining drywells are monitored upon request as is the case of monitoring by neutron-moisture sensors.

Starting in 1968, 28 double-shell tanks (DSTs) were built with a capacity of approximately 3,780 m³ (1,000,000 gallons) each. They are composed of an inner, freestanding, completely enclosed carbon steel tank which is referred to as the primary tank. The primary tank is located inside a reinforced concrete shell and dome with the walls covered with a steel liner, as shown in figure 2-18. The liner is

service. Through the useful life analysis it was concluded that the rate controlling mechanism for DST failure is primary tank breach by pitting corrosion in the vapor phase. The probability of this type of failure was estimated to range from 0.4 to 0.6 for the expected 40 to 60 yr of service life. In order to relate pit propagation with a parameter associated to tank operation, the following equation was derived to relate pit propagation rate (PPR) with CF (discussed previously) with a correlation coefficient $r^2 = 0.94$.

$$PPR \ (mils/yr) = 3.28 (CF)^{0.23}$$
 (2-1)

The expression was derived from a limited set of experimental data obtained with steel coupons exposed to the liquid phase, the vapor phase, and the interface between these phases of various waste types, including PUREX, REDOX, and BiPO₄, using both tank samples or laboratory simulated wastes (Ohl et al., 1996).

As a first level screening process to guide the integrity inspection and the corrosion monitoring of the tank, the DSTs were grouped into different categories (Ohl et al., 1996) according to their potential susceptibility to pitting corrosion and SCC. The screening for susceptibility to pitting corrosion of DSTs was performed using the CF to classify the DSTs into two groups (nonsusceptible and susceptible). On the basis of values of CF greater than 2.5, calculated from the estimated concentrations of NO₃⁻, NO₂⁻, and OH⁻, either for the supernatant or the sludge, only three DSTs (AN 107, AP 107, and AW 104) were considered to be susceptible to pitting corrosion. In order to screen for the susceptibility to SCC, the DSTs were classified into three groups by Ohl et al. (1996), using a different criterion developed by Ondrejcin (1978) at SRS. Ondrejcin determined the environmental domains of SCC at 50, 60, 70, 80, and 100 °C in terms of the concentrations of NO₃⁻ (1.5 to 5.5 mol/L), NO₂⁻ (0 to 3.5 mol/L), and OH⁻ (0 to 5.0 mol/L) using statistically designed, slow strain rate, SCC tests. An elongation to fracture lower than 13 percent was selected as indicative of SCC susceptibility and found to be consistent with the results of SCC tests using a fracture mechanics approach. Ohl et al. (1996) estimated the environment compositions and temperature for the DSTs, and classified them as: (i) Group I tanks, which included a majority of DSTs, that may have low susceptibility to SCC (i.e.elongation to failure greater than 13 percent), (ii) Group II tanks, which includes three DSTs (AW 104, SY 101, and SY 102), considered to be potentially susceptible to SCC (i.e. the elongation to failure was estimated to be lower than 13 percent), and (iii) Group III tanks (AY 101, AZ 101, and AZ 102), which exhibited temperatures above 100 °C, also considered to be potentially susceptible to SCC. The group III was created because Ondrejcin's tests did not address temperatures beyond 100°C and hence, as a conservative measure, the tanks with temperatures above 100°C were considered to be susceptible to SCC even if the chemistry of the wastes in these tanks did not indicate any SCC susceptibility. In summary, six tanks were evaluated as being potentially susceptible to SCC, but only Tank AW 104 was found to be susceptible to both pitting and SCC. However, as noted by Ohl et al. (1996), the screening approach needs to be verified through inspections, because it is based on current estimates of the bulk waste solution, without considering local concentrations in crevices or other areas. The screening approach also does not consider the potentially adverse conditions that may have existed in the past which could have initiated pitting or SCC.

Figure 2-19 shows the configuration of the instrumentation used in DSTs. The main difference with respect to the SSTs is the existence of the annulus. Monitoring leakages in the DSTs can be easily accomplished by continuous air monitoring in the annulus using radiation detectors or by conductivity probes that are activated in the presence of an electrolytic conductor. In DSTs, there are usually one or more thermocouple trees in risers in the primary tank.

is necessary to determine the effect of various TWRS design decisions on consequences, including selection of tanks for mixing prior to solidification. The details of the watch-list tanks are provided in chapter 4.

2.7.3 Unreviewed Safety Questions

The Unreviewed Safety Questions (USQ) program is a formal administrative program that aims to identify known or suspected operating conditions outside established safe limits. These limits form the authorization bases for continued operation of the tank farms. Some of the watch-list tanks were under this category until the safety issues associated with their operation were identified and they were placed under a specific watch-list category. Criticality with respect to the tank contents was originally an unreviewed safety question, but the criticality issue was closed in 1994. Currently, there are no tanks in the criticality watch-list. However, criticality during tank waste retrieval will be addressed on a tank-by-tank basis during remediation. Details of the watch-list categories are provided in chapter 4. The TPA requires that all unreviewed safety questions be resolved by September 1998. Recently, some tanks containing dry organic nitrates were placed under the USQ Program because methods of analyzing accident scenarios have become available for these (Hanlon, 1996).

2.7.4 Continued Operation of Tank Farms

In addition to routine operations, such as maintenance of facilities and equipment, a number of safety management activities are being conducted. Among these activities, those that may influence the NRC review of TWRS activities include: (i) combining compatible tank waste types through existing cross-transfer piping in order to provide tank space and address safety issues, (ii) screening and characterizing waste on a tank-by-tank basis for remedial actions, (iii) isolating and removing pumpable liquids from SSTs, and (iv) operating the 242-A evaporator to concentrate wastes and remove contaminants from residual liquids. These activities are not likely to fall within the Phase I remediation program but may be important in hazard analyses related to Phase II activities.

In addition to screening and characterizing waste on a tank-by-tank basis, monitoring of the effect of waste additions on the corrosion of the tanks has been initiated using electrochemical noise (EN) probes (Edgemon and Bell, 1996). Using these probes, the initiation and growth of pits was detected on prototype electrode probes placed in tank 241-AZ-101 following raw water additions (Edgemon et al., 1996; 1997). However, current transients related to pitting decreased in magnitude and frequency with increasing times after the water addition, suggesting that conditions leading to uniform corrosion were reestablished. Recommendations for improvements in the EN probes have been developed with an aim to extend monitoring to other tanks (Edgemon et al., 1996).

Details of the tank and evaporator designs are provided in section 2.6.2 and 2.6.4, respectively. Projections for future tank waste additions are shown in table 2-9 (U.S. Department of Energy, 1996b). This waste is expected to be added to the DSTs after being processed in the 242-A evaporator. The majority of the future waste additions would come from D&D activities at inactive facilities at the Hanford site and would be classified as dilute, noncomplexed wastes (meaning that they do not contain significant quantities of complexing organic chemicals). The 100 Area cleanout waste is classified as double-shell slurry feed waste. This is the waste that is concentrated in the evaporator to a point just below the sodium aluminate saturation boundary. Cleanout of the K Basins would result in the addition of approximately 54 m³ of sludge from spent nuclear fuel, corrosion products, iron and aluminum oxides,

Table 2-9. Projected future waste additions to double-shell tanks after processing in the 242-A evaporator (U.S. Department of Energy, 1996b)

Source	Waste Type	Volume (m³)	Duration of Accumulation		
PUREX: Deactivation waste	DN	5,700	FY94-97		
B Plant: Terminal cleanout waste (concentrated)	DN	2,100	FY97-01		
100 Area:Terminal cleanout waste (concentrated)	DSSF	2,200	FY95-99		
100 Area: Sulfate waste	DN	140	Not reported		
300 Area: Fuel supply cleanout	DN	45	Not reported		
105-F, 105-H: Basin cleanout	DN	850	Not reported		
Tank 107-AN: Caustic addition	DN	190	Not reported		
100-KE, KW: Basin cleanout	DN	1,200	Not reported		
TOTAL		12,400			
DN: Dilute noncomplexed waste					

DSSF:

Double shell slurry feed

concrete, fission and activation products and sand from the outside environment. The sludge waste would add about 11,000 Ci to the DSTs. This would include about 5,200 Ci of Pu-241, 260 Ci of Pu-239, 1,280 Ci of Sr-90, and 970 Ci of Cs-137. Following cleanout, the sludge would be transported in about 1200 m³ of water to the DSTs.

2.7.5 Cross-Transfer Piping

Since the solidification plants are planned to be constructed in the 200-East Area, wastes have to be transported from 200-West to 200-East. The existing cross-transfer piping is nearing the end of the 40-yr design life. Currently, four of the existing six lines are out of service due to plugging, and the two remaining lines do not meet engineering requirements such as double containment and leakage detection. Hence, the construction of regulatory-compliant cross-transfer piping has begun and is expected to be operational by 1998. The causes of plugging may vary, including fluid flow, thermal, and chemical factors. Additional information on the causes of plugging and the nature of precipitates in these pipes will benefit future safety analyses of existing and newly constructed cross-transfer piping systems. Details of the transfer piping are provided in section 2.6.3.

3 DESCRIPTION OF TANK CONTENTS

3.1 NATURE OF TANK WASTES

Effective and safe remediation of Hanford tank wastes requires an understanding of both the potential hazards and the potential responses of waste materials during processing. Therefore, it is important to describe and quantify the chemical and radiological contents of the tanks. Hanford tank wastes may be considered in four categories: SST wastes, DST wastes, MUST wastes, and future tank waste additions as discussed in chapter 2. The discussions of this chapter emphasize the inventory of SSTs and DSTs, which together comprise greater than 99 percent of the total waste volume and a majority of total radionuclide activity at the Hanford site (U.S. Department of Energy, 1996b). The other major contributors to radioactivity in Hanford wastes are the Cs and Sr capsules, which are beyond the scope of this report. The total MUST waste volume is minor and the MUST inventory, while not yet well documented, is expected to differ little in character from the SST and DST inventories (U.S. Department of Energy, 1996b). Future tank waste additions are discussed in chapter 2.

The tanks contain complex mixtures of solids and liquids. Liquids are either supernatant—easily pumped and floating above settled solids—or interstitial—confined to pore spaces of the solids. Solids are classified as sludge or saltcake (Gephart and Lundgren, 1995). Sludge is a thick, wet layer of settled and precipitated water insoluble solids at the tank bottom, with small pore spaces that do not allow removal of liquids. Saltcake is dryer with larger pore spaces, being a residue after evaporation of supernatant liquid; saltcake components are typically water soluble. Slurry is a water/liquid mixture that can be pumped. Figure 3-1 is a photograph of the interior of an SST, showing a variegated solid crust forming on the top of the waste characterization.

The wastes are products of several chemical processes (see chapter 2). Typically, records on the waste contents and waste volumes are incomplete or missing. Direct assay of the tank contents is complicated by (i) the inherent danger in sampling highly radioactive material that is potentially explosive (see chapter 4) and (ii) the heterogeneous nature of the tank wastes. Chemical and radiological characterization is, therefore, a challenging task. In this section, the general characteristics of the wastes are described, while section 3.2 addresses the inventories of chemical and radionuclide components of the wastes. Detailed inventories of the tanks are presented in appendix A. Unless otherwise noted, the following references were the sources for the information contained in this section: (Gephart and Lundgren, 1995; Golberg and Guberski, 1995; Agnew, 1997; U.S. Department of Energy, 1996b).

3.1.1 Single-Shell Tanks

As shown in table 2-3, the 149 SSTs were built from 1943 to 1964 and hold from 208 to 3,785 m³ (55,000 to 1,000,000 gal.) each. Of the combined 132,500 m³ (35 million gal.) of waste, 66 percent is wet saltcake, predominantly sodium nitrate, and 34 percent is sludge. Nearly all separable liquids have evaporated or been transferred from the SSTs to DSTs, but about 23,000 m³ (6 million gal.) of liquid are not easily pumped and will remain in the tanks. The solids and dissolved constituents of the SSTs are 90 percent sodium nitrates and nitrites, with the remainder consisting mostly of phosphates, carbonates, hydroxides, and sulfates. Radioactivity in the SSTs is dominated by Sr-90 (75 percent) and Cs-137 (24 percent); Sr is concentrated in the sludge, while Cs is located chiefly in the saltcake and interstitial liquids.

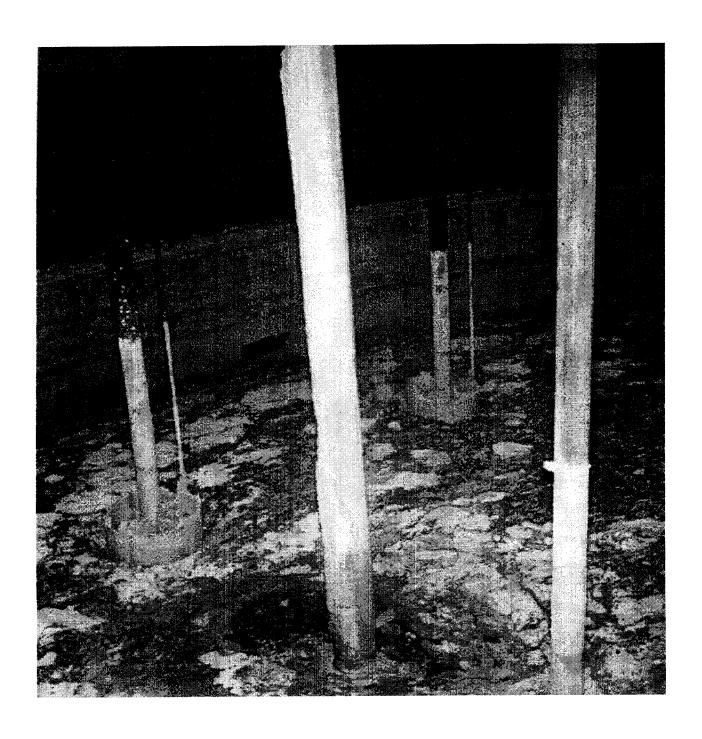


Figure 3-1. Photograph of the interior of tank 241-AX-101, from the web page at http://www.hanford.gov/twrs/char.pub/ax101big.gif

3.1.2 Double-Shell Tanks

The 28 DSTs are newer and larger than the SSTs, having been built between 1968 and 1986 and ranging in capacity from 3,785 to 4,390 m³ (1 to 1.16 million gal.) (table 2-4). Because they are volumetrically dominated by supernatant liquids transferred from SSTs, the 75,700 m³ (20 million gal.) of DST waste are 85 percent water. The waste is thus dominated by liquids and slurries, sometimes with a bottom layer of sludge. DST waste types have been delineated in greater detail than SSTs. Eight types have been defined, listed here in decreasing order of volume (Gephart and Lundgren, 1995; Hanlon, 1996):

- Double-shell slurry and double-shell slurry feed (31 percent of total DST waste volume; various sources)—suspension-rich, high-salt solutions from evaporation of SST and reprocessing plant wastes; includes solids comprising 19 percent of this waste type
- Concentrated complexant (23 percent; various sources¹)—liquid and solid alkaline waste with high organic and transuranic contents, resulting from evaporation of dilute complexed waste; includes solids comprising 17 percent of this waste type
- Dilute noncomplexed waste (21 percent; sources are T, B, REDOX, and PUREX plants, the N Reactor, the 300 Area, and the PFP)—low radioactivity liquid waste from a variety of processing operations; includes solids comprising 9 percent of this waste type
- Neutralized current acid waste (9 percent; PUREX)—93 percent liquid waste generated since 1983
- Concentrated phosphate waste (6 percent; N Reactor)—from decontamination of N Reactor; confined to tank AP-102
- Dilute complexed waste (5 percent; various sources)—high-organic liquids from the SSTs; includes solids comprising 10 percent of this waste type
- Neutralized cladding removal waste (4 percent; PUREX)—thick alkaline sludge, chiefly zirconium hydroxide
- **PFP sludge wash (0.7 percent; PFP)**—sludge from PFP recovery operations; confined to tank SY-102

According to the Westinghouse Hanford Company (WHC) Waste Tank Summary (Hanlon, 1996), most tanks contain only one of these waste types. Exceptions are: AW-103, AW-105, and SY-102, which contain neutralized cladding removal waste solids or PFP solids in addition to dilute noncomplexed waste; and SY-101 and SY-103, with both concentrated complexant and double-shell slurry.

¹There are differing definitions of concentrated complexant waste in the Hanford literature, but they appear to be compatible. The source definition used here—evaporation of dilute complexed waste which is itself derived from SSTs—is from Hanlon (1996) and appendix B in the TWRS EIS (U.S. Department of Energy, 1996b). Gephart and Lundgren (1995) and appendix L of the EIS (U.S. Department of Energy, 1996f) define concentrated complexant as being derived chiefly from Sr recovery operations in B Plant. Scheele et al. (1995) suggest that Sr recovery is the chief source of the organic complexants present in the tanks; thus, the definitions are compatible.

The chemistry of the solids and dissolved constituents of the DSTs is, like the SSTs, dominated by sodium nitrates and nitrites, with additionally 20 percent metal hydroxides and 10 percent phosphates, carbonates, oxides, and sulfates. Cs-137 comprises 72 percent of the DST waste radioactivity, while 27 percent is from Sr-90; this contrast with the SST proportions is due to the tendency for Sr to have settled out in the SST solids before waste transfer to the DSTs.

3.2 INVENTORY

Two approaches to tank inventory characterization have been employed, each complementing the other: direct sample measurement (or assay) and estimation based on facility records. The ongoing Hanford tank waste inventory effort combines both approaches: (i) analytical characterization work is being reported in a Tank Characterization Report (TCR) for each tank (e.g., Benar and Amato, 1996)—these reports include estimations of total tank inventories based on an informed combination of individual sample results; and (ii) historical characterization is reported in documents termed Historical Tank Content Estimates (HTCE), which are released for quadrant groupings of tank farms (e.g., Brevick et al., 1996) with reference to supporting summaries for each farm (e.g., Brevick and Newell, 1996). The HTCE reports summarize all available historical data on processing and waste transfers, and present the waste inventories based on those data. In terms of inventory characterization, the goal of Hanford's overall effort is to produce a unified "best-basis" inventory drawing on all available estimation and assay results. This work is still in progress, and the most recent results may be viewed on line at the PNNL Tank Waste Information Systems (TWINS) web site at

http://twins.pnl.gov:8001/TCD/main.html

(Permission for access to this database must be obtained from PNNL). This database also has all available assay data on tank waste samples, and should prove to be a valuable resource for ongoing tank waste familiarization.

The complex computational basis for HTCE inventories is being executed at Los Alamos National Laboratory (LANL). This effort, using what is termed the Hanford Defined Waste (HDW) model (Agnew, 1997), compiles historical process and waste transaction records in order to construct spreadsheets delineating time-dependent inventories of solid and liquid chemical inventories for each tank. In the HDW model, all possible sources of tank contents are classified among 48 different waste types, each with a given chemical/radionuclide profile based on knowledge of the processing from which it originated. Tank contents are then calculated from combinations of these waste types consistent with the historical records. The HDW estimation model for solids compositions is termed the Tank Layer Model (TLM) and that for liquids is called the Supernatant Mixing Model (SMM). The HDW model compiles estimates for 33 nonradioactive chemical species, 46 radionuclides (decayed to 1994), and four other properties (density, water weight percent, total organic carbon, and sludge void fraction). The radionuclide estimates are based on ORIGEN2 calculations for all the nuclear fuel batches processed at Hanford, with modifications for extraction and other processing.

The HDW total site inventories are shown in table A-1. Because the set of tables illustrating individual tank inventories would constitute 354 pages, we report here only a few selected tanks as

examples. These examples are shown in tables A-2 to A-7, which are reproductions from Agnew (1997). The example tanks were selected on the basis of the following (see chapters 2 and 4):

- A-101 is on both the Organic and Flammable Gas Watch-lists
- AW-104 has nitrate, nitrite, and hydroxide concentrations that make it particularly susceptible to pitting corrosion and stress corrosion cracking
- BY-106 is on the Ferrocyanide Watch-list
- C-106 is on the High-Heat Watch-list due to high fission product content
- SY-101 is a "burping" tank and is on the Flammable Gas Watch-list
- SY-102 is particularly high in Pu

The TWRS EIS (U.S. Department of Energy, 1996) used a different, preliminary set of data in reporting overall chemical and radionuclide inventories for SST and DST tanks. The EIS approach (Golberg and Guberski, 1995) was to use historical process records for estimation of SST inventories and measurement data augmented by historical data for the DSTs. Therefore, the methods employed for the EIS inventory were not fundamentally different from those in current use; the more recent historically based inventories are the products of further records research and more sophisticated modeling techniques, and their results are being critically compared with analytical data from all tanks. The EIS total inventories, not broken down by individual tank, are provided for comparative purposes in tables A-8 and A-9.² In the EIS (U.S. Department of Energy, 1996a), the argument is made that these total inventories, while not accounting for the considerable variations among tanks, are adequate for conceptual design of waste treatment options. The current Hanford inventory approach is compared with that utilized in the EIS in table 3-1. Comparison of inventory values is briefly discussed later in this chapter.

The general chemical characteristics of the SST and DST tank wastes were discussed in sections 3.1.1 and 3.1.2. In the following sections, the different chemical components of the waste—inorganic, organic, and radioactive—are separately addressed.

3.2.1 Inorganic chemicals

Table A-1 shows that, by far, the most abundant cation in the tank wastes is sodium; Na⁺ comprises around 80 percent of the cationic content by weight. Figure 3-2 shows the range of variation

²Note that there is a traceable discrepancy in DST soluble chemical components between the versions of table A-8 in the Golberg and Guberski report and the EIS itself; we report here the former, primary source. In EIS table A.2.1.2, values in the DSTs Soluble column from CrO_4^{2-} down to Zr^{4+} were erroneously shifted up one row.

Table 3-1. Comparison of inventory approaches for the TWRS Environmental Impact Statement and the ongoing inventory characterization effort ("Hanford Best-Basis")

Inventory	EIS	Hanford Best-Basis					
Coverage	site-wide only	Individual tank					
Approach	assay and records—DST records—SST	1	 Historical Tank Content Estimates—records; includes Hanford Defined Waste Model 				
		Best-Basis Inventory					
	The arrows signify that the Best-basis inventory is based on a comparative analysis of results of both Tank Characterization Reports and Historical Tank Content Estimates.						

in Na⁺ concentrations across all 177 tanks, based on the Revision 3 HDW model.³ The next most abundant cation overall is aluminum, with approximately 5 weight percent of the cationic inventory. There are relatively large concentrations of cations derived from construction materials: Fe^{3+} , Ni^{2+} , and Cr^{3+} and fuel claddings: Zr^{4+} and Al^{3+} [also presented as $Al(OH)_4^-$].

The anionic waste contents are not so dominated by a single constituent. The dominant anion, by weight, is nitrate (NO_3^-) at about 62 percent, and other abundant anions include hydroxide (OH^-) , nitrite (NO_2^-) , and carbonate (CO_3^{2-}) . However, a number of other anions such as phosphate (PO_4^{3-}) , CI^- , F^- , SiO_3^{2-} , SO_4^{2-} (table A-1) have significant concentrations and are important to waste chemistry. The EIS designates nitrate as the chief inorganic anion of significance to risk (U.S. Department of Energy, 1996b), because of its potential to oxidize ferrocyanide as well as organics in the tank, leading to explosion and radionuclide release. The range of variation in nitrate concentration among tanks based on the HDW revision 3 model is shown in figure 3-3.

3.2.2 Organic Chemicals

Interest in organic waste constituents arises from two considerations (Gephart and Lundgren, 1995; Turner et al., 1995). First, at elevated temperatures, organic compounds can combine with the abundant oxidizing materials in the waste, chiefly nitrates and nitrites, in exothermic reactions that pose risks of fire and/or explosion. (This issue is discussed in more detail in chapter 4.) Second, organic complexants can bind with waste constituents (e.g., radionuclides) and affect their chemical behavior during waste treatment processes. The overall EIS inventories (table A-8) report organic components only as total organic carbon (TOC). Ongoing individual tank inventory efforts such as HDW provide more detailed delineation of organic compound contents by tank (e.g., tables A-2 to A-7) and overall for the

³Note that revision 3 data are used in the histogram plots of figures 3-2, 3-3, and 3-4 because of the availability of the CNWRA database derived from revision 3 (Agnew, 1996). Electronic files of revision 4 concentration data are not readily available; the online TWINS database lists only total amounts in kg or Ci, rather than in concentrations. The differences between revisions 3 and 4 are not important for the purposes of these illustrative plots.

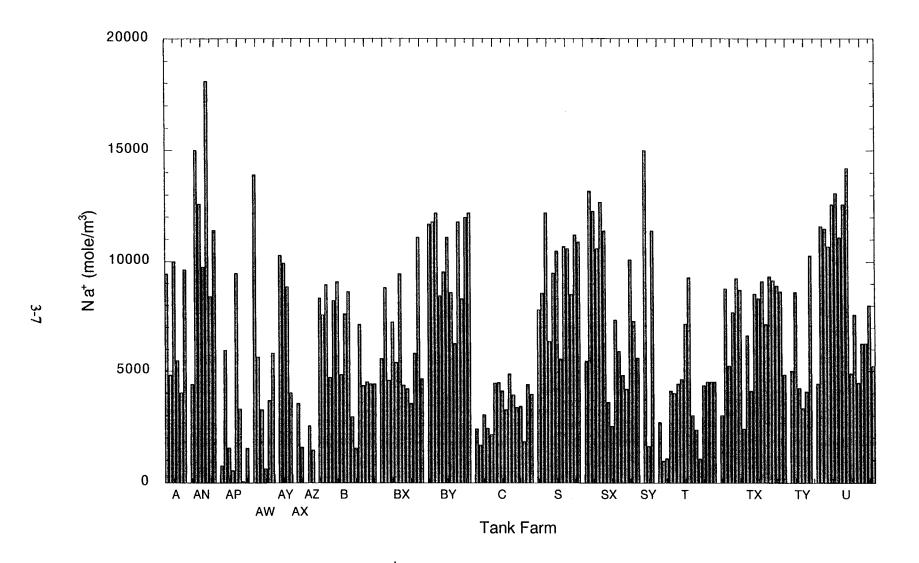


Figure 3-2. Bar chart of moles/m³ Na⁺ in Hanford tanks from the Hanford Defined Waste model, revision 3 (Agnew, 1996). Tank farms are separated by blank spaces along the x axis.



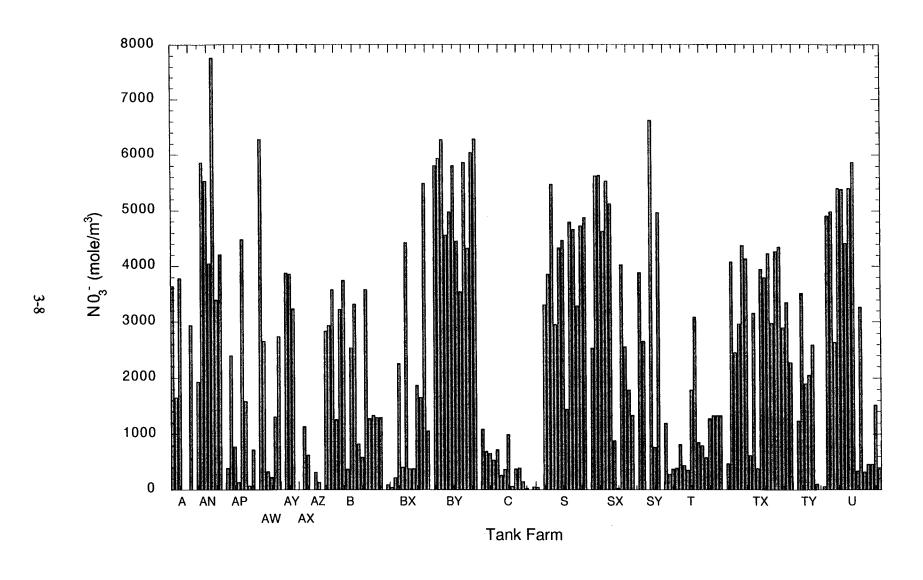


Figure 3-3. Bar chart of moles/m³ NO₃ in Hanford tanks from the Hanford Defined Waste model, revision 3 (Agnew, 1996)



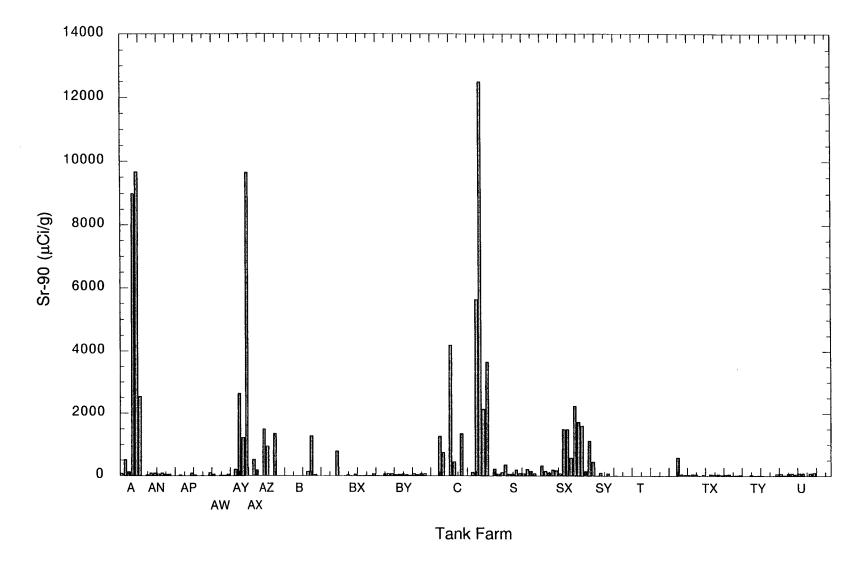


Figure 3-4. Bar chart of μ Ci/g Sr-90 in Hanford tanks from the Hanford Defined Waste model, revision 3 (Agnew, 1996). Many values are too small to be visible.

Hanford site (table A-1). The overall site inventories (table A-1) show that, on a molar basis, glycolate is the predominant organic complexant. The other listed organic anions—citrate, ethylenediaminetetra-acetic acid (EDTA), N-(2-hydroxyethyl)ethylenediaminetetra-acetate (HEDTA), acetate, oxalate, DBP, and butane—all have similarly low molar concentrations, ranging from approximately 1/20 to 1/4 of the total site glycolate value. On a weight basis, glycolate is rivaled in abundance by HEDTA. There are no major differences in organic inventories between SSTs and DSTs.

3.2.3 Radionuclides

The key radionuclides for risk assessment at Hanford site are C-14, Sr-90, Tc-99, I-129, Cs-137, and U (U.S. Department of Energy, 1996b). Table 3-2 lists the total tank HDW inventories for these radionuclides on a quadrant basis. Plutonium is added to the table because it is the key element for criticality hazard assessment (see section 4.2.5). More information on HDW total radionuclide inventories is available in table A-1, and EIS values are listed in table A-9. As mentioned above, Sr-90 and Cs-137 are by far the most abundant radionuclides on an activity basis. The daughters of Sr-90 and Cs-137—Y-90 and Ba-137m, respectively—are at or near a state of transient radioactive equilibrium, that is, equal radioactivity, with their parents and should be included in the inventory. However, both of these daughters are sufficiently short-lived that they decay away in a matter of days when separated from their parents. Figure 3-4 indicates the wide variability of Sr-90 concentrations in the tanks. Such large inter-tank variability is also noted in other tabulated radionuclides (tables A-2 to A-7).

Of the six key radionuclides listed in the EIS, C-14, Sr-90, I-129, and Cs-137 are considered to present exposure hazards during remediation (U.S. Department of Energy, 1996). Although the half-lives of Sr-90 and Cs-137 are short—29 and 30 yr, respectively—relative to the others, they are still long enough that these two radionuclides will remain the dominant sources of radioactivity exposure and heat generation hazard during waste retrieval and solidification. Consideration of longer term risk centers on those radionuclides—C-14, I-129, Tc-99, and U isotopes—which are deemed mobile in groundwater and have sufficiently long half-lives $(5 \times 10^3 \text{ to } 4 \times 10^9 \text{ yr})$ to persist well into the future. The longer term risk pertains to on-site storage of waste forms prior to disposal in a geologic repository and residual waste in the tank following remediation.

Table 3-2. Total HDW inventories of key radionuclides in Hanford tanks, listed by quadrant (modified from Table A-1). Values are in curies, with the exceptions of U and Pu (kg). Although Pu-239 and Pu-241 are the only fissile Pu isotopes, they comprise more than 95 percent of waste Pu by mass. Therefore, total Pu is reasonably representative of the relative fissile Pu content.

Quadrant	C-14	Sr-90	Tc-99	I-129	Cs-137	U (kg)	Pu (kg)
NE	8.4×10^{2}	2.3×10^{7}	5.3×10 ³	1.0×10 ¹	6.7×10 ⁶	1.1×10 ⁶	2.7×10^{2}
sw	1.5×10 ³	1.9×10 ⁷	1.1×10 ⁴	2.0×10 ¹	1.3×10 ⁷	6.2×10 ⁵	1.4×10^{2}
NW	4.1×10^{2}	1.9×10 ⁶	2.9×10^{3}	5.6×10°	4.5×10 ⁶	4.9×10 ⁵	1.2×10 ²
SE	2.1×10^{3}	1.8×10 ⁷	1.4×10 ⁴	2.7×10 ¹	2.3×10 ⁷	1.9×10 ⁵	2.5×10^{2}

3.2.4 Discussion

The nature of the tank waste inventories—chiefly their derivation in large part from reconstructions of waste histories—precludes attaching a large degree of certainty to any particular inventory estimation scheme without consideration of the times considered and the methods employed. For example, distribution of Cs-137 between SSTs and DSTs can be appreciably affected by pumping liquids from the former into the latter, which is an ongoing activity. Furthermore, it is projected that the 74,200 m³ (20 million gal.) of waste in the DSTs will be augmented by another 12,400 m³ (3.3 million gal.) during future waste transfers; no calculations of the effects on tank inventories were noted in the literature. The EIS (U.S. Department of Energy, 1996b) does note that the added wastes will be dominated by dilute noncomplexed waste types (see above). Note also that accurate radionuclide inventories require dates of calculation to account for decay; however, the five-year spread in dates used in the studies cited herein results in less than a 13 percent difference in Sr-90 and Cs-137 contents.

It has been noted by nearly all studies cited in this chapter that individual tank inventories have the highest degree of uncertainty. Agnew (1997) calculated estimated uncertainties in concentrations based on variability in knowledge of process and solubilities. The resultant variabilities are shown for individual tanks in tables A-2 to A-7. There is a wide range in relative variabilities, ranging up to nearly 100 percent of reported concentrations, but most appear to be in the range of 10 to 50 percent. A higher degree of confidence in individual tank inventories is the goal of the Hanford "best-basis" effort, which as noted is still incomplete. This effort will unify results from the HTCE/HDW estimation scheme with analytical data on the wastes themselves. See table 3-3 for a "grab bag" example of how divergent analytical data may be from inventory estimation for tank contents of major constituents. Note the rather good agreement for sodium and Cs-137, but the factor of two difference in nitrate and factor of five difference in Sr-90. Best-basis values must be critically reviewed when they are finalized.⁴

Table 3-3. Comparison of assay or Tank Characterization Report and records or Historical Tank Content Estimate (which incorporates Hanford Defined Waste results) inventory approaches for selected constituents in Tank SY-101. Data from TWINS online database and table A-6.

Constituent	HTCE/HDW	TCR
Na ⁺ (kg)	1.77×10 ⁶	1.39×10 ⁶
NO ₃ ⁻ (kg)	1.69×10 ⁶	8.50×10 ⁵
Sr-90 (Ci)	6.83×10⁵	1.31×10 ⁵
Cs-137 (Ci)	1.54×10 ⁶	2.31×10 ⁶

⁴Agnew (1997) notes that direct comparison of HDW values with assays for a given tank are not generally likely to be useful. He says instead that the comparison should be made among groups of tanks with similar waste histories (he does not, however, provide a list of such groups).

While the overall site tank waste inventories for constituents are subject to less uncertainty, significant differences emerge from one estimation scheme to another. (Note that TCR total site inventories are not yet available.) A comparison is made in the EIS (U.S. Department of Energy, 1996b) using an earlier HDW which does not generally differ markedly with the revision 4 version cited here. It is noted there that some constituents are listed at contents several times higher in the HDW than in the Golberg and Guberski (1995) WHC report. The EIS authors state that it is not possible, considering model complexities, to easily explain the source of these differences. It would seem, then, that use of the generally higher HDW inventories would be more conservative. An important exception, noted in the EIS, is nitrate, which is about twice as high in the Hanford overall inventory as in the HDW, and is potentially significant as a post-remediation pollutant (U.S. Department of Energy, 1996b). Table 3-4 compares the total site tank EIS values for selected major constituents with those from the more recent HDW revision 4 model (Agnew, 1997). Agreement is quite good for three of the four constituents, but the large difference in nitrate estimates persists.

In summary, the best tank waste inventory (i.e., the "best-basis" model currently under development) is not yet complete. Preliminary results are available at the online TWINS database site. Until completion of this inventory, use of the HDW model (Agnew, 1997) is generally preferred for constituents listed therein (table A-1 and example tables A-2 to A-7).

Table 3-4. Comparison of total site tank inventories as determined by Golberg and Guberski (1995) for the Environmental Impact Statement (EIS) and in the HDW revision 4 (Agnew, 1997) for selected constituents. EIS values are from tables A-8 and A-9, with values from the former converted from metric tons to kg. HTCE/HDW values are the "All Tanks" values in table A-1.

Constituent	EIS	HTCE/HDW
Na ⁺ (kg)	6.91×10^7	4.92×10 ⁷
NO ₃ (kg)	1.07×10 ⁸	5.04×10 ⁷
Sr-90 (Ci)	5.37×10 ⁷	6.16×10 ⁷
Cs-137 (Ci)	3.49×10^7	4.71×10 ⁷

4 HAZARDS POSED BY WASTE TANKS AND TANK WASTE REMEDIATION SYSTEM

4.1 PRIMARY HAZARDS ASSOCIATED WITH HANFORD WASTE TANKS

A number of safety issues associated with Hanford waste tanks have been identified by the DOE. Of primary importance, particularly with respect to the regulatory role of the NRC, are those having the potential for releasing radioactivity to the environment. The DOE developed a set of criteria to identify tanks with potential safety concerns as Watch-list tanks.¹ The four different Watch-list categories are flammable gas, ferrocyanide, high organic content, and high-heat generation. Information indicates that there are 50 tanks (44 SSTs, 6 DSTs) on the Watch-list (Hanlon, 1996), with 10 tanks listed in more than one of four different Watch-list categories. The safety issues associated with these Watch-list categories are discussed in the following sections, and the Hanford waste tanks identified for each Watch-list are given in table 4-1.

4.1.1 Flammable Gas Safety Issue

The risk associated with the release of flammable gases into the dome space of waste tanks at the Hanford site is a top priority safety issue (McDuffie, 1995). Although flammable gas production from radiolysis is always a concern for high-level radioactive waste storage, a special problem developed at the Hanford site when wastes were concentrated by evaporation to generate additional storage space in the million-gallon waste tanks. The volume of the slurry concentrate slowly increased due to retention of generated gases after being pumped into the tanks, which defeated the purpose of volume reduction. The real problem became evident when some tanks began to have rather large surface level drops accompanied by release of gas mixtures containing both fuel (hydrogen) and oxidant (nitrous oxide). These gas mixtures are flammable and potentially explosive even if not mixed with the oxygen in the ambient air. Tank SY-101, prior to installation of a mixer pump, exhibited the largest cyclic releases (as indicated by tank surface level drop and increase in tank pressure), and hydrogen concentrations in the tank dome space and ventilation header have exceeded the lower flammability limit (LFL)² for short periods of time (McDuffie, 1994). The presence of flammable concentrations of gases and an ignition source could lead to reactions that could cause a radioactive release or provide an energy source that

¹A separate but related formal administrative DOE program is in place to identify as an USQ known or suspected operating conditions that have not been analyzed or that fall outside of the established authorization bases. Following identification of a USQ, a review is conducted and corrective action is taken if applicable. The USQ may be closed from an administrative standpoint, which means that conditions surrounding the safety issue have been analyzed, although the safety issue may still exist and may require mitigation, controls, or corrective action. The safety issues that were identified under the Watchlist program were also previously analyzed as USQs. Technical evaluation has resulted in closing the USQs on ferrocyanide, floating organic layer, and criticality (U.S. Department of Energy, 1996a). There is a USQ associated with the Flammable Gas Watch-list tanks because of the potential consequences of a radiological release resulting from a flammable gas burn, an event not analyzed in the SST Safety Analysis Report. Hanlon (1996) reported that DOE declared a USQ on some tanks containing dry organic nitrate chemicals because methods for analyzing accident scenarios have become available for these.

²The lower and upper limits of flammability indicate the percentage of combustible gas in air below which and above which flame will not propagate. When flame is initiated in mixtures having compositions within these limits, it will propagate and therefore the mixtures are flammable (Avallone and Baumeister, 1996). Lower and upper limits of flammability for hydrogen are 4.0 and 75.0 vol.%, respectively. For ammonia, lower and upper limits are 15.0 and 28.0 vol.%, respectively. For methane, the lower and upper limits are 5.0 and 15.0 vol.%, respectively.

hydrophobic surfaces, capillary channel gas accumulation, mechanical trapping of crystal clusters, and tight engulfment in bubbles attached to solid particles (McDuffie, 1995). Retention of gas within the waste appears to present a greater problem than gas generation, which would not be a problem if the tank ventilation can successfully remove the gases from the tank dome space. Gas retention or accumulation, on the other hand, can result in a serious situation if it leads to a sudden release of large inventories of gases, such as those which have occurred in tank SY-101 and, more recently, in tank SY-103 (Hanlon, 1996). In the latter case, hydrogen gas concentration increased from a 60 ppm baseline to 500 ppm over two days, then subsequently increased from 470 to 1720 ppm within one minute. The LFL for hydrogen is 40,000 ppm and the safety criterion for dome space is 10,000 ppm.

Because there is inadequate information regarding tank waste processes that generate, retain, and release flammable gases, efforts are ongoing at Hanford to collect information about the basic chemical and physical properties of the tank wastes. This information is needed to gain knowledge about the behavior of the waste so that effective mitigation methods can be developed and implemented. Mitigation methods may involve mechanical processes, chemical treatment, or a combination of both. Tank SY-101 is currently being mitigated by using a mixer pump to stir the waste and allow hydrogen gas to be released gradually and prevent episodic releases that are above the LFL. The pump is operated for 25 min three times a week.⁴ Other tanks are being screened and evaluated to assess the magnitude of their risk from flammable gas generation, retention, and intermittent release. Gas monitoring systems are also being installed that will provide continuous monitoring of hydrogen and periodic monitoring of other gases. In addition, efforts are under way to upgrade instruments for surface level and temperature measurements.

Recently, all 177 tanks (Watch-list and non-Watch-list) were placed under flammable gas controls, which means that flammable gas may exist in all 177 tanks and special safety measures will be taken during maintenance, monitoring, and waste transfer activities (Hanlon, 1996; U.S. Department of Energy, 1996a). Final resolution of how many tanks present a risk due to flammable gas has not occurred.

4.1.2 Organics Safety Issue

A variety of organic compounds were used at the Hanford site during fuel reprocessing, metal recovery operations, and waste management operations. The principal sources for the majority of the organics were the solvent extraction processes that were used to recover Pu and U, which include Uranium Recovery, PUREX, and REDOX processes, and the waste management operations which involved removal of Cs-137 and Sr-90 from the wastes to improve the safety of radioactive waste storage. The major organics added to the tanks as a result of these operations include the solvent tributyl phosphate (TBP, 30 vol%) in a normal paraffin hydrocarbon (NPH) diluent, the radiolytic degradation products of TBP [dibutyl phosphate (DBP) and butanol], di-2-ethylhexyl phosphoric acid (D2EHPA), sodium ethylenediaminetetra-acetate (EDTA), sodium hydroxyacetate. Estimated quantities of organic chemicals used at Hanford are listed in table 4-2.

In addition to the organics, wastes contain large amounts of sodium nitrate and nitrite, with the nitrite arising principally from radiolysis of nitrate. Since these organic-bearing wastes are mixtures of organic fuels, strong inorganic oxidants, and heat-producing radionuclides, the potential exists for rapid

⁴ Wodrich, D. 1997. U.S. Department of Energy. Personal communication, January 14, 1997.

energetic reactions that could result in radioactive release to the environment. Such a reaction resulted in a major explosion in a radioactive waste tank in Kyshtym, Russia, in 1957 (Medvedev, 1979) resulting in radiation contamination of an estimated 23,000 sq km. The Kyshtym explosion occurred when the tank cooling system failed and the radioactive decay heat raised the temperature of a sodium acetate-sodium nitrate radioactive waste mixture to the point at which a thermal-runaway reaction occurred between acetate and nitrate. Fisher (1990) evaluated Hanford tank waste data and available reactivity data with respect to the Kyshtym accident and concluded that the temperatures of Hanford tank wastes are well below those required to initiate reactions between sodium acetate and sodium nitrate/nitrite. The organic chemical safety issue associated with the Hanford site and TWRS operations is the possibility of local ignition sources or radioactive decay heat that could lead to exothermic reactions between the heated organic waste components and oxidizing salts (e.g., sodium nitrate and sodium nitrite) under conditions of low moisture.

Based on reviews of waste transfer records (Babad and Turner, 1993) and available sampling data (Webb et al., 1995), 36 tanks were considered to possibly contain greater than 3 wt% TOC on a dry-weight basis (or 480 J/g of exothermic energy), which is the minimum fuel concentration considered necessary to support a propagating reaction based on empirical data (Fisher, 1990). Those tanks had controls put in place to prevent propagating reactions (Westinghouse Hanford Company, 1995) and were placed under the scope of the DOE Data Quality Objectives⁵ (DQO) to Support Resolution of the Organic Complexant Safety Issue (Turner et al., 1995). The tanks were evaluated using criteria described in the DQO to determine whether: (i) the wastes have enough fuel to support a propagating reaction when dried, (ii) enough moisture is present in the wastes to prevent a propagating reaction, and (iii) the wastes have the potential to dry during interim storage. Of the 36 tanks, only 20 are still in a recent (July 31, 1996) High Organic Watch-list (Hanlon, 1996). These tanks are listed in table 4-1.

If the waste has sufficient fuel-nitrate mixture and low moisture content and energy source raised the temperature to the ignition point, a propagating reaction could be initiated. A variety of ignition sources were evaluated (Westinghouse Hanford Company, 1985). All of the potential initiators (e.g., vehicle operation above the tanks) would occur near the waste surface with the exception of rotary core drilling and lightning strikes. Proper safety interlocks to limit drill bit temperature rise and grounding of tanks can mitigate these events. The temperature of the waste in these tanks is either monitored continuously or measured manually on a weekly basis. The tanks are also checked for the presence of entrained or floating organic layers that might pose a risk from a slow pooled or wicked fuel burn. Studies are also under way to gain a better understanding of high organic safety issues. Current characterization efforts are focused on testing tank waste samples to confirm that the current safe storage criteria (i.e., fuel energy value, TOC, moisture content) for tank wastes are conservative for actual waste

⁵ The DQO Process, defined by the EPA, is a series of planning steps to identify and design more efficient and timely data collection programs. It provides a systematic procedure for defining the criteria that a data collection design should satisfy, including when and where to collect samples, the tolerance level of decision errors for the study, and how many samples to collect. It is the policy of the DOE Office of Environmental Management (EM) to apply up-front planning, where practical, to ensure safer, better, faster, and cheaper environmental sampling and analysis programs for all EM projects and operations (memo from Thomas P. Grumbly, Assistant Secretary for Environmental Management, September 7, 1994). Specifically, it is EM policy that the DQO process be used in all environmental projects where there may be a need to collect significant environmental data. The EPA "Guidance for the Data Quality Objectives Process" (U.S. Environmental Protection Agency, 1994a,b) provides excellent guidance on the steps of the DQO process for developing data quality criteria and performance specifications for data operations.

(U.S. Department of Energy, 1996c). Waste from selected tanks will be tested for reaction propagation using an adiabatic calorimeter.

4.1.3 Ferrocyanide Safety Issue

During the 1950s, additional tank storage space for high-level radioactive waste from defense operations was generated using precipitation processes for scavenging Cs and other soluble radionuclides from tank waste liquids. In the Cs-137 scavenging processes, waste solutions were adjusted to a pH between 8 and 10, and sodium or potassium ferrocyanide and nickel sulfate were added to coprecipitate Cs with the insoluble alkali-metal nickel ferrocyanide. Because the waste solutions had high nitrate and radiolytically produced nitrite concentrations, these ions became incorporated into the precipitates. After allowing the radioactive precipitates to settle, the decontaminated solutions were pumped to disposal cribs, thereby providing additional tank storage volume. Later, some tanks were found to be leaking; pumpable liquids were removed from these tanks, leaving behind a wet solid (sludge) residue containing the ferrocyanide precipitates (Burger et al., 1991). In implementing the scavenging process, approximately 140 metric tons (154 tons) of ferrocyanide [calculated as Fe(CN)₆] were added to waste that was later routed to 18 Hanford site SSTs.

The explosive nature of ferrocyanides in the presence of oxidizers has been known for decades, but the conditions under which impure mixtures of ferrocyanide, nitrate, and nitrite can undergo propagating reactions had not been thoroughly studied. The potential reactivity of these mixtures was first recognized at the Hanford site when the Cs-137 scavenging process using ferrocyanide was investigated for application to radioactive wastes produced by the next generation processing technology. The investigation found that cesium zinc ferrocyanide and nitrate exploded when heated (Hepworth et al., 1957). In the laboratory, mixtures of ferrocyanide and oxidants, such as nitrates and nitrites, have been shown to undergo energetic reactions when heated to high temperatures (above 250 °C) or exposed to an electrical spark of sufficient energy to heat the mixture (Cady, 1993; Epstein et al., 1994). Because the scavenging process precipitated ferrocyanide from solutions containing nitrate and nitrite, an intimate mixture of ferrocyanides and nitrates and/or nitrites is likely to exist in some regions of the ferrocyanide tanks. Despite the fact that the measured temperatures in the Hanford waste tanks continue to drop, there has been speculation as to the possibility of "hot spots" forming in the tanks from radiolytic heating.

Efforts have been under way since the mid-1980s to evaluate the potential for ferrocyanide reactions in Hanford site SSTs (Burger, 1984; Burger and Scheele, 1990; Meacham et al., 1995). The 1987 EIS (U.S. Department of Energy, 1987) included an environmental impact analysis of potential explosions involving ferrocyanide-nitrate mixtures. The EIS postulated that an explosion could occur during mechanical retrieval of saltcake or sludge from a ferrocyanide waste tank. The EIS concluded that this worst-case accident could create enough energy to release radioactive material to the atmosphere through ventilation openings, exposing persons offsite to a short-term radiation dose of approximately 200 mrem. A General Accounting Office study (Peach, 1990) postulated a greater worst-case accident, with independently calculated doses of one to two orders of magnitude greater than postulated in the DOE EIS.

Three different flowsheets (and variations of them) were used in ferrocyanide waste scavenging campaigns. Approximately 66 percent of the total ferrocyanide used at the Hanford site was used in the U-Plant flowsheet, which treated "metal waste" dissolved in nitric acid after the U had been recovered using the tributyl phosphate process. Simulant sludge produced by this flowsheet contained approximately 8.3 wt% sodium nickel ferrocyanide on a dry basis. The T-Plant flowsheet, used to treat first-cycle waste

from the BP process, consumed approximately 8 percent of the ferrocyanide used at the Hanford site, and simulant sludge produced by this flowsheet contained 8.8 wt% sodium nickel ferrocyanide. The In-farm flowsheet, which treated the basic waste from recovery of U, consumed approximately 26 percent of the ferrocyanide used at Hanford and produced sludge containing up to 25.8 percent sodium nickel ferrocyanide (Postma and Dickinson, 1995). A more detailed review of ferrocyanide waste production is presented by Postma et al. (1994) and by Jeppson and Wong (1993).

Reviews of process flowsheets and waste transfer records (Borsheim and Simpson, 1991) indicated that eighteen tanks received ferrocyanide waste. These tanks were placed under the scope of the DQO on Ferrocyanide Safety Issue (Meacham et al., 1995) for further evaluation using criteria described in the DQO. The Ferrocyanide Safety Program was implemented in 1990 to address this safety issue (Bryan et al., 1995) and comprised four major components. The first, tank monitoring, involves developing, deploying, and maintaining instrumentation for continuous monitoring of the tank contents. Specifically, waste temperatures in the tanks are being monitored continuously to detect increasing temperature trends. The second program component, modeling and analyzing existing tank data, allows predictive calculations of, for example, the existence of hot spots within the waste or concentrations of gases within the tank dome space. Ferrocyanide waste characterization using waste simulants and actual tank samples is the third program component and focuses on the chemical analysis (e.g., fuel, moisture, and nickel concentrations) of gas space, surface samples, and core samples from the ferrocyanide tanks. The fourth component is research and development designed to provide an understanding of potentially hazardous reactions of precipitated ferrocyanides and their aging products within the SST ferrocyanide waste.

Four of the 18 tanks that received ferrocyanide waste (tanks C-108, C-109, C-111, and C-112) were classified as safe based on criteria described in the DQO on Ferrocyanide Safety Issue and were removed from the Ferrocyanide Watch-list in June 1996 (Hanlon, 1996). As of July 31, 1996, fourteen tanks remained on the Ferrocyanide Watch-list (Hanlon, 1996) and are listed in table 4-1. These tanks contain > 8 wt% sodium nickel ferrocyanide on an energy equivalent basis but meet conditionally safe criteria established in the DQO which preclude sustainable, rapid exothermic ferrocyanide reactions (Hanlon, 1996, table A-2 footnote). However, because the ferrocyanide sludge has been exposed for many years to other highly caustic wastes, as well as to elevated temperatures and both gamma and beta radiation, DOE investigators believe that ferrocyanide decomposition may have occurred in the tanks which would lead to ferrocyanide concentrations much less than that predicted by tank inventory records. Tank waste samples that have been analyzed to date support the conclusion that ferrocyanide decomposition has occurred and that the sludge in the Ferrocyanide Watch-list tanks is too dilute to support a sustained reaction, even if dried out and ignited (Postma and Dickinson, 1995). This conclusion has recently been accepted by the DOE.⁶ All tanks have been removed from the Ferrocyanide Watch-list and the Ferrocyanide Safety Issue was officially closed as of October 1996.

4.1.4 High-Heat Safety Issue

Radioactive decay of stored waste can result in elevated temperatures of Hanford tanks. If waste tank structural damage occurs due to overheating of the waste tank concrete structure, release of high-level nuclear waste may occur. According to Hanlon (1996), 10 SSTs have high-heat loads [>42,000 kJ/h (>40,000 Btu/h)] namely: A-104, A-105, C-106, SX-107, SX-108, SX-109, SX-110, SX-111, SX-112, and SX-114. All of these tanks are on active ventilation except for A-104 and A-105.

⁶ Kinzer, J. 1996. U.S. Department of Energy. Reported in Tri-City Herald, October 31, 1996.

Tank C-106, the only tank on the High-Heat Watch-List at present (Hanlon, 1997), requires more than active ventilation to keep the temperature below 150 °C (300 °F), which is the maximum temperature limit established in the DOE Operating Safety Document (Wodrich, 1992). The rate of heat generation in tank C-106 is estimated at more than 105,000 kJ/h (100,000 Btu/h) and arises primarily from radioactive decay of Sr-90 waste that was transferred into the SST in the late 1960s. For this tank, water is periodically added to maintain a liquid cover (supernate) over the liquid sludge for enhanced thermal conductivity and evaporative cooling (DeFigh-Price and Wang, 1993). The amount of cooling liquid currently maintained in tank C-106 exceeds the interstitial holdup of the tank sludge; the excess cooling liquid is a primary concern because it could release radionuclides to the surrounding soil and groundwater if a tank leak develops. Although the method of active ventilation supplemented by water addition is effective for the short term, the long-term resolution for tank cooling is removal of the heat-generating waste in the tank. This solution is being pursued as the only remediation method for this safety issue, and tank C-106 has been selected as the first SST for retrieval and transfer of radioactive waste to a selected DST. Sluicing of tank C-106 is scheduled to begin in 1997.

4.2 OTHER HAZARDS

4.2.1 Crust Burn Issue Associated with Flammable Gas Tanks

In addition to the potential for ignition of flammable gases such as hydrogen/air and/or hydrogen/nitrous oxide, as discussed in section 4.1.1, another scenario of significant concern associated with the tank wastes is the potential for secondary ignition of organic-nitrate/nitrite mixtures in the crust layer initiated by the burning of flammable gases or by a mechanical in-tank energy source. This scenario has been called a "crust burn" issue. Crust heating by a burning gas or by mechanical energy (e.g., from friction during core sampling) could initiate an exothermic reaction between organic carbon and the nitrate or nitrite compounds. If the crust material gets too hot, volatile components could be released into the atmosphere as aerosols which could entrain and release radionuclides to the environment.

The crust burn problem was first evaluated for tank SY-101 based on visual observations of the waste surface with a television camera, chemical analyses of crust samples, and calorimetry tests of waste samples. Results of crust analyses and analytical modeling of crust heating were used to show that a "crust burn" was not a safety issue for tank SY-101 (Fox et al., 1992). Sampling activities have been scheduled for other tanks on the Flammable Gas Watch-list (Johnson, 1994). Primary data needed to determine the potential for a crust burn of the waste material are derived from calorimetry tests, including differential scanning calorimetry (DSC) which involves heating small samples at a programmed rate, by measuring differential temperatures between the sample and a reference chamber. The heat flow into or out of the sample is used to determine (i) whether an exothermic reaction exists, (ii) the temperatures required for it to occur, and (iii) the net amount of heat produced. A relatively recent characterization report by Baldwin et al. (1995) concluded that DSC measurements on crust samples from tank AW-101 show exotherms in nearly every subsample, but none of the observed exotherms exceeded the 586 J/g threshold set forth in the DQO on the crust burn issue (Johnson, 1994).

4.2.2 High-Efficiency Particulate Air Filter Blow-Out Issue Associated with Flammable Gas Tanks

Another scenario associated with the presence of flammable gas mixtures in waste tanks that needs to be addressed is that of a pressure pulse which can occur even without ignition of the gas

(e.g., sudden release of gas accumulated in the waste). The HEPA filters on the tanks have an operating limit of +2.5 kPa (+10 in. of water) (McDuffie, 1995). If the gas pressure exceeds this value the filter seal could be breached and there would be an open pathway for release of radionuclides to the environment. Studies are under way to better understand mechanisms of gas accumulation and release in tank wastes. Plume burn analyses are also ongoing to determine the size of flammable gas release which can burst a HEPA filter upon ignition (McDuffie, 1995).

4.2.3 Organic Solvent Safety Issue

Various separation processes employed at the Hanford site involved the use of organic solvents which were inadvertently and/or purposely sent to the waste tanks. Subsequent waste transfer operations also distributed organic solvents among several of the Hanford tanks. The potential hazards associated with organic solvents are (i) contributing to headspace flammability (as discussed in section 4.1.1), (ii) igniting an organic solvent pool, and (iii) igniting an organic solvent that is entrained in waste solids.

Currently, one tank (C-103) is known to contain an organic solvent pool. Current characterization efforts include continued vapor sampling of the tank headspace to identify additional tanks that may contain an organic solvent pool or entrained organic solvent. If vapor sampling suggests the presence of organic solvent, liquid grab samples and/or near-surface samples will be obtained to better quantify the potential for an organic solvent fire.

4.2.4 Known and Assumed Leaking Tanks

Liquid waste from past tank leaks has resulted in vadose zone contamination beneath the leaking tanks and may be adversely affecting the groundwater in the vicinity of the tanks. As mentioned in section 2, potential groundwater impacts are currently being investigated as part of the RCRA Groundwater Assessments for the T Farm Waste Management Area and will be ongoing soon for the S-SX and B-BX-BY Waste Management Areas (U.S. Department of Energy, 1996d).

Leak monitoring is ongoing for the 177 waste tanks, and reports on waste inventory and surveillance are released monthly and quarterly. The report for the month ending July 31, 1996 (Hanlon, 1996) indicated that 67 of the 149 SSTs are assumed leakers. There are no reported leaks from the 28 DSTs. Table 4-3 provides a list of tank identification number, date at which the tank was declared a leaker, estimated leak volume, estimated activity of leak, and date the tank was interim stabilized. The leak volume ranges from approximately 1,300 L (350 gal.) from tank C-204 in the 200-East Area to 436 m³ (115,000 gal.) from tank T-106 in the 200-West Area. Estimates of total leak volume from all 67 assumed leakers range from 2,300 m³ to 3,400 m³ (600,000 to 900,000 gal.). To minimize further releases to the environment, the DOE removed all SSTs from service in 1980 and initiated a program to transfer all pumpable liquid into DSTs and stabilize the SST tank wastes until final disposition. This effort, known as interim stabilization, is currently ongoing. Interim stabilization has been completed on all but five assumed leaking tanks. All SSTs (including nonleaking) are expected to be interim stabilized by the year 2000 (U.S. Department of Energy, 1996d).

4.2.5 Criticality

In the DOE Final TWRS EIS, it is stated that:

"Of the actions evaluated in the Final Safe Interim Storage EIS, only the retrieval of solids from tank SY-102 was affected by the technical uncertainties regarding criticality." (U.S. Department of Energy, 1996c; p. E-12).

As a result, the DOE has suspended retrieval of wastes from this tank, possibly for transfer into a DST, pending the outcome of a criticality safety evaluation process outlined for the Defense Nuclear Facility Safety Board. Based on these statements, it could be assumed that the only significant risk of criticality known at this time is from retrieval of wastes from tank SY-102.

In order to determine if the statements made in the preceding paragraph accurately depict the criticality potential of other Hanford TWRS tank wastes relative to tank SY-102, a survey was performed to obtain information about the fissile nuclide content and criticality potential of other tanks. A report by Perry et al. (1994) contains information about the Pu-239 content and criticality potential of tank SY-101, a tank that one might expect to also have nontrivial criticality concerns. In this report, the authors used core sample data and the S_N^7 code ONEDANT to calculate the k_∞^8 corresponding to the Pu concentrations in sedimentary layers of this tank. They found that these layers have a k_∞ of about 0.012 percent and that an increase in the Pu-239 concentration by a factor of about 16,000 is necessary before the sediments would approach criticality. The analyses seem credible considering that the total tank inventory of Pu-239 in this tank was estimated to be 910 g (Perry et al., 1994) and the minimum critical masses of Pu-239 are 9,800 g for pure Pu metal, 900 g for Pu metal and light-water-moderated bare spherical reactor, and 320 g for a light-water-moderated and reflected spherical reactor (Knief, 1992).

Since the publication of Perry et al. (1994), additional information about the contents of the tanks has become available (Agnew, 1996). As discussed in section 3.2, the Agnew (1996) report estimates the total inventory of Pu-239 in tank SY-101 to be about 3,580 g and the total inventory of U-235 to be about 89,500 g, implying that the major criticality concern with the tank wastes in SY-101 may be the U-235 concentration and not the Pu-239 concentration.

To determine if there is a risk of a U-based criticality in the tanks, the tanks were ranked based on the fissile enrichment of the U which they contain and it was found that tank AW-104 contains U with the highest enrichment at 0.866 percent. The calculated U enrichments assume that the tank inventories of U isotopes listed in Agnew (1996) are accurate. In some cases, these isotopic inventories were calculated using the ORIGEN2 code (Croff, 1980). Calculations of k_{∞} were performed using the MCNP Version 4A code. The results indicated that even with optimal moderation in a pure, homogeneous, U-light water system, k_{∞} is only 0.94 for this system with U at an enrichment of 0.866 percent. The calculations used ENDF/B-VI cross-section files and the optimal relative abundance of UO₂ to water molecules was 0.43 (with a corresponding "solution" density of 4.8 g/cm³). This value of k_{∞} implies that U at the assumed enrichment would have a net poisoning effect on a critical system and that a U-based criticality is not possible. Also, adding U with the enrichments found in the Hanford tanks

 $^{^{7}}$ S_N theory discretizes the Boltzman equation for neutron transport in solid angle as well as space to find the eigenvalue of the system (k_{eff}).

⁸ k_∞ is the neutron multiplication factor for a material with infinite extent, that is, neutron leakage out of the system is zero.

(Agnew, 1996), to a critical system, would cause the system to become subcritical, meaning that addition of U lessens the probability of an accidental criticality.

As a result of these findings, the tanks in the Hanford TWRS were ranked based on their total Pu fissile nuclide content (Pu-239 plus Pu-241) in grams using data from Agnew (1996). The results of this rank ordering and the total fissile Pu content of each tank are shown in table 4-4a. Table 4-4b lists the same information with the tanks grouped by tank farm. It is noted that tank SY-102 is high on the list presented in table 4-4a but is not the highest ranking tank, implying that the waste in tank SY-102 may not pose the greatest risk of a nuclear criticality of all tank wastes, as assumed in the beginning of this section.

It is noted that the DOE has stated that the margin of subcriticality in the HLW tanks is maintained by two independent criticality parameters; (i) the low plutonium concentration and (ii) the amount of soluble and insoluble neutron absorbers (Braun et al., 1994). The calculated k_{∞} values of representative waste samples is below 0.2, which is highly subcritical (Braun et al., 1994). Based on these statements, it could be concluded that an accidental criticality of in-tank wastes is highly unlikely.

Ex situ vitrification of wastes currently being stored in tank SY-102 may present some hazard for accidental criticalities during the removal, transportation, and solidification of these wastes. Since the exact solidification process that would be used is not known at this time, possible criticality hazards of similar processes that have been or are occurring elsewhere have been reviewed. These reviews are described in the following paragraphs.

Processes with the potential to cause accidental criticalities in the Plasma Hearth Process (PHP) were evaluated (Slate and Santee, 1996). The PHP is a technique currently under development by Science Applications International Corporation (SAIC) for the DOE that destroys the organic component of the waste and vitrifies the inert fraction into glass or slag. Three stages of the process that were susceptible to accidental criticalities were found: (i) the concentration of Pu in the crucible as multiple waste streams are processed, (ii) the pouring of molten slag into a collection drum, and (iii) the arranging of cooled collection drums into matrices that allow for neutron communication between drums during transportation and storage. For example, the maximum safe weight limit of weapons grade Pu was found to be 27 kg for the four-drum (of DOT-17C specifications) arrangement. This was the most reactive arrangement of weapons grade Pu slag drums that was studied. Although the actual numerical limits found by the authors have little meaning to the Hanford TWRS, the three stages of the process that they found susceptible to criticality may represent potential processes that need to be studied in criticality safety analyses at the Hanford vitrification operation.

The potential for accidental criticalities at the defense waste processing facility at the Savannah River Site (SRS) was assessed by Ha et al. (1996). The subprocesses or mechanisms that the authors studied which may lead to criticality during vitrification are: (i) chemical reactions that concentrate U and Pu with respect to iron and manganese neutron absorbers, (ii) fissile material adsorbed onto monosodium titanate, (iii) fissile material entrained in the sludge solids, (iv) Pu solubility in mercury, (v) process cleaning procedures, and (vi) melter accumulation. The authors concluded that in all of the aforementioned subprocesses criticality had a negligible chance of occurring due mainly to the low fissile content of the wastes, the presence of neutron absorbers such as iron and magnesium, and the lack of an identifiable chemical process that can cause the concentration of fissile nuclides relative to the neutron absorbers. Although the authors found that the risk of criticality was insignificant for the SRS, similar subprocesses or mechanisms should be examined for their potential significance for the Hanford TWRS.

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Table 4-4a. A rank ordering of the tanks based on their fissile plutonium content

	Fissile Pu		Fissile Pu	T1-	Fissile Pu
Tank	Content (g)	Tank	Content (g)	Tank	Content (g)
TX-118	7.10E+04	SX-114	4.44E+03	BY-102	1.82E+03
C-102	5.98E+04	U-109	4.33E+03	AP-102	1.74E+03
SY-102	4.54E+04	SX-107	4.22E+03	SX-106	1.74E+03
C-104	3.94E+04	AN-103	4.01E+03	T-110	1.69E+03
AZ-102	3.56E+04	TX-101	3.93E+03	C-109	1.70E+03
S-107	2.89E+04	SX-105	3.88E+03	TX-116	1.64E+03
AY-101	2.82E+04	SX-102	3.78E+03	BY-112	1.64E+03
AZ-101	2.55E+04	C-103	3.78E+03	B-110	1.60E+03
A-106	2.41E+04	SX-112	3.77E+03	C-111	1.57E+03
AW-103	2.37E+04	BY-103	3.75E+03	TX-113	1.55E+03
C-107	2.27E+04	SX-109	3.74E+03	S-108	1.53E+03
C-106	1.86E+04	SX-108	3.74E+03	U-105	1.51E+03
AW-105	1.88E+04	BX-101	3.73E+03	U-103	1.51E+03
AW-104	1.55E+04	BY-106	3.17E+03	B-104	1.42E+03
AW-101	1.42E+04	AN-107	3.16E+03	T-106	1.41E+03
C-105	1.29E+04	SX-110	3.14E+03	TX-112	1.40E+03
B-101	1.33E+04	A-101	3.05E+03	TX-117	1.36E+03
T-101	1.17E+04	A-105	2.99E+03	BY-110	1.37E+03
U-108	1.04E+04	AN-104	2.93E+03	BX-103	1.34E+03
U-107	8.88E+03	SX-103	2.85E+03	TX-105	1.27E+03
AY-102	8.79E+03	T-103	2.82E+03	TX-114	1.27E+03
AX-102	7.38E+03	T-102	2.82E+03	U-102	1.19E+03
C-101	7.01E+03	S-109	2.70E+03	TX-115	1.19E+03
S-101	6.77E+03	AP-105	2.69E+03	U-111	1.16E+03
S-104	6.45E+03	A-102	2.58E+03	BX-111	1.15E+03
AX-101	6.23E+03	BY-111	2.51E+03	S-103	1.12E+03
S-110	5.93E+03	SY-103	2.48E+03	T-104	1.09E+03
SX-101	5.58E+03	AW-106	2.48E+03	AP-106	1.12E+03
SX-111	5.30E+03	B-109	2.43E+03	AX-104	1.10E+03
B-111	5.13E+03	BY-109	2.24E+03	A-103	1.07E+03
AN-105	5.12E+03	T-111	2.01E+03	BY-104	1.05E+03
SX-104	4.97E+03	BY-101	2.04E+03	S-105	1.02E+03
SY-101	4.93E+03	C-112	2.02E+03	BY-107	9.65E+02
AN-102	4.90E+03	BY-105	1.99E+03	TX-110	9.61E+02
S-111	4.71E+03	AW-102	1.98E+03	TX-109	9.42E+02
S-106	4.48E+03	S-112	1.87E+03	U-106	9.62E+02

Table 4-4a. A rank ordering of the tanks based on their fissile plutonium content (cont'd)

Fissile Pu			Fissile Pu
Tank	Content (g)	Tank	Content (g)
S-102	9.13E+02	BX-104	1.84E+02
TX-111	8.76E+02	T-107	1.78E+02
U-112	8.54E+02	TY-102	1.78E+02
TY-101	8.42E+02	U-110	1.73E+02
TX-106	8.45E+02	BX-106	1.76E+02
AN-101	8.37E+02	B-107	1.70E+02
BX-102	8.31E+02	B-103	1.69E+02
BX-107	6.80E+02	TX-103	1.67E+02
BX-110	6.44E+02	B-112	1.64E+02
BX-112	5.57E+02	BX-109	1.45E+02
TY-103	5.27E+02	U-104	1.34E+02
B-105	5.03E+02	T-108	1.10E+02
TX-102	5.01E+02	TX-104	1.10E+02
T-105	4.08E+02	TY-105	6.36E+01
U-202	4.08E+02	SX-113	5.98E+01
U-201	4.07E+02	BX-108	5.87E+01
BY-108	4.01E+02	AN-106	4.71E+01
B-106	3.50E+02	C-108	3.83E+01
T-112	2.99E+02	BX-105	2.33E+01
AP-103	3.09E+02	TX-107	1.36E+01
TY-104	2.79E+02	U-101	4.52E+00
AP-108	2.75E+02	AX-103	1.25E+02
A-104	2.62E+02	B-203	1.37E+00
AP-101	2.50E+02	B-204	1.34E+00
B-102	2.40E+02	T-204	1.04E+00
C-201	2.39E+02	T-203	9.56E-01
C-202	2.39E+02	B-201	7.65E-01
C-203	2.39E+02	T-201	7.65E-01
C-204	2.39E+02	B-202	7.39E-01
T-109	2.22E+02	T-202	5.74E-01
B-108	2.15E+02	TY-106	1.70E-01
SX-115	2.08E+02	AP-104	0.00E+00
U-203	2.06E+02	AP-107	0.00E+00
TX-108	2.06E+02		
U-204	2.04E+02		
C-110	1.95E+02		

Table 4-4b. The plutonium content of the tanks grouped by tank farm

Tank	Fissile Pu Content (g)	Tank	Fissile Pu Content (g)	Tank	Fissile Pu Content (g)
241-A-101	3.05E+03	241-B-102	2.40E+02	241-BY-110	1.37E+03
241-A-102	2.58E+03	241-B-103	241-B-103 1.69E+02 241-		2.51E+03
241-A-103	1.07E+03	241-B-104	1.42E+03	241-BY-112	1.64E+03
241-A-104	2.62E+02	241-B-105	5.03E+02	241-C-101	7.01E+03
241-A-105	2.99E+03	241-B-106	3.50E+02	241-C-102	5.98E+04
241-A-106	2.41E+04	241-B-107	1.70E+02	241-C-103	3.78E+03
241-AN-101	8.37E+02	241-B-108	2.15E+02	241-C-104	3.94E+04
241-AN-102	4.90E+03	241-B-109	2.43E+03	241-C-105	1.29E+04
241-AN-103	4.01E+03	241-B-110	1.60E+03	241-C-106	1.86E+04
241-AN-104	2.93E+03	241-B-111	5.13E+03	241-C-107	2.27E+04
241-AN-105	5.12E+03	241-B-112	1.64E+02	241-C-108	3.83E+01
241-AN-106	4.71E+01	241-B-201	7.65E-01	241-C-109	1.70E+03
241-AN-107	3.16E+03	241-B-202	7.39E-01	241-C-110	1.95E+02
241-AP-101	2.50E+02	241-B-203	1.37E+00	241-C-111	1.57E+03
241-AP-102	1.74E+03	241-B-204	1.34E+00	241-C-112	2.02E+03
241-AP-103	3.09E+02	241-BX-101	3.73E+03	241-C-201	2.39E+02
241-AP-104	0.00E+00	241-BX-102	8.31E+02	241-C-202	2.39E+02
241-AP-105	2.69E+03	241-BX-103	1.34E+03	241-C-203	2.39E+02
241-AP-106	1.12E+03	241-BX-104	1.84E+02	241-C-204	2.39E+02
241-AP-107	0.00E+00	241-BX-105	2.33E+01	241-S-101	6.77E+03
241-AP-108	2.75E+02	241-BX-106	1.76E+02	241-S-102	9.13E+02
241-AW-101	1.42E+04	241-BX-107	6.80E+02	241-S-103	1.12E+03 -
241-AW-102	1.98E+03	241-BX-108	5.87E+01	241-S-104	6.45E+03
241-AW-103	2.37E+04	241-BX-109	1.45E+02	241-S-105	1.02E+03
241-AW-104	1.55E+04	241-BX-110	6.44E+02	241-S-106	4.48E+03
241-AW-105	1.88E+04	241-BX-111	1.15E+03	241-S-107	2.89E+04
241-AW-106	2.48E+03	241-BX-112	5.57E+02	241-S-108	1.53E+03
241-AX-101	6.23E+03	241-BY-101	2.04E+03	241-S-109	2.70E+03
241-AX-102	7.38E+03	241-BY-102	1.82E+03	241-S-110	5.93E+03
241-AX-103	1.25E+02	241-BY-103	3.75E+03	241-S-111	4.71E+03
241-AX-104	1.10E+03	241-BY-104	1.05E+03	241-S-112	1.87E+03
241-AY-101	2.82E+04	241-BY-105	1.99E+03	241-SX-101	5.58E+03
241-AY-102	8.79E+03	241-BY-106	3.17E+03	241-SX-102	3.78E+03
241-AZ-101	2.55E+04	241-BY-107	9.65E+02	241-SX-103	2.85E+03
241-AZ-102	3.56E+04	241-BY-108	4.01E+02	241-SX-104	4.97E+03
241-B-101	1.33E+04	241-BY-109	2.24E+03	241-SX-105	3.88E+03

Table 4-4b. The plutonium content of the tanks grouped by tank farm (cont'd)

Tank	Fissile Pu Content (g)	Tank	Fissile Pu Content (g)
241-SX-106	1.74E+03	241-TX-108	2.06E+02
241-SX-107	4.22E+03	241-TX-109	9.42E+02
241-SX-108	3.74E+03	241-TX-110	9.61E+02
241-SX-109	3.74E+03	241-TX-111	8.76E+02
241-SX-110	3.14E+03	241-TX-112	1.40E+03
241-SX-111	5.30E+03	241-TX-113	1.55E+03
241-SX-112	3.77E+03	241-TX-114	1.27E+03
241-SX-113	5.98E+01	241-TX-115	1.19E+03
241-SX-114	4.44E+03	241-TX-116	1.64E+03
241-SX-115	2.08E+02	241-TX-117	1.36E+03
241-SY-101	4.93E+03	241-TX-118	7.10E+04
241-SY-102	4.54E+04	241-TY-101	8.42E+02
241-SY-103	2.48E+03	241-TY-102	1.78E+02
241-T-101	1.17E+04	241-TY-103	5.27E+02
241-T-102	2.82E+03	241-TY-104	2.79E+02
241-T-103	2.82E+03	241-TY-105	6.36E+01
241-T-104	1.09E+03	241-TY-106	1.70E-01
241-T-105	4.08E+02	241-U-101	4.52E+00
241-T-106	1.41E+03	241-U-102	1.19E+03
241-T-107	1.78E+02	241-U-103	1.51E+03
241-T-108	1.10E+02	241-U-104	1.34E+02
241-T-109	2.22E+02	241-U-105	1.51E+03
241-T-110	1.69E+03	241-U-106	9.62E+02
241-T-111	2.01E+03	241-U-107	8.88E+03
241-T-112	2.99E+02	241-U-108	1.04E+04
241-T-201	7.65E-01	241-U-109	4.33E+03
241-T-202	5.74E-01	241-U-110	1.73E+02
241-T-203	9.56E-01	241-U-111	1.16E+03
241-T-204	1.04E+00	241-U-112	8.54E+02
241-TX-101	3.93E+03	241-U-201	4.07E+02
241-TX-102	5.01E+02	241-U-202	4.08E+02
241-TX-103	1.67E+02	241-U-203	2.06E+02
241-TX-104	1.10E+02	241-U-204	2.04E+02
241-TX-105	1.27E+03		
241-TX-106	8.45E+02		
241-TX-107	1.36E+01		

4.2.6 Lightning Strikes

The Hanford site has an average of 10 days per year during which thunderstorms occur. Based on measurements made worldwide of the relationship between thunderstorm days per year and the number of lightning strikes to the ground, one can expect, on average, about one flash to ground per square kilometer per year at Hanford (Cowley and Stepnewski, 1994). Although actual lightning strikes in the tank farms have not been documented, tank farm operations personnel have indicated that lightning strikes do occur within the tank farms. This observation gives credence to the estimate that strikes occur within the tank farms about once a year. Thus a lightning strike to either an underground storage tank or a piece of support equipment is a credible event.

Studies described in a report by Cowley and Stepnewski (1994) were conducted to evaluate whether or not lightning strikes are a credible accident initiator in Hanford site tank farms and if lightning strikes could result in any unique accidents that are not already addressed in existing safety guidelines for the Hanford site tank farms [e.g., Interim Safety Basis (Leach and Stahl, 1993)]. The study included a survey of equipment that supports the underground storage tanks in order to identify potential consequences of a lightning strike on support equipment and to relate the consequences to existing controls. A walkdown of all of the tank farms was also performed to help identify equipment and structural configurations that could result in the release of radioactive material from lightning strikes. All types of accidents that might be initiated by a lightning strike on support equipment or facilities were analyzed. The studies concluded that lightning strikes on support equipment and facilities do not result in any new accidents, that is, accidents not already addressed in the Interim Safety Basis. Lightning strikes increase the probability of occurrence of some accidents, but the potential damage could be limited by better equipment grounding and bonding techniques and increased use of transient protection on signal and power wiring.

No significant problems resulting from lightning strikes on DSTs were found. The accident with the greatest potential consequence for DSTs, ignition of a flammable gas mixture, was shown to have a calculated probability of occurrence in the 10^{-6} range. The actual probability may be less because the calculated probability does not account for the DST acting as a faraday cage (Cowley and Stepnewski, 1994). Because the DST would act as a faraday cage, not every lightning strike would result in a spark inside the tank. The accident with the greatest potential consequence for SSTs, ignition of a flammable gas mixture, has a calculated probability of occurrence of 1.9×10^{-5} . However, this probability does not give any credit for the effectiveness of grounding and bonding of equipment. If proper bonding and grounding of equipment inserted through risers were ensured, the probability of an external lightning strike causing a spark inside a tank could be reduced below the credible range. Increased ventilation to the tanks would also reduce the probability of an ignition in SSTs because the increased ventilation flow would ensure that gases released to the tank headspace would remain below the LFL.

4.3 POTENTIAL SAFETY CONCERNS ASSOCIATED WITH RETRIEVAL, MIXING, AND TRANSFER OF TANK WASTES

The retrieval and transfer of wastes stored in Hanford waste tanks will require an assessment of waste compatibility. This assessment may be particularly important in connection with future retrieval and transfer of wastes for pretreatment and solidification. The overall problem relates to the potential incompatibility of wastes that are either stored in, or will be received into, the Hanford site DST system, which could result in safety and operations problems. The DOE has formalized the process for assessing waste compatibility for transfers into and within the DST system in its *Tank Farm Waste Transfer*

Compatibility Program (Fowler, 1995a), and data needs for assessing waste transfers are specified in the Data Quality Objectives for Tank Farms Waste Compatibility Program (Fowler, 1995b). The primary goal of these programs is to assure that safety and operations problems such as flammable gas accumulation, tank corrosion, or transfer line plugging do not result due to or during waste transfers in the DST system.

There are two main issues of importance to waste compatibility assessment: (i) safety problems may arise as a result of commingling wastes under interim storage, and (ii) continued operability may be jeopardized during waste transfer and waste concentration/minimization (i.e., plugged transfer or process lines, trapped flammable gas, exothermic reactions, corroded lines or DSTs, or thermally-stressed DSTs). Potential safety problems that need to be considered include:

- Criticality
- Flammable gas generation and accumulation
- Energetics9
- Corrosion and leakage
- Unwanted chemical reactions

Considerations of the above processes help determine whether wastes may be transferred, combined, and stored in DSTs without causing any safety problem.

Evaluation of criticality safety requires information on fissile material concentration (Pu equivalent¹⁰), and in some cases, volume percent of solids. An estimation of solids density may also be needed for comparison of criticality limits given in g/L with measurable quantities such as μ Ci/L or μ g/g. Criticality control in the DSTs is achieved by conducting operations in compliance with criticality prevention specifications (CPS) (Vail, 1994) which limit the Pu equivalent concentration in each DST. The CPS limit the fissile mass available and require a large concentration factor before safety is jeopardized.

The generation of hydrogen or other flammable gas does not by itself pose a safety problem. Safety becomes a concern when flammable gases accumulate to a level above their LFL. Specific gravity is currently used by the DOE as an indicator for potential flammable gas accumulation based on data on the specific gravities for the six DSTs currently on the Flammable Gas Watch-list. Although a direct correlation between specific gravity and gas accumulation has not been established, an evaluation of the method indicated that specific gravity is an appropriate limiting factor for formation of flammable

⁹ Energetics refers to the ability of a waste to sustain a self-propagating exothermic reaction. This is generally measured via thermal analysis (e.g., DSC and TGA) (Fowler, 1995b).

¹⁰ For purposes of criticality control, one gram of Pu is treated as one gram of ²³⁹Pu. For the most part, waste generators need only consider the ^{239,240}Pu concentration when determining Pu equivalent concentration mass. Under certain circumstances, other fissile materials will have to be measured. These materials include ²³³U, ²³⁵U, ²⁴¹Pu, and (if present in sufficient quantities) ²³⁷Np, ²³⁸Pu, and ²⁴¹Am. Treatment of these materials on a Pu equivalent basis is defined in chapter 2 of the Nuclear Criticality Safety Manual, WHC-CM-4-29 (Westinghouse Hanford Company, 1994a).

accumulations (Reynolds, 1994). Other methods of evaluating gas accumulation potential are being investigated (Fowler, 1995b).

The components necessary to oxidize fuel are generally present in tank waste and incoming waste streams. If the temperature increases enough to dry out the waste and initiate a chemical reaction (~ 200 °C), an in-tank reaction could occur. Data needs for evaluating energetics include identification of separable organic material and characterization of waste by DSC and thermo-gravimetric analysis (TGA) augmented, when necessary, by adiabatic calorimetry.

Waste compatibility assessment is also needed to minimize corrosion of steel components of the DSTs, transfer piping, and support facilities and reduce the possibility of leakage to the environment. DOE operating specification documents establish waste composition limits to keep corrosion rates below 1 mil per yr and to inhibit stress corrosion cracking (Fowler, 1995b). These limits are specified in Westinghouse Hanford Company (1994b) and Westinghouse Hanford Company (1994c). Data needs for determination of the possible occurrence of corrosion and leakage include pH, temperature, and concentrations of hydroxide, chloride, nitrate, and nitrite. These issues are briefly discussed in section 2.6.

For determining the potential of unwanted chemical reactions, DOE procedures call for determining chemical compatibility based on the reactivity group number of the source waste. This information is to be provided by the waste generator on a waste profile sheet in accordance with the Waste Analysis Plan (Mulkey and Jones, 1994). Source wastes will be categorized according to EPA compatibility matrix (U.S. Environmental Protection Agency, 1994a,b) and potential chemical compatibility hazards will be identified prior to acceptance into a DST (Fowler, 1995b).

In addition to safety concerns, waste compatibility assessment is also needed to address operational concerns including

- Heat load limits on receiving tank (tank farm ventilation capacity issue)
- Plugged pipeline and equipment (unanticipated precipitation)

Consideration of the above processes will help determine whether wastes may be transferred/combined without exceeding the physical constraints of the transfer piping and tanks in the DST system and will be instrumental in identifying safety issues and regulatory controls that are needed to assure safe operations.

DOE procedures place limits on the heat generation rates of the wastes to prevent localized boiling. This is necessary because the ventilation systems in tank farms AN, AP, AW, and SY were not designed to handle boiling, and internal boiling arising from excessive heat generation rates could lead to release of radionuclides. Although the other two tank farms, AY and AZ, have ventilation systems designed to handle boiling, waste heat generation rates in these tanks need to be kept below the vent system design limit $(10\times10^6 \text{ Btu/h per tank})$ (Fowler, 1995b). The heat generation rate is usually estimated based on the mean Sr-90 and Cs-137 concentrations. These are generally measured using beta counting and gamma energy analysis.

Waste compatibility analysis is needed to ensure pumpability of the source waste to the receiving tank and that no reactions occur that could lead to plugging of process lines and equipment. Plugging of process lines and equipment may be considered to be a safety issue because (i) rupturing of pipes may

occur due to sudden overpressurization; not all transfer lines are equipped with appropriate rupture disks and leak catchment systems; and (ii) the measures taken to remove plugging and restart the system may need safety analyses. Pumpability of the source waste is estimated by determining the Reynolds number for the transfer system. Data needs for calculating the Reynolds number are density of the waste, viscosity of the waste, pipe diameter, and pump velocity (flow rate). Volume percent solids (measured and/or estimated) and the cooling curve verification of precipitating solids as a function of temperature may also be needed to aid in the determination of waste pumpability.

The DOE approach to evaluating waste compatibility for the DST system has been developed based on engineering process knowledge and observations of operational problems. Basic information needs and decision criteria established by DOE for compatibility assessments are discussed in Fowler (1995b). The chemical and physical data needed for these assessments are listed in table 4-5.

Table 4-5. Analytical data needs for compatibility assessment (taken from Fowler, 1995b)

	Safety Concerns				
Parameter	Criticality	Flammable Gas	Energetics	Corrosion	Operations Concerns
Aluminum					Х
Americium-241					X
Carbonate					X
Cesium-137					X
Chloride				X	
Cooling Curve					X
Exotherm/Endotherm Ratio			X		
Fluoride				Xª	X
Hydroxide			-	X	X
Nitrate				X	X
Nitrite				X	X
Organic Carbon					X
Organic, Separable			X		
pН				X	
Phosphate					X
Plutonium-239/240*	X				
Solids, Vol. %	X				X
Specific Gravity		X			X
Strontium-90					Х
Sulfate				X ^a	X
Uranium	X				
Viscosity					X
Water, wt. %					X

a Although not included by Fowler (1995b), concentrations of fluoride and sulfate should be considered in the case of corrosion as potential aggressive anions.

^{*} Total alpha may be used for this determination. Other fissile elements may be needed as noted in footnote 12 in section 4.3.1.

5 SUMMARY AND ADDITIONAL INFORMATION REQUIREMENTS

5.1 SUMMARY

The large amount of information pertaining to Hanford site, history of processes, the TWRS, and tank waste contents was reviewed in the previous chapters. This review is neither exhaustive nor critical. The privatization contractor activities related to waste retrieval and solidification are part of a much larger TWRS program. However, the objective of the report is to provide the reader sufficient background information to assess the safety aspects of TWRS privatization contractor activities and the performance of onsite waste disposal systems.

Chapter 2 describes the Hanford site geographical and geological features, natural hazards, site contamination, the tank farms, reactors, and associated facilities, and ongoing activities related to TWRS. The geological and geographical features were provided in the form of a map generated by ArcView. This technique of generating the site map, in addition to its enhanced accuracy over image reproduction from previous reports, provides a particular advantage because as further information on the site characteristics and coordinates of various facilities is acquired, this can be overlaid on the existing maps electronically. The current state of knowledge of the groundwater, soil, and surface water contamination is also reviewed in chapter 2. A review of available information of site contamination suggested that, in general, concentrations of radionuclides in effluents has not changed significantly over the last few years since decommissioning of most production facilities. Onsite surface and near-surface soils had concentrations above applicable regulatory limits of Co-60, Sr-90, Cs-137, Pu-239, and Pu-240, with the highest levels near waste disposal sites. Borehole data show that below the 200-West Area, Cs was recorded to a depth of at least 38 m, which is the top of a low-permeability confining bed. Surface water analyses in the 200 Areas indicate that the radionuclide and the hazardous chemical contaminant concentrations are below regulatory limits. Analyses of surface water near the 100 Area reactors indicate elevated levels of Sr-90 and H-3. Only in the 100-K Area do the concentrations of nonradiological chemicals exceed the regulatory limits. Mapping of the groundwater contaminant plumes suggests that these are clustered around the various processing plants. Near the proposed privatization facilities, elevated levels of H-3 have been found. The only nonradiological components being discharged at elevated levels offsite to the Columbia River are chromium and nitrate.

The engineering systems of importance to the TWRS include the waste tanks, transfer systems (including valve pits, jumpers, and transfer lines), evaporator/crystallizer, and the solidification facilities. The solidification systems have not been constructed and will be reviewed as part of subtask 1.2. The new waste transfer line under construction between the 200-West and -East areas (known as the cross transfer line) was reviewed. Pitting, erosion, and malfunctioning of the cathodic protection system are considered to be the three most important performance limiting failure processes for the cross transfer piping system. Plugging of the lines due to chemical reactions or hydrodynamic changes is also an important consideration for performance. The same performance issues may be relevant to the new pipe transfer systems constructed to transfer waste from Tanks AP-106 and AP-108 to the privatization facilities, as well as the piping systems within the privatization facilities. The evaporator/crystallizer is essential in reducing the waste volumes prior to solidification. At present, the expected life of the evaporator/crystallizer is 8 to 10 yr before replacement of the construction materials is needed.

The privatization facilities will be constructed in the area adjacent to the AP tank farm east of 200 East. At present, the site for disposal of the LAW products is not known. The ongoing programs pertaining to TWRS include vadose zone characterization, tank waste characterization, evaluation of watch-list tanks, resolution of unreviewed safety questions, continued operation of tank farms, construction and operation of cross-transfer piping, upgrades to the tank farms and transfer lines, development of the initial tank retrieval system, determining the disposition of Cs and Sr capsules, and initiating resolution of other issues pertaining to tank closure, retrieval of tank waste residuals, and contamination around tanks. Chapter 2 also surveyed the information available on other facilities that are under construction or will be constructed by DOE. These facilities are outside the scope of the contracts associated with the two privatization contractors, but are part of the DOE TWRS program. The CSB that is being constructed at the site of the former vitrification facility is an important component of the disposition of spent fuel from the K-basins. In the phased alternative of the TWRS, the CSB may also be used to store some of the high-level waste glass logs. Unfortunately, at the time of writing of this report, detailed design information regarding the CSB was not available.

Identification and quantification of Hanford tank waste contents are subjects of extensive study. Chapter 3 of this report includes a general description of DST and SST waste characteristics, and a discussion of tank inventories of chemicals and radionuclides. The wastes have been produced over a long period of time by a variety of processes; characterizing tank contents chemically and radiologically is therefore a challenging task. Two approaches to this question are being employed by Hanford, each complementing the other: direct sample assay and estimation based on facility records. The former is limited by the extreme physical and chemical heterogeneity of the tank contents, while the latter may be unreliable due to incomplete or inaccurate documentation of process and waste transfer transactions. The Hanford effort is centered on determination of a "best-basis" value for each constituent in each tank, based on a combination of the assay and historical data. Until that evaluation process is completed, the historically based HDW model being developed at LANL is the most complete and thorough data set of tank inventory estimates. The CNWRA has prepared a database (based on inventories from the HDW model) allowing access of tank information utilizing ARC/INFO geographical information system software. The available tank inventory estimates show that sodium is by far the most abundant metal, comprising 80 percent by weight of the metal cation population. Important inorganic anions are nitrate, hydroxide, nitrite, and carbonate, while the most abundant organic complexants are glycolate and HEDTA. The radionuclide inventory is dominated by Sr-90 (75 percent of the SST radioactivity and 27 percent of the DST radioactivity) and Cs-137 (24 percent of the SST and 72 percent of the DST).

Chapter 4 reviewed the various hazards posed by tank wastes and associated with the retrieval and mixing of wastes prior to solidification. The safety issues associated with solidification will be discussed in another report (Jain, 1997). A partial list of the hazards posed by tank wastes and the TWRS activities includes:

- (i) Flammable gas: flammable gases such as hydrogen are generated due to radiolytic as well as organic reactions. Recently all 177 tanks were placed on the flammable gas watch-list. Final resolution of how many tanks present risk due to flammable gas has not occurred.
- (ii) Organics: the potential for exothermic and explosive reaction between organics and oxidants such as nitrates is a concern. Of the original 36 tanks placed under this list, only 20 tanks are still on the watch-list.

- (iii) Ferrocyanide: the potential for explosion when ferrocyanide left over from Cs and Sr extraction combines with oxidants is a concern. At present all tanks have been removed from the ferrocyanide watch-list.
- (iv) High-heat: radioactive decay may generate high heat that may in turn result in structural damage of the concrete walls. Only tank C-106 was placed on this watch-list and long-term resolution is being pursued.
- (v) Crust burn associated with secondary ignition of organic-nitrate/nitrite mixtures in the crust layer
- (vi) HEPA filter blowout associated with flammable gas
- (vii) Organic solvent leading to possible flammability and environmental contamination
- (viii) Known and unknown leaking tanks
- (ix) Criticality
- (x) Lightning strikes

The report also evaluated potential safety concerns associated with retrieval, mixing, and transfer of tank wastes. Two important concerns are (a) safety problems arising from commingling wastes under interim storage and (b) operability of waste transfer systems that may be impeded by plugging, trapped flammable gas, exothermic reactions, and corroded lines. The physical and chemical data needs for compatibility assessments are provided in chapter 4.

5.2 ADDITIONAL INFORMATION REQUIREMENTS

Information requirements for hazard analyses will evolve as further knowledge is gained regarding the processes for tank waste retrieval, separations, feed preparation, solidification, and disposal. The additional information requirements are categorized below in terms of various components of the TWRS. Topics related to solidification processes and chemical reactions are not listed, since they are discussed in other reports (Jain, 1997; Pabalan et al., 1997).

5.2.1 Site Contamination

At the time of this document's production, limited data were available regarding the extent and magnitude of radionuclide and chemical contamination of surface soils and subsurface soils of the vadose zone at the Hanford site. As planned site characterization activities proceed, specifically, the work to be accomplished by US Ecology near the 200 Areas, study of contaminant migration in the 200-West area under the SX tank farm, and study of the 200-East area (under one of the potential LAW disposal sites), new data made available from these investigations need to be evaluated, summarized, and incorporated in this document.

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5.2.2 Waste Characteristics

Important information needs pertain to the chemical and physical characteristics of the wastes in each of the Hanford tanks. Data on tank chemical composition is needed for safety analysis and assessment of waste compatibility. Information on radionuclide composition of individual tanks is also needed for safety and waste compatibility analyses, as well as for determining shielding requirements for TWRS equipment. Waste rheology and shear strength need to be known to determine dilution requirements, equipment needs, and efficiency of retrieval of tank wastes. However, as pointed out in chapter 3, characterization of the chemical and radionuclide inventories of the tank wastes is not a straightforward task. Estimation of inventories based on facility records may be unreliable due to incomplete or inaccurate documentation of tank additions and waste transfers, whereas direct sample measurement is limited by the extreme physical and chemical heterogeneity of the tank contents. Inventories also change due to waste degradation, radioactive decay, and waste transfers. Although a relatively sophisticated and systematic estimation of individual tank contents is being conducted (e.g., Agnew, 1996), future NRC analyses of Hanford TWRS operations should cross-check and verify these estimates with actual data resulting from DOE waste characterization efforts.

5.2.3 Transfer Lines

As described in chapter 2, the transfer lines within the 200-East and -West Areas have a variety of designs. These designs include (i) a pipe-in-pipe system which is the design for the new cross-transfer piping between the 200-East and 200-West areas; (ii) a pipe-in-concrete system which was the design for the original cross-transfer piping and many of the other piping systems within the 200-East and 200-West areas; and (iii) direct buried pipes, which were used in some of the transfer lines within the 200-East and 200-West areas. Some of these transfer lines are considered to be arterial lines, in that a leak or blockage may have a significant impact on the continued operation of the tank farms and the TWRS. In addition to the cross-transfer lines, selected transfer lines within the 200 Areas are planned to be upgraded to double-walled piping, and equipped with leak detection and corrosion protection systems. The transfer lines selected for replacement are (i) the line connecting the T plant to the SY farm in the 200-West Area, (ii) the line connecting the PFP to the SY tank farm, (iii) other piping in the SY tank farm, (iv) the distribution piping in the A-tank farm, and (v) the piping in the AY and AZ tank farms. Two areas of additional information needs are (i) the analyses of causes of old piping failure including blockage and corrosion, and (ii) the location of all existing piping systems, especially in the 200-East Area. The former is important to understanding the changes in the design of the piping systems and to determining if blockage would occur upon retrieval of wastes mixed from different tanks. The latter is important to understanding which transfer lines are most important to the safe functioning of the TWRS.

5.2.4 Tank Waste Contents

The information on tank waste contents described in chapter 3 has focused on the SSTs and DSTs. Information on the appryoximately 20 MUSTS and 40 Inactive Miscellaneous Underground Storage Tanks (IMUSTs) is sparse. While these are not of concern for Phase I of the TWRS, the wastes in the MUSTs and IMUSTs are planned to be retrieved and processed in Phase II (US Department of Energy, 1996b). The volume of wastes in these tanks is less than 1 percent of the total waste inventory, and the chemistry is expected to be similar to that of the SSTs. Nevertheless, the details of tank contents are not available at present.

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APPENDIX B

GEOGRAPHIC INFORMATION SYSTEM DATABASE OF TANKS AND TANK WASTES

GEOGRAPHIC INFORMATION SYSTEM DATABASE OF TANKS AND TANK WASTES

A computerized database for the Hanford tanks and tank wastes project was constructed using the ARC/INFO Geographic Information System (GIS) developed by Environmental Systems Research Institute, Inc. (ESRI). The purpose of the GIS is to provide a computer-based information source on data related to underground storage tanks and tank wastes located within the 200 Areas at the Hanford site. The GIS currently contains information on waste tank status, characteristics, and waste chemistry. The information within the database was collected from reports prepared for the DOE by contractors and laboratories. The GIS provides an avenue for rapid retrieval and evaluation of this data. Display, examination, and analysis of the data contained in the GIS are best achieved using the ArcView desktop mapping software package also developed by ESRI. Following are brief descriptions of the ARC/INFO GIS system, the data collected and entered into the GIS database to date, the ARC/INFO coverages specifically constructed for the project, and the ArcView desktop mapping software.

The ARC/INFO GIS system is designed to link quantitative and qualitative data contained within a database to a common spatial reference or geographic location. In the ARC/INFO environment, discrete data types intrinsically tied to geographic locations are separated into layers or coverages. Coverages usually consist of a single data format: points, lines, or areas. Points, lines, or areas can be digitized or edited into coverages. When a coverage is created, ARC/INFO builds an associated attribute table which is linked to the coverage. Spatial information about each point, line, or area added to the coverage is automatically entered into the attribute table by ARC/INFO. Data items and associated data are then added to the attribute table to complete the coverage. In short, an ARC/INFO coverage is linked to a table which contains spatial data and associated attribute data for each point, line, or area in the coverage.

The GIS for the Hanford tanks and tank wastes project contains information on the configuration, status, surveillance, liquid and solid contents, and chemical and radionuclide composition of each of the existing 177 large underground storage tanks in the 200 Areas at the Hanford site. Coverages for the GIS are composed of line and area data types which represent streets, buildings, tank farms, and underground storage tanks within the 200 Areas at the Hanford site. The geographic locations of streets, buildings, tank farms, and storage tanks at the site were digitized directly from an existing map of the 200 Areas (figure 3.2.3; U.S. Department of Energy, 1996b). Data on tank status, characteristics, and contents (e.g., shell type, total capacity, liquid and solid waste volumes) were manually entered into the GIS from charts and tables in a waste tank summary report prepared for the DOE by WHC (Hanlon, 1996). An electronic version of the HDW report (Agnew, 1996) was obtained and used to input the estimated chemical and radionuclide inventories of tanks into the GIS. The chemical and radiological constituents contained in the GIS are listed in appendix A. Development of the database accessed by the GIS [e.g., incorporation of data on radionuclides not addressed by the Agnew model (see above)] will continue.

The GIS is currently composed of six coverages (coverage name is in parentheses): streets (HANSTREETS), buildings (HANBLDGS), tank farms (HANFARMS), underground storage tanks (HANTANKS), a summary of storage tank status and characteristics (TANKSUMM), and a chemical and radionuclide inventory (TANKCHEM). A schematic diagram of the GIS showing the coverages and data items contained within each coverage is shown in figure B-1. Notice that information on tank status, characteristics, and chemistry are contained in the TANKSUMM and TANKCHEM coverages; the other coverages provide data about the site infrastructure (e.g., street and building names). Data can be added to these existing coverages as additional information concerning the site geography or tanks if needed or

requested. As waste is retrieved from the tanks for processing, information on the status, content, and chemistry of the tanks can be modified. New coverages can also be constructed and added to the GIS. For example, a coverage is planned which will show the location and describe the characteristics of cross-transfer piping for the retrieval and transport of tank wastes to staging, sampling, and treatment facilities.

Although coverages can be displayed and examined using the ARC/INFO system, its data management and analysis features are limited. ArcView is a sophisticated desktop mapping software package which has extremely flexible data management, analysis, and reporting features. ArcView can read ARC/INFO coverages without translation or recompilation. A major advantage of ArcView is that it permits access to spatial data in ARC/INFO format to users not directly associated with an ARC/INFO site. In addition, unlike the UNIX-based ARC/INFO, ArcView can be installed and executed on either Windows or UNIX platforms.

The ArcView graphical user interface allows users to quickly display coverages, dynamically examine data, perform spatial and logical data queries, and create maps. A map of the site constructed with ArcView using the Hanford tanks coverages is shown in figure B-2. ArcView is an excellent tool for performing spatial analyses and presenting information graphically (in charts, tables, and maps). For the Hanford tanks and tank wastes GIS, waste tanks with certain characteristics or waste contents and tank wastes with certain chemistries can be identified using query operations. To demonstrate, a simple example is presented. A logical query was performed to identify waste tanks in the 200-East Area with a Pu-239 concentration greater than $1.0~\mu\text{Ci/g}$. The information resulting from this query operation is presented graphically in figure B-3. In this figure, the tanks with Pu-239 concentration greater than $1.0~\mu\text{Ci/g}$ are highlighted on a map of the 200-East Area. The figure also includes a table which lists the tank numbers and estimated Pu-239 concentrations of the highlighted tanks.

Further work in this area was halted in January 1997, due to resource constraints.

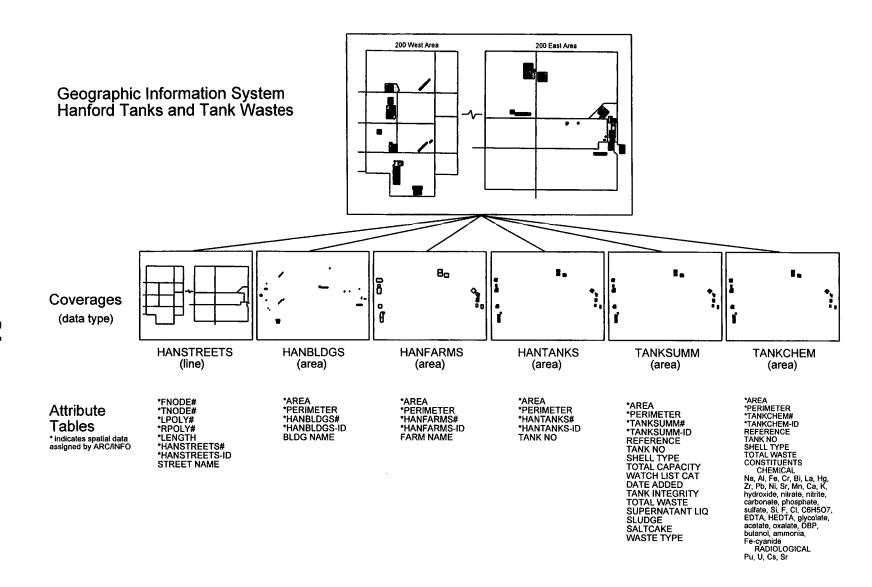


Figure B-1. Schematic diagram of the Hanford tanks and tank waste Geographic Information System showing the coverages, data types, and data items contained within each coverage

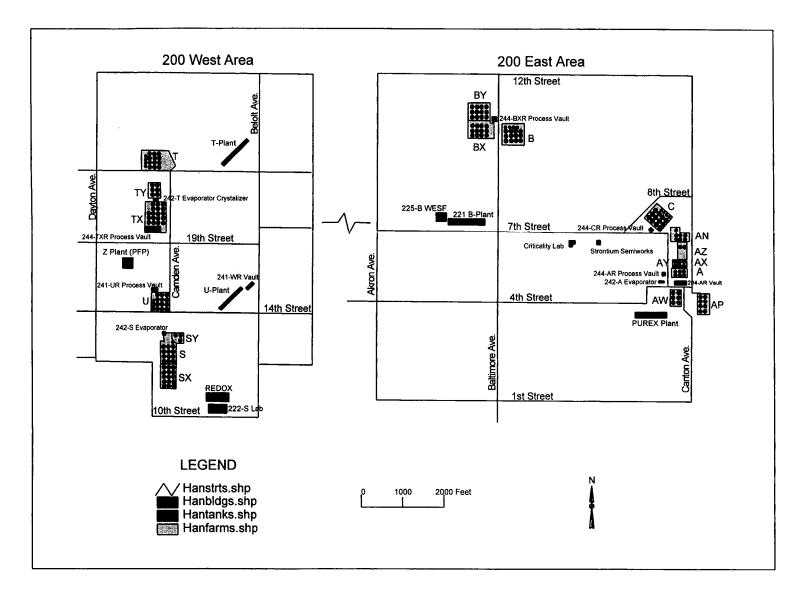


Figure B-2. Map of the 200 Areas at the Hanford site constructed with ArcView using the Hanford tanks ARC/INFO coverages

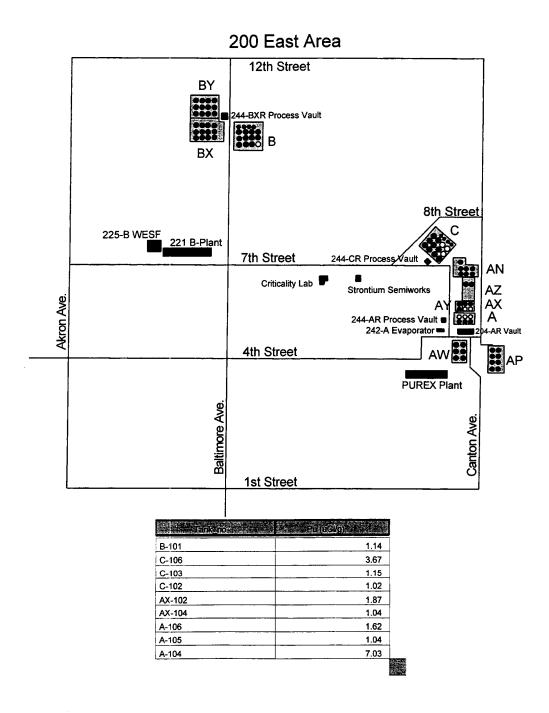


Figure B-3. Map of the 200-East Area at the Hanford site highlighting tanks (open circles) with a Pu-239 concentration greater than 1.0 μ Ci/g. The tank numbers and estimated Pu-239 concentrations of the highlighted tanks are listed in the table at the bottom.