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Origin of Wastes in C-200 Series Single-Shell Tanks

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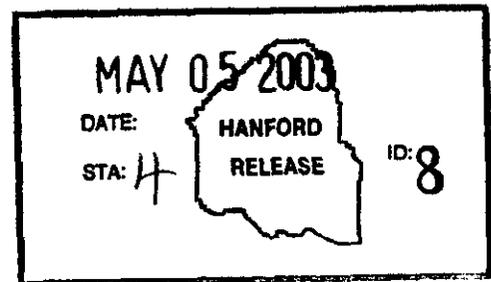
Abstract: A review of historical documents was conducted to determine the origin of wastes transferred into Hanford site tanks 241-C-201 through 241-C-204. Metal waste from the B-Plant was originally transferred into these tanks from 11/1947 to 01/1948 and then subsequently removed from 03/1953 to 01/1955. Waste from pilot-scale testing of the PUREX process at the Hot Semi-Works was transferred into these tanks from 05/1955 to 11/1956 and subsequently removed in 1970, leaving a sludge heel.

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ORIGIN OF WASTES IN C-200 SERIES SINGLE-SHELL TANKS

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EXECUTIVE SUMMARY

A review of historical documents was conducted to determine the waste transfers into and waste removal from single-shell tanks 241-C-201 through 241-C-204. These tanks were originally constructed as part of the Manhattan Project from 1944 to 1945. Single-shell tanks 241-C-201 through 241-C-204 sat unused until November 1947 when they were activated to store metal waste (high-level waste) from operation of the bismuth phosphate process in the 221-B Separations building. These tanks were filled with metal waste by January 1948.

Metal waste was hydraulically mined from these tanks from March 1953 through January 1955. The metal waste sludge and supernatant was dissolved in acid in the 244-BXR vault and then transferred to the 221-U building where uranium was recovered from these wastes using a tributyl phosphate-based solvent extraction process. After completing the removal of metal waste, each tank was visually inspected and determined to be empty. However, given the inspection method, periscope optics, residual metal waste could have been left in each tank.

Tanks 241-C-203 and 241-C-204 received cold uranium (i.e., uranium that had not been irradiated in a reactor) waste from plutonium-uranium extraction (PUREX) startup testing in November 1955. The cold uranium waste was removed from tanks 241-C-203 and 241-C-204 in December 1955. Tanks 241-C-201 through 241-C-204 were then used from May 1955 through October 1956 to receive and store waste originating from research and development activities conducted at the 201-C Hot Semiworks facility in the 200 East Area of the Hanford Site. The cold uranium waste was removed from tanks 241-C-203 and 241-C-204 before transfers of Hot Semiworks waste into these tanks was conducted.

Tanks 241-C-201 through 241-C-204 were not used to receive waste after being filled with waste from the Hot Semiworks. The liquid in tanks 241-C-201, 241-C-202, and 241-C-204 was transferred to single-shell tank 241-C-104 in 1970. The liquid in tank 241-C-204 was transferred to single-shell tanks 241-C-104 and 241-C-109 in 1970. Residual liquids were subsequently transferred from these tanks into single-shell tank 241-C-106 in 1980.

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LIST OF TERMS

1C	first cycle decontamination waste
2C	second decontamination cycle wastes
CW	coating waste
DST	double-shell tank
g/L	grams per liter
HDW	Hanford Defined Waste
kg	kilograms
lbs	pounds
MW	metal waste
NCRW	neutralized PUREX cladding waste
PUREX	plutonium-uranium extraction
REDOX	reduction-oxidation
SST	single-shell tank
TBP	tributyl phosphate
$\mu\text{Ci/cc}$	microcurie per cubic centimeter
$\mu\text{g/cc}$	microgram per cubic centimeter
$\mu\text{g/L}$	microgram per liter

1.0 INTRODUCTION

This document discusses the waste transfers into and waste removal from single-shell tanks 241-C-201 through 241-C-204. Section 2.0 provides a description of the different types of wastes that were generated at the Hanford Site chemical processing plants and transferred to the C-200 series single-shell tanks (SST). A basic understanding of the different types of wastes that were generated at the Hanford Site is provided for the reader to comprehend the waste types transferred into and waste removed from tanks 241-C-201 through 241-C-204, as discussed in Section 3.0.

2.0 TYPES OF TANK WASTE GENERATED AT THE HANFORD SITE CHEMICAL PROCESSING PLANTS

There are 149 single-shell and 28 double-shell underground storage tanks located at the Hanford Site. These tanks received supernatants and precipitated sludges originating from the reprocessing of spent nuclear fuels, research and development, and waste management activities.

There were numerous spent nuclear fuel reprocessing, research and development, and waste management activities conducted at the Hanford Site starting in 1944. 221-T Plant, first used for reprocessing of spent nuclear fuel in December 1944, operated until March 1956 using the bismuth phosphate process. T Plant was reactivated in July 1960 (HW-66271-DEL, page C-2) and continues to be used to date as a decontamination facility. 221-B Plant reprocessed spent nuclear fuel from April 1945 to June 1952 using the bismuth phosphate process. The bismuth phosphate process was based on carrier precipitation batch chemistry. B-Plant was later renovated and used from 1963 through 1986 to recover the fission products cesium and strontium from the wastes stored in SSTs.

Later, reduction-oxidation (REDOX) (January 1952 through November 1966) and plutonium-uranium extraction (PUREX) (1956 through 1988) plants replaced B- and T-Plants using continuous solvent extraction processes for separating uranium and plutonium from dissolved, spent nuclear fuels. Uranium was recovered from the wastes stored in the SSTs from operation of the bismuth phosphate plants using a tributyl phosphate solvent extraction process in the Tributyl Phosphate (TBP) Plant (221-U building). The Hot Semiworks, building 201-C, was operated from 1949 through 1967 as a research and development facility for many of the Hanford Site chemical processes (e.g., REDOX, TBP, B-Plant strontium separations, PUREX process tests). All of these facilities generated numerous sources of radioactive mixed wastes that are stored in the SSTs and double-shell tanks (DST).

In addition to the operations conducted in the processing plants, there were numerous activities conducted within the underground storage tanks, including evaporation, cesium precipitation using ferrocyanide, and discharge of supernatants to specific activity retention cribs and trenches. These numerous spent nuclear fuel reprocessing, research and development, and

waste management activities resulted in the mixing and alteration of the different waste types within several (but not all) of the 149 SSTs and 28 DSTs.

These spent nuclear fuel reprocessing, research and development, and waste management activities conducted in the processing plants are discussed further in the following sections. Refer to DOE/RL-97-02, *National Register of Historic Places Multiple Property Document Form - Historic, Archaeological and Traditional Cultural Properties of the Hanford Site, Washington February 1997*, for additional details on these processes.

2.1 BISMUTH PHOSPHATE PROCESS - B AND T PLANTS

B- and T-Plants were constructed in 1944 through 1945 to separate plutonium from spent nuclear fuel using the bismuth phosphate process. Figure 2-1 shows a summary of the 221-B/T Plant bismuth phosphate process, which is referred to throughout this discussion.

In the bismuth phosphate process, the aluminum cladding of spent nuclear fuel elements was dissolved in boiling sodium nitrate solution, to which sodium hydroxide was slowly added (HW-10475-C, page 403). The cladding removal waste, sometimes referred to as coating waste (CW), was transferred to single-shell underground storage tanks. (See item 1 in Figure 2-1.)

The fuel element uranium cores (see item 2 in Figure 2-1) were then dissolved in nitric acid (HW-10475-C, chapter IV, page 405). Water and sulfuric acid were added to the dissolved uranium metal solution, and the mixture was then transferred to the plutonium extraction section. The sulfuric acid formed a uranyl sulfate complex that prevented its precipitation as a phosphate in the subsequent plutonium extraction step (HW-10475-C, page 418).

Plutonium was extracted from the acid solution by addition of bismuth nitrate and phosphoric acid to form a bismuth phosphate carrier precipitate (HW-10475-C, page 503). The plutonium and bismuth phosphate carrier precipitate was centrifuged and washed with water to separate the acidic supernatant from the precipitate, (see item 3 in Figure 2-1). The acidic solution remaining after the plutonium precipitation contained about 99 percent of the uranium, about 90 percent of the fission products. This separation process also removed and reduced the gamma radiation activity level in the plutonium precipitate by a factor of 10. However, zirconium phosphate is insoluble and zirconium-95 (10 percent of the activity) stayed with the plutonium product. The acidic uranium solution was then neutralized and transferred to the underground SSTs as metal waste (MW). Recent laboratory testing of the bismuth phosphate flowsheet confirms this partitioning of radionuclides (internal letter 7G300-02-NWK-024, "Bismuth Phosphate Process Radionuclide Partition Factors for the Hanford Defined Waste Model"). The laboratory tests indicate the percentage of cesium-137 and strontium-90 partitioned to the metal waste may have been as high as 100 percent and 89 percent, respectively.

The plutonium-bearing cake was then dissolved in nitric acid and further decontamination of the plutonium to separate fission products was conducted (HW-10475-C, chapter VI). Sodium bismuthate, sodium dichromate, or potassium permanganate was added to oxidize the plutonium to the +6 valence-state. This step caused the bismuth phosphate to precipitate phosphate

insoluble fission products ("by-product precipitation"), leaving the plutonium in solution. The precipitate was separated from the plutonium-bearing solution using centrifuges and washed to remove soluble plutonium. The plutonium was reduced to the +4 valence state to form a precipitate that could be separated from the remaining soluble fission products by centrifugation. The fission products separated from the plutonium product during this first cycle of the decontamination process (designated as 1C) were combined with the coating removal waste and transferred to SSTs. The 1C waste (see item 4 in Figure 2-1), contained approximately 10 percent of all fission products and approximately 1.4 percent of the plutonium present in the original fuel charged to the plant (HW-23043, pages 20 and 22). After 1951, the Bismuth Phosphate process flowsheet was modified to include cerium and zirconium scavenger precipitation in the 1C by-product step to remove lanthanide and zirconium radionuclides from the plutonium product (HW-23043, page 16).

The plutonium solids were again dissolved in nitric acid. A second decontamination cycle (see item 5 in Figure 2-1) was conducted to reduce the gamma activity level by a factor of 10,000 from that in the previous dissolved metal solution, giving an overall process decontamination factor of 100,000 below that of the original solution (HW-10475-C, page 627). The second decontamination step essentially repeated the steps previously described for the first cycle decontamination. The second decontamination cycle wastes (designated as 2C) were also transferred to the SSTs. The 2C waste contained less than 0.1 percent of the uranium and fission products and about 0.4 percent of the plutonium present in the original fuel charged to the plant (HW-23043, pages 26 and 28).

Table 2-1 provides the estimated compositions of the neutralized CW, MW, 1C, and 2C waste solutions generated from the bismuth phosphate plants based on the October 1, 1951 flowsheet (HW-23043). Additional analyses of the supernatant fraction of MW, 1C, and 2C that was stored in SSTs are provided in Tables 2-2 and 2-3. These sample analyses support previous statements regarding the partitioning of fission products to the various Bismuth Phosphate Plant Waste Streams. Specifically, 90 percent of the fission products were partitioned to the metal waste as evident by the cesium-137 concentration provided in Table 2-3. About 10 percent of the fission products partitioned to the 1C waste, as demonstrated by the gross beta and gross gamma radionuclides analyses provided in Table 2-2 and the cesium-137 analyses provided in Table 2-3. Note that the coating waste batch size shown in Table 2-1 is based on 6,600-lbs uranium, but that the metal waste dissolution batch size is based on 2,200-lbs uranium. These sample analyses support that the 2C waste contained less than 0.1 percent of the fission products.

2.1.1 224-B and 224-T Concentration Buildings

The plutonium product from the bismuth phosphate process was subsequently concentrated in the 224-T and 224-B buildings using a lanthanum fluoride precipitation process. The process steps executed in the 224 buildings were as follows (HW-10475-C, chapter VII and HW-23043, pages 34 to 55):

- The starting batch size received from the 221 buildings was 330 gallons.
- Plutonium solution from the 221 buildings was oxidized with sodium bismuthate.

- Phosphoric acid was added to produce a by-product precipitation (with the plutonium still in solution).
- The solution and precipitate were separated by centrifugation.
- Nitric acid was added to dissolve the by-product cake, and this solution was removed as waste.
- The plutonium was oxidized with potassium permanganate.
- Hydrogen fluoride and lanthanum salts were added to the plutonium solution (the "crossover" step), producing plutonium lanthanum fluoride. Lanthanum was such a good carrier solution that plutonium could be carried with very little bulk or volume of carrier.
- Impurities were precipitated in a by-product cake (as the plutonium was oxidized at this point). Fission products were carried with the lanthanum. This by-product cake contained all the lanthanides (cerium, lanthanum, etc.) and residual ruthenium, samarium, europium, americium, and curium that the bismuth phosphate could not carry out of the stream.
- The cake was dissolved in nitric acid, neutralized with sodium hydroxide, and sent to tanks within the 224 buildings for settling.
- Plutonium was reduced to the +4 valence-state by adding oxalic acid.
- Potassium hydroxide was added to convert the plutonium lanthanum fluoride to solid plutonium lanthanum oxide.

The liquid was removed by centrifugation. The solid plutonium lanthanum oxide was then dissolved in nitric acid, making plutonium nitrate. By this time, each original 330-gallon batch of plutonium-bearing solution that had entered the 224 buildings was concentrated down to eight gallons. The liquid waste (designated as "224") from the lanthanum fluoride and barium sulfate precipitation process was neutralized and transferred to the SSTs. Table 2-1 provides the estimated compositions of the neutralized 224 waste solutions based on the October 1, 1951 flowsheet (HW-23043). The resulting purified plutonium material was transferred to the 231-Z building and subsequently to the 234-5 building (Z-Plant) beginning in 1949 for further processing.

Figure 2-1. Fission Product Distribution in Bismuth Phosphate Process.

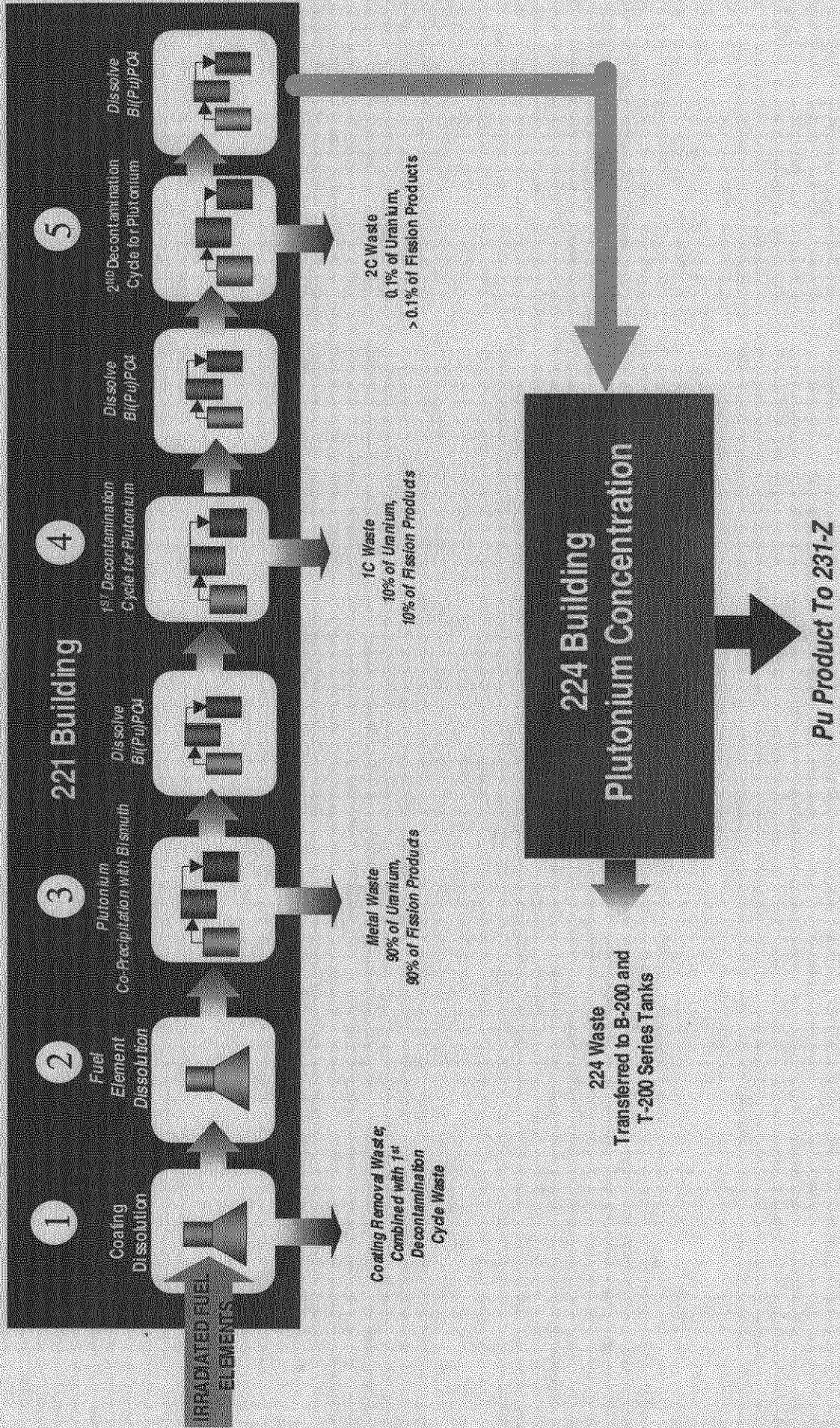


Table 2-1. Estimated Composition of Bismuth Phosphate Plant Wastes
From October 1, 1951 Flowsheet ⁽¹⁾

Analyte g/L (except as noted)	Coating Removal Waste	Metal Waste	First Decontamination Cycle (1C) Waste	Second Decontamination Cycle (2C) Waste	224 Building Waste
Plutonium	3.3E-04	2.0E-04	6.0E-07 ⁽⁴⁾	1.6E-07 ⁽⁵⁾	1.68E-04 ⁽⁶⁾
Uranium	0.15		0.235 ⁽⁴⁾	Not reported	2.04E-05
Gamma counts/minute/mL	6.6E+04	1.3E+07	2.3E+06 ⁽⁴⁾	1.13E+04 ⁽⁵⁾	1.13E+02 ⁽⁶⁾
Sodium Aluminate (NaAlO ₂)	95.1				
Sodium Hydroxide (NaOH)	43.6				
Sodium Nitrate (NaNO ₃)	61.8				
Sodium Nitrite (NaNO ₂)	56.0				
Sodium Silicate (NaSiO ₃)	4.3				
Uranyl nitrate (UHN) ⁽³⁾		132			
Fluorine (F)					5.6
Nitrate (NO ₃)		9.7	93.1	61.3	42.4
Sulfate (SO ₄)		24.4	4.73	3.61	0.35
Phosphate (PO ₄)		25.2	26.2	23.0	3.05
Sodium (Na)		83.2	47.3	36.7	36.8
Bismuth (Bi)			2.59	1.31	1.18
Cerium (Ce)			0.030		
Lanthanum (La)					0.49
Manganese (Mn)					0.33
Zirconium (Zr)			0.030		
Iron (Fe)			1.37	1.82	
Chrome (Cr)			0.16	0.06	0.17
Ammonia (NH ₄)			1.98	1.71	0.12
Silicon Hexa-Fluoride (SiF ₆)			4.35	3.67	
Volume per Batch (gallons)	795	2,380	2,040	2,090	2,200

Notes:

⁽¹⁾ See HW-23043⁽²⁾ HW-23043 page 31 notes that uranium is not actually present in this form, but is probably as NaUO₂PO₄ and Na₄(UO₂)₃CO₃.⁽⁴⁾ Pu and Gamma concentrations were calculated from the compositions of tanks 13-4 and 14-3 (HW-23043 pages 20 and 22).⁽⁵⁾ Pu and Gamma concentrations were calculated from the compositions of tanks 18-4 and 19-3 (HW-23043 pages 26 and 28).⁽⁶⁾ Pu and Gamma concentrations were calculated from the compositions of tanks A-4, D-4, B-3, and F-8 (HW-23043 pages 39, 44, 48, and 54)

Table 2-2. Analyses of Bismuth Phosphate Process Supernatants Stored ^(1,2)

Waste Type	Tank	pH	Pu μg/L	Gross Beta millicuries/liter	Gross Gamma millicuries/liter	Date Sampled
Metal Waste	T-101	10.1	70	200 ⁽⁵⁾	70 ⁽⁵⁾	12-12-1946
Metal Waste	T-101	10	35	110	25	7-01-1947
Metal Waste	T-102	9.9	60	120	20	7-01-1947
Metal Waste	T-103	9.8	60	150	20	7-01-1947
1C/CW	B-109	9.9	40	0.65	0.28	3-18-1947
1C/CW	C-112	9.9	12	12	4.4	3-18-1947
2C	B-111	6.9	7.2E-02	2.0E-03	3.0E-03	7-1-1947
2C	B-112	6.8	4.32E?? ⁽³⁾	1.5E-03	3.0E-03	7-1-1947
2C	T-110	Not reported ⁽⁴⁾	15	4.9E+04	30	7-13-1945
2C	T-110	9.8 ⁽⁴⁾	19	6.9E+04	55	7-25-1945
2C	B-110	9.6 ⁽⁴⁾	8.5	7.0E+04	55	7-25-1945

Notes:

⁽¹⁾ See HW-10728 and HW-3-3220.⁽²⁾ Solids formed in each of wastes, settling to the bottom of each tanks. These sample analyses are for the supernatant only and are not representative of the sludges.⁽³⁾ The reported Pu sample analyses for tank B-112 seems to be in error and lacking an exponent in HW-10728.⁽⁴⁾ Prior to October 1945, the 2C waste was neutralized to a pH of approximately 10. The waste collected in tanks 241-T-110, 241-T-111, and 241-T-112 were neutralized to about pH 7 after October 1945 to precipitate bismuth and plutonium (HW-3-3220, page 13).⁽⁵⁾ Reduction in the gross gamma and beta analyses for the metal waste in tank T-101 from sampling in 12-12-1946 to 07-01-1947 is due to decay of short-lived fission products.

Table 2-3. Radionuclide Analyses of Metal Waste and First Decontamination Cycle Waste Supernatants. (2 Sheets)

Tank	Date Filled	Pu µg/cc	Gross Beta µCi/cc	Gross Gamma µCi/cc	Sr µCi/cc	Cs µCi/cc	Ru µCi/cc	Rare Earths + Y - Ce µCi/cc	Ce µCi/cc	Nb µCi/cc	Zr µCi/cc	Te µCi/cc
Analyses of Metal Waste Supernatant Following Uranium Extraction (1)												
C-106	Not specified				0.44	54.2						
BX-108	Not specified				0.26	132.4						
BX-109	Not specified				1.08	56.3						
C-112	Not specified				1.20	25.8						
C-109	Not specified				0.46	40.7						
C-111	Not specified				0.10	34.5						
Average Concentrations for Metal Waste												
					0.59	57.3						
Analyses of First Decontamination Cycle Waste Mixed with Coating Removal Waste (2)												
B-107	8-1945	17E-03	0.135	0.055	0.011	0.10						
T-107	9-1945	1.5E-03	0.170	0.093	0.0013	0.20						
B-108	12-1945	2.0E-02	0.183	0.044	0.022	0.12						
T-108 (Top)	12-1945	2.0E-02	0.25	0.073	0.12	0.17	0.0066	0.047	0.007	0.0018	0	1.2E-05
T-108 (Bottom)	12-1945	2.0E-02	0.25	0.070	0.12	Not reported	0.0065	0.029	0.0066	0.0024	0	3E-05
T-109	3-1946	2.6E-03	0.14	0.082	0.00038	0.15						
B-109	4-1946	1.8E-02	0.16	0.051	0.01	0.11						
T-104 (Top)	7-1946	3E-03	0.51	0.130	0.00013	0.13	0.058	0.004	0.051	0.028	0.010	2.4E-05
T-104 (Bottom)	7-1946	3E-03	0.52	0.160	0.00037	Not reported	0.059	0.003	0.050	0.028	0.015	3.6E-05
C-110	8-1946	2E-03	0.14	0.0067	0.00026	0.11						
C-111	11-1946	4.2E-03	0.16	0.069	0.01	0.13						
C-112	4-1947	3.1E-03	0.14	0.064	0.005	0.13						

Table 2-3. Radionuclide Analyses of Metal Waste and First Decontamination Cycle Waste Supernatants. (2 Sheets)

Tank	Date Filled	Pu µg/cc	Gross Beta µCi/cc	Gross Gamma µCi/cc	Sr µCi/cc	Cs µCi/cc	Ru µCi/cc	Rare Earths + Y - Ce µCi/cc	Ce µCi/cc	Nb µCi/cc	Zr µCi/cc	Te µCi/cc
U-110	4-1947	2.1E-04	0.13	0.069	0.00011	0.17						
U-111	10-1947	3.4E-04	0.12	0.060	0.00023	0.14						
TX-109 ⁽³⁾	9-1949	2.7E-05	2.8	2.2	0.00087	0.27	0.34	0.0085	0.0035	0.34	1.2	8E-05
Average Concentrations for IC / CW		7.67E-03	0.39	0.22	0.02	0.15						

Notes:

- (1) HW-36717, 1955, *Decontamination of Uranium Recovery Process Stored Wastes Interim Report*, General Electric Company, Richland, Washington.
(2) HW-20195, 1951, *Radioactive Content of Stored Bismuth Phosphate First Cycle Waste Supernatants*, General Electric Company, Richland, Washington.
(3) Tank TX-109 exhibits higher gross beta and gross gamma radioactivity since this tank was sampled shortly after filling and the short-lived fission products (e.g., Ru, Nb, and Zr) had not decayed appreciably.

2.2 CONTINUOUS SOLVENT EXTRACTION PROCESSES - REDOX AND PUREX

The REDOX plant was operated from 1952 through 1966, while the PUREX plant was operated from 1956 through 1988 to reprocess spent nuclear fuels. Spent nuclear fuel elements were dissolved to remove the fuel cladding. The cladding removal waste (designated as CW) from the aluminum clad fuel was inherently alkaline and did not require neutralization before transfer to single-shell and later (after 1983) double-shell underground storage tanks. Zirconium (Zr) clad fuel, Zircaloy^{®1} (98.5% Zr and 1.5% Sn) was also processed in the PUREX and REDOX plants. The Zircaloy[®] clad fuel was dissolved in a solution of ammonium fluoride and ammonium nitrate. The cladding waste was neutralized (designated as NCRW) by addition of sodium hydroxide solution before transfer to single-shell and later (after 1983) double-shell underground storage tanks (PFP-P-020-00001). A solvent extraction process was operated at each facility to separate uranium and plutonium from dissolved spent nuclear fuels.

The REDOX plant used a methyl isobutyl ketone solvent whereas the PUREX plant used a tributyl phosphate solvent in a normal paraffin hydrocarbon diluent. The fission products and impurities separated during the uranium and plutonium solvent extraction process were neutralized and transferred initially to single-shell, and later (after 1983), to double-shell underground storage tanks, forming supernatant and sludges within the tanks. The supernatant, known as REDOX supernatant neutralized (RSN) and PUREX supernatant neutralized (PSN) were stored separately in the 200 West and 200 East Area tank farms, respectively. The plutonium solutions generated at REDOX and PUREX plants were initially transferred to the 231-Z building and subsequently to the 234-5Z building (Z Plant) for further processing. Uranium solutions were transferred to 224-U building (UO₃ Plant) for conversion to an oxide and transfer to offsite facilities for re-use in the fabrication of nuclear fuel.

2.3 HOT SEMIWORKS

The Hot Semiworks, 201-C building, was constructed in 1951 to 1952 as a research and test facility for the REDOX and TBP Plant chemical separations processes (HW-22955). The Hot Semiworks was originally operated from November 1952 to October 1953 as a pilot facility to research and demonstrate the REDOX chemical separations process (HW-31767). The facility was modified in 1953 and operated from May 1955 (HW-38768-RD) through March 1956 (HW-49673-RD) as a research and demonstration facility for the PUREX chemical separations process. Following completion of the PUREX chemical separation process research and development activities, a maintenance program was initiated at the Hot Semiworks facility for plant improvements. This maintenance program was completed in July 1957 and the Hot Semiworks was placed in standby mode in July 1957 (HW-52860).

The highly radioactive waste from the REDOX process research runs were concentrated and transferred to an underground storage tank, TK-70 (also designated as 241-CX-70) located at the

¹ Zircaloy[®] is a trademark of Teledyne Wah Chang, Albany, Oregon.

Hot Semiworks 241-CX tank farm facilities. The highly radioactive wastes from the PUREX process research runs were concentrated and transferred to C-200 series tanks (see Section 3.4). Process condensates and cooling water from equipment in the Hot Semiworks were transferred to crib 216-C-1 (HW-48518, page 21). Process condensates from the evaporation of highly radioactive waste were transferred to crib 216-C-6 (HW-48518, page 21). Organic solvent waste from the PUREX process research runs was transferred from Hot Semiworks building 276-C to crib 216-C-4 starting in October 1955 (HW-48518, page 22).

In 1961, the Hot Semiworks was modified to incorporate solvent extraction and ion exchange columns for demonstrating strontium purification processing (HW-68786). The Hot Semiworks was operated from May 1961 to October 1961 to demonstrate strontium separation from PUREX waste and transfer the separated strontium to Oak Ridge National Laboratory (HW-72666-RD Part 1, page 6). The Hot Semiworks facility was renamed the Strontium Semiworks facility in 1961 and was operated until 1967 to separate strontium from various waste solutions.

The Strontium Semiworks along with a renovated portion of B-Plant, PUREX head-end, and the 244-CR vault were used from 1961 through 1967 to separate strontium-90, cesium-137, cerium-144, and promethium-147 from various waste solutions (HW-71179). The head-end of PUREX was used to separate strontium-90 from high-level waste with the strontium-90 solution transferred to the 244-CR vault for storage and decay of strontium-89 and eventual transfer to the Strontium Semiworks. The 244-CR vault was also used to transfer a solution containing strontium and rare earths from PUREX to B-Plant for separating the strontium-90 and rare earths (mixture of cerium-144 and promethium-147) into separate solutions (HW-77016).

The strontium-90 solution and rare earths solution were transferred separately to the Strontium Semiworks for further purification and load-out onto casks (HW-78987-REV, page 12 and HW-81481, page 38). Solvent extraction equipment was operated under various flowsheet conditions to purify separate batches of strontium-90, cerium-144, and promethium-147.

PUREX high-level waste solutions that were stored in C Farm were also passed through a shielded cask that contained Decalso^{®2} ion exchange material to separate cesium-137. Building 801-C in C-Farm was used to contain the shielded cask while cesium was loaded onto the ion exchange material (HW-71333). A cask station at the PUREX facility was also used to load cesium onto ion exchange material.

The Strontium Semiworks was also used in conjunction with the 801-C cask station to demonstrate the separation of technetium-99 from alkaline high-level waste solutions. Approximately 1-kg of technetium-99 was separated from high-level waste that was stored in C-Farm SSTs in October 1963 (HW-79377-C, page C-7 and HW-79480, page G-2). The high-level waste solution was passed through a shielded cask in the 801-C building that contained Decalso[®] ion exchange material to separate cesium. The effluent solution from the cesium cask was then passed through a separate shielded cask in the 801-C building that contained IRA-401^{®3} ion exchange material, which adsorbed technetium from the waste solution.

² Decalso[®] is a synthetic, sodium aluminosilicate gel manufactured by the Permutit Company, New York.

³ IRA-401[®] is a styrene, di-butyl benzene ion exchange bead manufactured by the Rohm and Haas Company, Philadelphia, Pennsylvania.

The Strontium Semiworks received the cask that was loaded with technetium in November 1963, eluted and concentrated the technetium, which was then loaded into a smaller cask for transfer to the Hanford Laboratories located in the 300 Area (HW-79768, page G-2). A second campaign to recover an additional 1-kg of technetium-99 from high-level waste stored in C-Farm was conducted in August through September 1964 in the same manner as the first campaign (HW-83876, page B-2 and HW-84354, page B-1).

In addition to the technetium-99 campaigns, the Strontium Semiworks used the installed solvent extraction equipment to purify a solution containing a mixture of americium, cerium, and rare earths (HAN-98529-DEL, page AIII-3). The solution containing a mixture of americium, cerium, and rare earths had been separated at the REDOX facility while reprocessing the irradiated fuel from the Shippingport reactor (ARH-1354). The americium, cerium, and rare earths were shipped the Hanford Laboratories in the 300 Area.

Tanks 241-C-107, 241-C-108, 241-C-109, 241-C-111, and 241-C-112 all received highly radioactive waste solutions from the Strontium Semiworks from 1961 through 1967 (HW-72625, page 4, HW-74647, page 4, HW-76223, page 4, HW-78279, page 4, HW-80379, page 4, HW-83308, page 4, RL-SEP-260, page 4, RL-SEP-659, page 4, RL-SEP-821, page 4, RL-SEP-923, page 4, ISO-226, page 4, ISO-404, page 4, ISO-538, page 4, ISO-674, page 4, ISO-806, page 4, ISO-967, page 4, ARH-95, page 5, and ARH-326, page 5). The C-200 series tanks did not receive any waste from the Strontium Semiworks operations.

After being used successfully to separate various fission products from waste solutions, the Strontium Semiworks was deactivated beginning from October 1967 (HAN-98918-DEL, page AIII-3) through November 1967 (HAN-99196-DEL, page AIII-3).

3.0 WASTE TRANSFER INTO AND WASTE REMOVAL FROM TANKS 241-C-201 THROUGH 241-C-204

This section provides a brief description of tanks 241-C-201 through 241-C-204 and summarizes waste transfers into and waste removal from these tanks. In order to determine the origins of the wastes presently stored in tanks 241-C-201 through 241-C-204, declassified, historical reports for the Hanford Site were reviewed. With the exception of the waste status summary reports, all reports cited in this section are available electronically from the Hanford Declassified Document Retrieval System at <http://www2.hanford.gov/declass/>. Full-text copies of the waste status summary reports cited in this section available from RIM services (509-376-5440). The results of the present review of historical records are compared with a previous review of historical report that was conducted in 1980 as part of WHC-MR-0132, *A History of the 200 Area Tank Farms*.

3.1 DESCRIPTION OF TANKS 241-C-201 THROUGH 241-C-204

The SSTs 241-C-201 through 241-C-204 were originally constructed in 1944 as part of the Manhattan Project (HW-10475-C, chapter IX). These four tanks, along with twelve larger SSTs, comprise the C Tank Farm. Tanks 241-C-201 through 241-C-204 are twenty-foot diameter underground tanks made of re-enforced concrete with a steel liner on the bottom and sides. Each tank has a design capacity of 55,000 gallons at a liquid depth of twenty-four feet.

3.2 METAL WASTE TRANSFERS INTO TANKS 241-C-201 THROUGH 241-C-204

The C Tank Farm was originally constructed to receive waste from the planned, 221-C Bismuth Phosphate Separations plant and the planned 224-C Concentration building. However, the 221-C and 224-C buildings were never constructed. The C Tank Farm was originally isolated from all other tank farms since underground piping from the 241-B-154 diversion box to the 241-C-154 diversion box in C Tank Farm was not installed (HW-10475-C, chapter IX pages 904).

In September 1947, construction activities were completed to permit utilization of the C-200 series tanks for storage of waste from the 221-B Bismuth Phosphate Separations plant (HW-7795-DEL, page 26). Tanks 241-C-201 through 241-C-204 began to receive metal waste from the 221-B Bismuth Phosphate Separations plant in November 1947, with these tanks reported as 25 percent full on November 30, 1947 (HW-8267-DEL, page 29). Metal waste continued to be transferred from the 221-B Bismuth Phosphate Separations plant into tanks 241-C-201 through 241-C-204 in December 1947 (HW-8438-DEL, page 27) and January 1948 (HW-8931-DEL, page 28). On December 31, 1947, these tanks were reported as 68.6 percent full (HW-8438-DEL, page 27); and on January 31, 1948, these tanks were reported as being 100 percent full of metal waste (HW-8931-DEL, page 28). No additional transfers of metal waste were made into these tanks. Sludge and liquid level measurements were made in each of the C-200 series tanks on May 27, 1948 (HW-10154) and are summarized in Table 3-1.

Table 3-1. Metal Waste Sludge and Liquid Level Measurements.

Tank	Sludge Level	Liquid Level ⁽¹⁾
241-C-201	4-feet, 9-inch	17-feet, 3-inch
241-C-202	4-feet, 3-inch	17-feet, 9-inch
241-C-203	4-feet, 3-inch	17-feet, 9-inch
241-C-204	3-feet, 3-inch	19-feet, 9-inch

Note: ⁽¹⁾ Calculated by difference from reported sludge level and total waste level

3.3 METAL WASTE RETRIEVAL FROM TANKS 241-C-201 THROUGH 241-C-204

Beginning in February 1952, metal waste was removed from these SSTs. Removal of metal waste sludges from the SSTs was accomplished using a hydraulic mining technique known as sluicing. The slurry of metal waste solids and liquid was transferred through a series of

underground tanks contained in concrete vaults to the 221-U building for recovery of uranium (WHC-SD-WM-TI-302, *Hanford Waste Tank Sluicing History*).

In December 1953, the metal waste supernatant present in SSTs 241-C-201 through 241-C-204 was transferred to SST 241-C-106 in preparation for sluicing the metal waste solids present in these tanks (HW-45165-RD, page 69 and HW-30498, page 4). Sluicing operations in tank 241-C-202 were conducted from January 9 through January 14, 1954 (HW-30851, page 4), with a total of approximately 13 tons of uranium removed from this tank (HW-45163-RD, page 2). Sluicing operations were next initiated in tank 241-C-203 on January 15, 1954 (HW-45163-RD, page 4 and HW-36979-B, page 94) and completed the week of January 28, 1954 (HW-45163-RD, page 8). Sluicing of tank 241-C-204 was to be initiated in February 1954, but a failure of the sludge jet precluded sluicing in this tank until later in calendar year 1954 (HW-45163-RD, page 10). Instead, sluicing operations were initiated on February 15, 1954 in tank 241-C-201 (HW-45163-RD, page 12 and HW-36979-B, page 86) and continued until the tank was declared empty on March 17, 1954 (HW-31374, page 4 and HW-36979-B, page 78).

Metal waste sludge removal from tank 241-C-204 was delayed until November 1954 due to other higher priority tasks in the tank farms. A pump was installed in tank 241-C-204 in November 1954 (HW-45163-RD, page 74 and HW-36979-B, page 13) and approximately 7,000-gallons of waste was transferred to SST 241-C-104 in preparation for resuming sluicing operations (HW-33904, page 3). Sluicing operations in tank 241-C-204 were started in January 1955 (HW-36979-C, page 2) and completed in February 1955 (HW-36979-C, page 7). An inspection of the tank interior in February 1955 indicated that all sludge had been removed (HW-36979-C, page 18).

The waste status summary report for February 1955 indicates that each of the C-200 series tanks was empty as of February 28, 1955 (HW-35628, page 4). Uranium inventory records indicate that of the 52 tons of uranium in these tanks, that 54 tons were removed (HW-63090, pages 83 – 86). Hanford Site personnel used a periscope optic system to inspect inside SSTs to verify the removal of metal waste sludges as shown in Figure 3-1. The recovery of metal waste sludge from SSTs was very successful and was summarized in January 1957 (HW-47988) as follows:

“... we are forced to seriously question the validity of the residual heels as determined by visual inspection. A removal efficiency of greater than 99 percent is difficult to give credence when one considers the mining technique employed. In addition, it was not expected at any time that the tank inspection method which had to be used would give this degree of accuracy within a one percent range.”

“Sluicing was carried on until uranium removal was essentially zero ... Visual inspections through periscope were made for mounds of material and when found, these were removed by further sluicing ... Estimates of residual material cannot be made with a high degree of accuracy, but it can be said that no mounds of material were left and, within the capability of equipment, methods and the sluicing media, all recoverable material was removed.”

3.4 HOT SEMIWORKS WASTE TRANSFERS INTO TANKS 241-C-201 THROUGH 241-C-204

From May 1955 (HW-38768-RD) through March 1956 (HW-49673-RD), the Hot Semiworks was used as a research and demonstration facility for the PUREX chemical separations process. The highly radioactive waste from the solvent extraction process conducted in the Hot Semiworks was concentrated to recover nitric acid, neutralized by addition of sodium hydroxide solution and transferred to tanks 241-C-201 through 241-C-204.

Tank 241-C-201 first received highly radioactive waste from the Hot Semiworks beginning in May 1955 (HW-37143, page 4) and was reported as being filled with 54,500-gallons of waste as of November 30, 1955 (HW-40208, page 4). Tank 241-C-202 was reported to first receive waste from the Hot Semiworks beginning in November 1955 (HW-40208, page 4) and was reported as being filled with 54,500-gallons of waste in May 1956 (HW-43420, page 4). Tanks 241-C-203 and 241-C-204 were also reported to have received Hot Semiworks waste beginning in December 1955 (HW-40816, page 4), and each tank was reported to contain 34,500-gallons of waste as of November 1956 (HW-47052, page 4).

Tanks 241-C-203 and 241-C-204 also received in November 1955 (HW-40208, page 4) waste from cold uranium runs conducted as part of startup operations at the PUREX plant. The PUREX plant was using uranium that had not been irradiated (i.e., cold uranium) as feed for the startup tests, as documented in the Atomic Energy Commission 200 Area weekly reports for November 1955 and December 1955 (HAN-61678, reports #44 thru 50). As noted in the Tank Farm Waste Status Summary Report for November 1955 (HW-40208, page 4) and the Summary Report – PUREX Plant Cold Runs (HW-43454, page 9), the waste from the cold uranium runs at the PUREX plant was discharged from tanks 241-C-203 and 241-C-204 to a crib.

Tanks 241-C-201 through 241-C-204 did not receive any additional waste after November 1955.

3.5 SUPERNATANT REMOVAL FROM TANKS 241-C-201 THROUGH 241-C-204

Approximately 19,000-gallons of supernatant was transferred from tank 241-C-203 to tank 241-C-109 in January through March 1970 (ARH-1666 A, page 5). The supernatant contained in tanks 241-C-201 through 241-C-204 was pumped to tank 241-C-104 in April through June 1970 (ARH-1666 B, page 5). The volume of supernatant removed from tanks 241-C-201, 241-C-202, 241-C-203, and 241-C-204 was 54,000, 55,000, 12,000, and 14,000-gallons, respectively. With the exception of tank 241-C-204, these tanks contained only a heel of supernatant and sludge following these transfers. Tank 241-C-204 was reported to still contain 41,000-gallons of supernatant in June 1970.

On July 10, 1977, the supernatant present in tank 241-C-204 was transferred to an unidentified location, leaving only 3,000-gallons of supernatant in this tank (RHO-CD-213, page 30-24-02). In October 1980, supernatant was again pumped from tanks 241-C-201 through 241-C-204 into tank 241-C-106 using a submersible pump (RPT-120180, RPT-100180, and 65260-80-0931).

3.6 CURRENT REVIEW OF WASTE TRANSFER RECORDS COMPARED WITH OTHER REVIEWS

Historical records of waste transfers into, from, and among the 200 Area tank farms were compiled and reported in WHC-MR-0132, *A History of the 200 Areas Tank Farms*, LA-UR-96-3860, *Hanford Tank Chemical and Radionuclide Inventories: HDW Model Rev. 4*, and in particular for the C Tank Farm in WHC-SD-WM-ER-313, Revision 1, *Supporting Document for the Historical Tank Content Estimate for the C-Tank Farm*. These previous reviews to determine the tank contents for tanks 241-C-201 through 241-C-204 generally refer to WHC-MR-0132 and provide no new references regarding waste transfers into these tanks.

In general, the waste transfer records summarized in WHC-MR-0132 and WHC-SD-WM-ER-313, Revision 1 for SSTs 241-C-201 through 241-C-204 are in agreement with the information presented in this report. Specific discrepancies between these previous reports and the information present in this document are discussed in the following subsections.

3.6.1 COLD URANIUM WASTE DISCHARGE TO TANKS 241-C-203 AND 241-C-204

WHC-MR-0132 and WHC-SD-WM-ER-313, Revision 1 do not identify the transfer of cold uranium run waste from startup of the PUREX plant into tanks 241-C-203 and 241-C-204 and the subsequent discharge of this waste to a crib, as in November 1955. This probably is because WHC-MR-0132 reported the status of each tank on a quarterly basis instead of on a monthly basis. WHC-SD-WM-ER-313, Revision 1 relied upon WHC-MR-0132 for waste transfer information and also did not identify the transfer of cold uranium run waste from startup of the PUREX plant into tanks 241-C-203 and 241-C-204.

3.6.2 UNEXPLAINED WASTE VOLUME INCREASE IN TANK 241-C-204

WHC-MR-0132 reports the volume of waste contained in tank 241-C-204 increased from 36,000-gallons to 57,000-gallons in the fourth quarter of 1967, without an explanation. The historical waste transfer record for this period report the increase in the volume of waste contained in tank 241-C-204 without an explanation (ARH-326, page 5). The historical waste transfer record for the fourth quarter of 1967 also report that waste from the Strontium Semiworks was transferred to tank 241-C-107, not 241-C-204 (ARH-326, page 5). The waste level measure reported for tank 241-C-204 in the fourth quarter of 1967 seems to be in error.

Supporting this assumption are previous increases and decreases reported for the waste volume in tank 241-C-204 without any known waste transfers. Tank 241-C-204 was reported as containing 54,000-gallons of waste in March 1957 (HW-49523, page 4) and then only 33,000-gallons of waste in April 1957, without any waste removal occurring (HW-50127, page 4). The decrease in the measured volume of waste in tank 241-C-204 reported in 1957 was attributed to a new electrode measurement of the waste level. It seems that difficulties were encountered in obtaining an accurate measurement of the waste volume in tank 241-C-204. The Hanford Defined Waste (HDW) model, *Hanford Tank Chemical and Radionuclide Inventories: HDW Model Rev. 4*, LA-UR-96-3860, assumes the increase in the volume of waste contained in tank

241-C-204 is attributed to waste transferred from the Strontium Semiworks. This assumptions now seems incorrect.

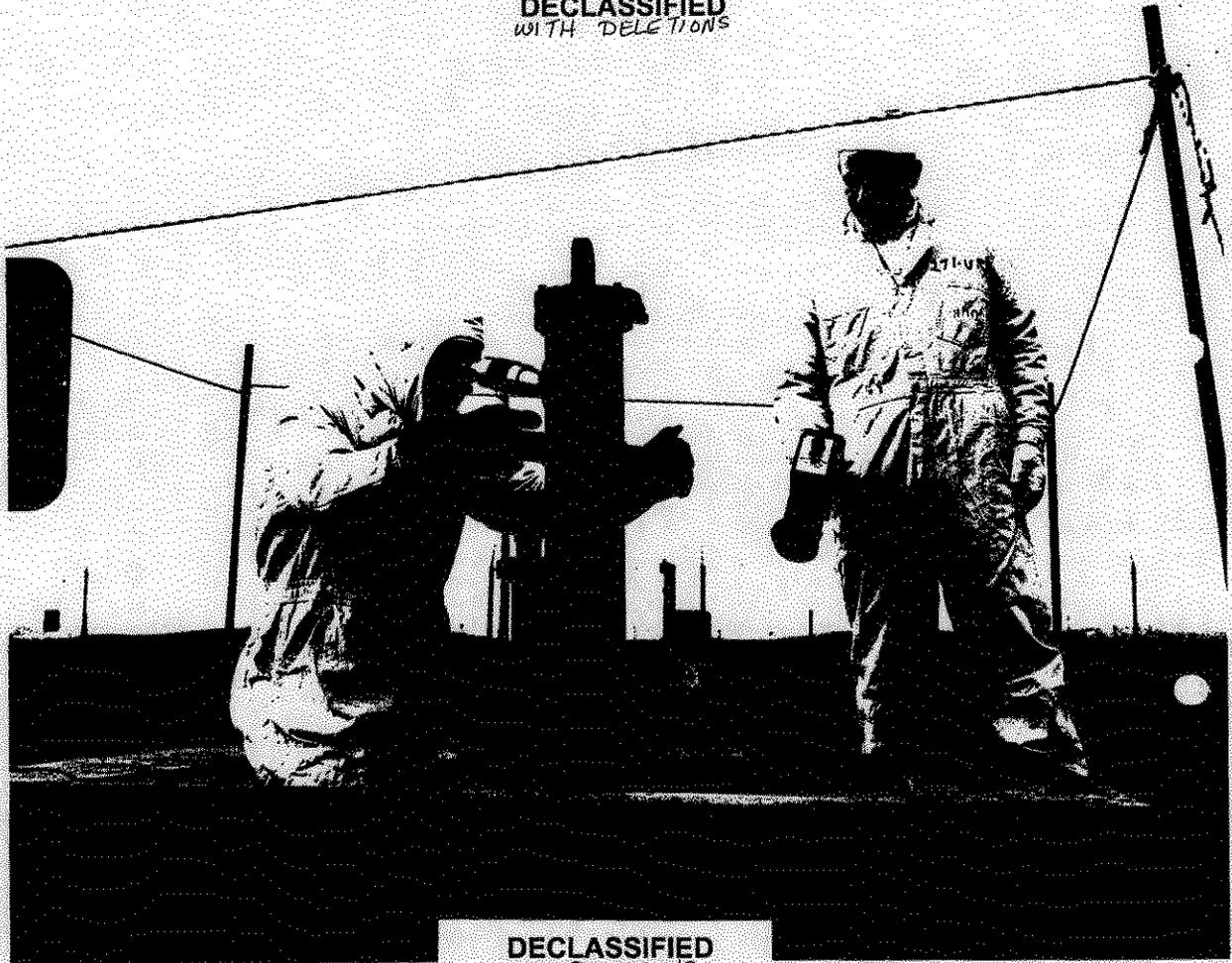
3.6.3 METAL WASTE HEELS

The Hanford Defined Waste (HDW) model, *Hanford Tank Chemical and Radionuclide Inventories: HDW Model Rev. 4*, LA-UR-96-3860, indicates that tanks 241-C-201 through 241-C-204 presently contain Hot Semiworks waste. The HDW model, Revision 4 also indicates that tanks 241-C-201, 241-C-203, and 241-C-204 each contain a heel of metal waste sludge of 1,000-gallons (~5 inches), 4,000-gallons (~20 inches), and 2,000-gallons (~10 inches), respectively. It is interesting to note that the HDW model, Revision 4 does not indicate a heel of metal waste in tank 241-C-202.

The HDW model, Revision 4 does not provide a basis for assuming a heel of metal waste sludge remains in tanks 241-C-201, 241-C-203, and 241-C-204 and seems to be inconsistent, since tank 241-C-202 is not identified as containing a heel of metal waste sludge. The assumption that tanks 241-C-201, 241-C-203, and 241-C-204 each contain a heel of metal waste seems arbitrary, given the fact that these tanks were sluiced in 1954 through 1955 (see Section 3.3) to remove metal waste. As noted in section 3.5, visual observations of these tanks following sluicing was conducted and no metal waste heels were observed. The visual observations made of the interiors of tanks 241-C-201 through 241-C-204 would have identified the 5 to 20 inches of metal waste if present in each tank. The visual observation of each tank following sluicing operations reported that each tank was empty. Therefore, it is unlikely that the volume of metal waste assumed in HDW model, Revision 4 was left as a heel in tanks 241-C-201, 241-C-203, or 241-C-204.

Figure 3-1. Hanford Employee Using Periscope Optics Unit to Inspect Single-Shell Tank Interior Following Sludge Removal by Sluicing.

DECLASSIFIED
WITH DELETIONS



DECLASSIFIED
WITH DELETIONS

A final look through a periscope into the interior of an underground waste storage tank is taken by a General Electric atomic worker checking the position of an accumulation of uranium-bearing sludge. Sluicing operations to recover the valuable material create fog and haze, cutting visibility to zero. Lack of visibility and the presence of radiation were two of the biggest problems General Electric engineers had to overcome during their uranium recovery program. Recover of valuable uranium from underground tanks was successful.

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4.0 SUMMARY

The SSTs 241-C-201 through 241-C-204 were filled with metal waste from September 1947 through January 1948. Metal waste was hydraulically mined from these tanks from March 1953 through January 1955. The metal waste sludge and supernatant was dissolved in acid in the 244-BXR vault and then transferred to the 221-U building where uranium was recovered from these wastes using a tributyl phosphate-based solvent extraction process. After completing the removal of metal waste, each tank was visually inspected and determined to be empty. However, given the periscope optics inspection method, residual metal waste could have been left in each tank.

Tanks 241-C-203 and 241-C-204 received cold uranium (i.e., uranium that had not been irradiated in a reactor) waste from plutonium-uranium extraction (PUREX) startup testing in November 1955. The cold uranium waste was removed from tanks 241-C-203 and 241-C-204 in December 1955. Tanks 241-C-201 through 241-C-204 were then used from May 1955 through October 1956 to receive and store waste originating from research and development activities conducted at the 201-C Hot Semiworks facility in the 200 East Area of the Hanford Site. The cold uranium waste was removed from tanks 241-C-203 and 241-C-204 before transfers of Hot Semiworks waste into these tanks was conducted.

Tanks 241-C-201 through 241-C-204 were not used to receive waste after being filled with waste from the Hot Semiworks. The liquid in tanks 241-C-201, 241-C-202, and 241-C-204 was transferred to single-shell tank 241-C-104 in 1970. The liquid in tank 241-C-204 was transferred to single-shell tanks 241-C-104 and 241-C-109 in 1970. Residual liquids were subsequently transferred from these tanks into single-shell tank 241-C-106 in 1980.

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