Origin and Classification of Wastes in U-200 Series Single-Shell Tanks

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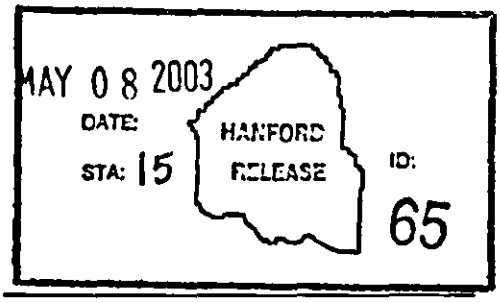
U.S. Department of Energy Contract DE-AC27-99RL14047

EDT/ECN: EDT N.S. 5-8-03 ECN-636113 UC: 2000
Cost Center: 7G300 Charge Code: 501546
B&R Code: N/A Total Pages: 24

Key Words: Waste, Classification, Origin, SST, Single-Shell Tanks, U-201, U-202, U-203, U-204

Abstract:
Tanks U-201, U-202, U-203, and U-204 contain waste from dissolving the coating present on irradiated nuclear fuel at the REDOX plant. Tank U-204 also contains waste from flushing a diversion box. These wastes are not high-level waste. Based on available sample analyses, these wastes are low-level waste.



Nancy A. Fouad 5-8-03
Release Approval Date

Release Stamp

Approved For Public Release

RPP-13362
Revision 0

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Date Published
January 2003

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Prepared for the U.S. Department of Energy
Office of River Protection

Contract No. DE-AC27-99RL14047

EXECUTIVE SUMMARY

A review of waste transfer documents was conducted to determine the origin of waste transferred into single-shell tanks 241-U-201 through 241-U-204. Based on this review, the wastes stored in single-shell tanks 241-U-201 through 241-U-204 can be classified as coating removal waste generated from chemical dissolution of irradiated nuclear fuel at the Hanford site REDOX plant.

Tanks 241-U-201 through 241-U-204 were originally constructed as part of the Manhattan Project from 1944 to 1945, but were not used until 1954. Single-shell tank 241-U-204 received approximately 35,000-gallons of waste resulting from a flush of a diversion box in January 1954. The flush solution from the diversion box was categorized as second decontamination cycle waste from operation of the 221-T Bismuth Phosphate plant. Tank 241-U-204 then received coating removal waste from operation of the reduction-oxidation (REDOX) process in the 202-S building in April and May 1956. Single-shell tanks 241-U-201 through 241-U-203 sat unused until January 1956 when they were activated to store coating removal waste from operation of the REDOX process in the 202-S building. These tanks were filled with coating removal waste from February 1956 through March 1956. No additional transfers of waste into these tanks occurred. The supernatant present in tanks 241-U-201 through 241-U-204 was removed in 1977 and processed through the 242-S Evaporator, leaving a heel of approximately 4,000 to 5,000 gallons in total of supernatant and sludge in each tank.

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1C	first decontamination cycle
2C	second decontamination cycle
CW	coating removal waste
g/L	grams per liter
lbs	pounds
REDOX	reduction-oxidation
MW	metal waste
µCi/cc	microcuries per cubic centimeters
µCi/g	microcuries per gram
µCi/L	microcuries per liter
µg/cc	micrograms per cubic centimeters
µg/L	micrograms per liter
ηCi/g	nanocuries per gram
%	percent

1.0 INTRODUCTION

The origin of the wastes in tanks 241-U-201 through 241-U-204 is important to determining the disposition of these wastes and the waste storage tanks.

Section 2.0 discusses the origin of waste transferred into and removed from single-shell tanks 241-U-201 through 241-U-204. Section 3.0 provides a description of the different types of wastes that were generated at the Hanford Site chemical processing plants and transferred to the underground storage tanks 241-U-201 through 241-U-204. Section 4.0 discusses the gross alpha analyses of these wastes. Section 5.0 summarizes the waste transfer records and gross alpha analyses for the wastes stored in single-shell tanks 241-U-201 through 241-U-204.

2.0 WASTE TRANSFER INTO AND WASTE REMOVAL FROM TANKS 241-U-201 THROUGH 241-U-204

This section provides a brief description of tanks 241-U-201 through 241-U-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-U-201 through 241-U-204, publicly available 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/. The waste status summary reports are available only as photocopies from the Hanford Site central files (509-376-5440).

2.1 DESCRIPTION OF TANKS 241-U-201 THROUGH 241-U-204

Single-shell tanks 241-U-201 through 241-U-204 were originally constructed in 1944 as part of the Manhattan Project (see HW-10475-C, *Hanford Technical Manual Section C*, chapter IX). These four tanks, along with twelve larger single-shell tanks, comprise the U Tank Farm. Tanks 241-U-201 through 241-U-204 are twenty-foot diameter underground tanks made of reinforced 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.

2.2 WASTE TRANSFERS INTO TANKS 241-U-201 THROUGH 241-U-204

The U Tank Farm was originally constructed to receive waste from the 221-U Bismuth Phosphate Separations plant and the planned 224-U Concentration building. However, the 221-U and 224-U buildings were not used to reprocess spent nuclear fuel using the bismuth phosphate flowsheet (HW-10475-C, chapter IX, page 904). Instead, U Tank Farm was connected to receive waste from the 221-T Bismuth Phosphate plant and the 202-S Reduction-

Oxidation (REDOX) plant (see Section 3.0). Single-shell tanks 241-U-201 through 241-U-204 were not used to receive any waste solutions until January 1954.

The Atomic Energy Commission monthly report for January 1954 (HAN-62359-DEL, February 4, 1954, page 44) indicates that all four tanks were empty. The Atomic Energy Commission monthly report for February 1954 indicates that tank 241-U-204 received 35,000 gallons of waste from a diversion box flush (HAN-62359-DEL, March 3, 1954, page 58). This is substantiated by the waste transfer records for the Hanford Site tank farms which indicate that tank 241-U-204 received 35,00-gallons of 2C (second decontamination cycle) waste as a result of a diversion box flush (HW-30851, page 4). Waste transfer records for the Hanford Site tank farms from February 1954 through January 1956 indicate that no additional transfers of waste into tanks 241-U-201 through 241-U-204 for this period (HW-31126, HW-31374, HW-31811, HW-32110, HW-32389, HW-32697, HW-33002, HW-33396, HW-33544, HW-33904, HW-34412, HW-35022, HW-35628, HW-36001, HW-36553, HW-37143, HW-38000, HW-38401, HW-38926, HW-39216, HW-39850, HW-40208, HW-40816, and HW-41038).

In February 1956, approximately 48,000-gallons of REDOX coating removal waste was transferred from single-shell tank 241-U-110 into tank 241-U-201 (HW-41812, page 6, and HW-83906B-RD, pages 13 and 16). In March 1956, two, approximately 51,000-gallon batches of coating removal waste were transferred from tank 241-U-110 into tanks 241-U-202 and 241-U-203 (HW-42394, page 6, and HW-83906B-RD, page 30). In April 1956, no waste was added to any of the U-200 series tanks. In May 1956, approximately 18,000-gallons of REDOX coating removal waste was transferred from tank 241-U-110 into tank 241-U-204 (HW-43490, page 6, and HW-83906B-RD, pages 45 and 52). Over time, solids formed in the coating removal waste present in tanks 241-U-201 through 241-U-204 and settled to the bottom of each tank.

It should be noted that tank 241-U-110 also contained first decontamination cycle (1C) / coating waste (CW) sludge from spent nuclear fuel reprocessing activities at the 221-T Bismuth Phosphate plant. Tank 241-U-110 had previously contained 1C/CW supernatant as well as 1C/CW sludge, but the supernatant was removed in April 1952 and processed in the 242-T Evaporator for concentration and storage in another single-shell tank (241-TX-113). Tank 241-U-110 was reported as being "down to sludge on April 28, 1952" (HW-27838, page 11), leaving approximately 336,000 gallons of 1C/CW sludge in this tank. By August 1955, the 1C/CW sludge had settled further in tank 241-U-110 and was reported to be approximately 319,000-gallons (HW-38926, page 6). Beginning in September 1955, tank 241-U-110 was used to receive coating removal waste from the REDOX plant (HW-39216, page 6). The REDOX coating removal waste was transferred to tanks 241-U-201 through 241-U-204, leaving the 1C/CW sludge in tank 241-U-110. No 1C/CW sludge was transferred to tanks 241-U-201 through 241-U-204 as evident by the sludge volume reported at 319,000 gallons in tank 241-U-110 following each of the coating waste transfers.

A review of available waste transfer records for the 200 Area tank farms (LA-UR-96-3860, LA-UR-97-311, WHC-MR-0132, and HNF-SD-WM-ER-325) indicates that no additional transfers of waste into tanks 241-U-201 through 241-U-204 occurred. Review of these documents also supports the previously discussed waste transfers into these tanks.

2.3 WASTE TRANSFERS FROM TANKS 241-U-201 THROUGH 241-U-204

The supernatant present in tanks 241-U-201 through 241-U-204 was removed in 1977 and processed through the 242-S Evaporator. Tanks 241-U-201 through 241-U-204 contained approximately 1,000-gallons of supernatant and approximately 2,000 to 4,000 gallons of sludge per tank following the removal of the supernatant in 1977 (WHC-MR-0132). Table 1 indicates recent measurements of the sludge and supernatant volumes in each tank, as report in June 2002 (HNF-EP-0182). Table 2 provides the results of gross alpha analyses that were conducted on core samples obtained in 1995 from the waste in each tank. Analyses of the individual transuranic elements present in the waste stored in tanks 241-U-201 through 241-U-204 were not conducted.

Table 1. Waste Volume Estimates for U-200 Series Single-Shell Tanks (HNF-EP-0182).

Tank	Total Volume (Gallons)	Sludge Volume (Gallons)	Liquid (Gallons)
241-U-201	5,000	4,000	1,000
241-U-202	4,000	3,000	1,000
241-U-203	4,000	3,000	1,000
241-U-204	4,000	3,000	1,000

Table 2. Gross Alpha Analyses [1].

Tank Name	Primary Result	Duplicate Result	Mean	Standard Units	QC Flags [2]	Sample Location	Portion
241-U-201	<0.00109	<8.38E-04	<9.64E-04	µCi/g	QC: c	Core 70: 2; Riser 2	Segment Lower Half; Total.
241-U-201	<0.00308	<0.00379	<0.00343	µCi/g	QC: a, c	Core 70: 1; Riser 2	Segment Upper Half; Total.
241-U-201	<0.00311	<0.00311	<0.00311	µCi/g	QC: a	Core 70: 1; Riser 2	Segment Lower Half; Total.
241-U-201	<7.39E-04	<9.00E-04	<8.20E-04	µCi/g	QC: a, c	Core 73: 2; Riser 6	Segment; Total.
241-U-201	<8.39E-04	<0.00116	<10.00E-04	µCi/g	QC: c	Core 70: 2; Riser 2	Segment Upper Half; Total.
241-U-201	<9.72E-04	<6.78E-04	<8.25E-04	µCi/g	QC: a, c	Core 73: 1; Riser 6	Segment; Total.
241-U-202	<0.00118	<0.00116	<0.00117	µCi/g	QC: a, c	Core 78: 2; Riser 2	Segment Lower Half; Total.
241-U-202	<0.00118	<0.0012	<0.00119	µCi/g	QC: a, c	Core 75: 2; Riser 2	Segment Lower Half; Total.
241-U-202	<0.00532	<0.00471	<0.00502	µCi/g	QC: a, c	Core 78: 2; Riser 2	Segment Upper Half; Total.
241-U-202	<7.40E-04	<0.00122	<9.80E-04	µCi/g	QC: a, c	Core 75: 2; Riser 2	Segment Upper Half; Total.
241-U-202	0.00119	0.00107	0.00113	µCi/g	QC: a, c, e	Core 75: 1; Riser 2	Segment; Total.
241-U-202	0.00125	<0.00137	<0.00131	µCi/g	QC: a, c	Core 78: 1; Riser 2	Segment Upper Half; Total.
241-U-202	0.00152	<0.0011	<0.00131	µCi/g	QC: a, c	Core 78: 1; Riser 2	Segment Lower Half; Total.
241-U-203	<0.00121	<0.00131	<0.00126	µCi/g	QC: c	Core 80: 1; Riser 6	Segment Upper Half; Total.
241-U-203	<0.00151	<0.00147	<0.00149	µCi/g	QC: c	Core 79: 1; Riser 2	Segment Lower Half; Total.
241-U-203	9.75E-04	<0.00115	<0.00106	µCi/g	QC: c	Core 80: 1; Riser 6	Segment Lower Half; Total.
241-U-204	0.0967	<0.0623	<0.0795	µCi/g		Core 81: 1; Riser 2	Segment Upper Half; Total.
241-U-204	<0.0436	<0.044	<0.0438	µCi/g		Core 82: 1; Riser 6	Segment Upper Half; Total.

Notes:

[1] Analyses were obtained from the Tank Waste Information System on October 21, 2002. See http://twins.pnl.gov:8001/twins.htm

[2] Quality Control (QC) Flags Explanation:

a -- indicates that the standard recovery was below the QC range.
b -- indicates that the standard recovery was above the QC range.
c -- indicates that the spike recovery was below the QC range.
d -- indicates that the spike recovery was above the QC range.
e -- indicates that the RPD was greater than the QC limit range.
f -- indicates that there was blank contamination.
g -- indicates that this is a tentatively identified compound.
h -- indicates that the serial dilution exceeds the acceptance limit.
i -- indicates that the matrix spike failed and the serial dilution passed.

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

There were numerous spent nuclear fuel reprocessing, research and development, and waste management activities conducted at the Hanford Site starting in 1944. These spent nuclear fuel reprocessing, research and development, and waste management activities conducted in the processing plants are discussed in 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.*

It has been established in Section 2.0 that only coating removal waste from the REDOX plant was transferred into tanks 241-U-201 through 241-U-203. Tank 241-U-204 received a diversion box flush consisting of second decontamination cycle waste from the 221-T Bismuth Phosphate plant and coating removal waste from the REDOX plant. Therefore, the following sections provide a discussion of the origin of only these specific waste types.

3.1 BISMUTH PHOSPHATE PROCESS – B- AND T-PLANTS

B- and T-Plants were constructed in 1944 – 1945 to separate plutonium from spent nuclear fuel using the bismuth phosphate process. Figure 1 depicts a simplified process flow diagram for the bismuth phosphate process. In the bismuth phosphate process, the aluminum coating of spent nuclear fuel elements were dissolved in boiling sodium nitrate solution, to which sodium hydroxide was added (HW-10475-C, chapter IV, page 403). The coating removal waste (designated as CW) was transferred to various single-shell underground storage tanks.

The fuel element uranium cores 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 the precipitation of uranium as a phosphate in the subsequent plutonium extraction step (HW-10475-C, chapter IV, 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, chapter V, page 503). The plutonium and bismuth phosphate carrier precipitate was centrifuged and washed with water to separate the acidic supernatant from the precipitate. The acidic solution remaining after the plutonium precipitation contained 99 percent of the uranium, about 90 percent of the short-lived fission products (e.g., niobium-95, yttrium-91, strontium-89, and cerium-141), and about 1 percent of the plutonium (HW-10475-C, chapter V, page 503). This separation process also removed and reduced the gamma radiation activity level in the plutonium precipitate by a factor of 10. Uranium along with strontium, cesium, and technetium readily partition to the metal waste. However, zirconium is phosphate 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 single-shell tanks as metal waste (designated as MW). Recent

laboratory testing of the bismuth phosphate flowsheet confirm this partitioning of radionuclides (internal letter 7G300-02-NWK-024, "Bismuth Phosphate Process Radionuclide Partition Factors for the Hanford Defined Waste Model").

The plutonium precipitate and bismuth phosphate carrier were 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 transferred to single-shell tanks. The 1C waste contained approximately 10 percent of the 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).

A second decontamination cycle was conducted to reduced 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, chapter VI, 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 single-shell tanks. The 2C waste contained less than 0.1 percent of the fission products and about 0.4 percent of the plutonium present in the original fuel charged to the plant (HW-23043, pp 26 and 28). 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.

Table 3 provides the flowsheet estimated compositions of the neutralized coating removal waste (CW), metal waste (MW), first decontamination cycle (1C), and second decontamination cycle (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 single-shell tanks are provided in Tables 4 and 5. The CW was combined with the 1C waste in the same single-shell tanks. Note that the coating waste batch size shown in Table 3 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 as evident by the gamma emitting radionuclide analyses reported in Table 4. Analyses of the 2C supernatant stored in tank 241-T-112 conducted on August 6, 1952 and September 24, 1952 indicate that the total beta emitters was comprised of 35 to 50 percent ruthenium, 35 to 50 percent cesium, 4 to 8 percent cerium, yttrium, and other rare earths, and 6 to 11 percent undetermined (HW-27035, page 8).

Figure 1. Bismuth Phosphate Process

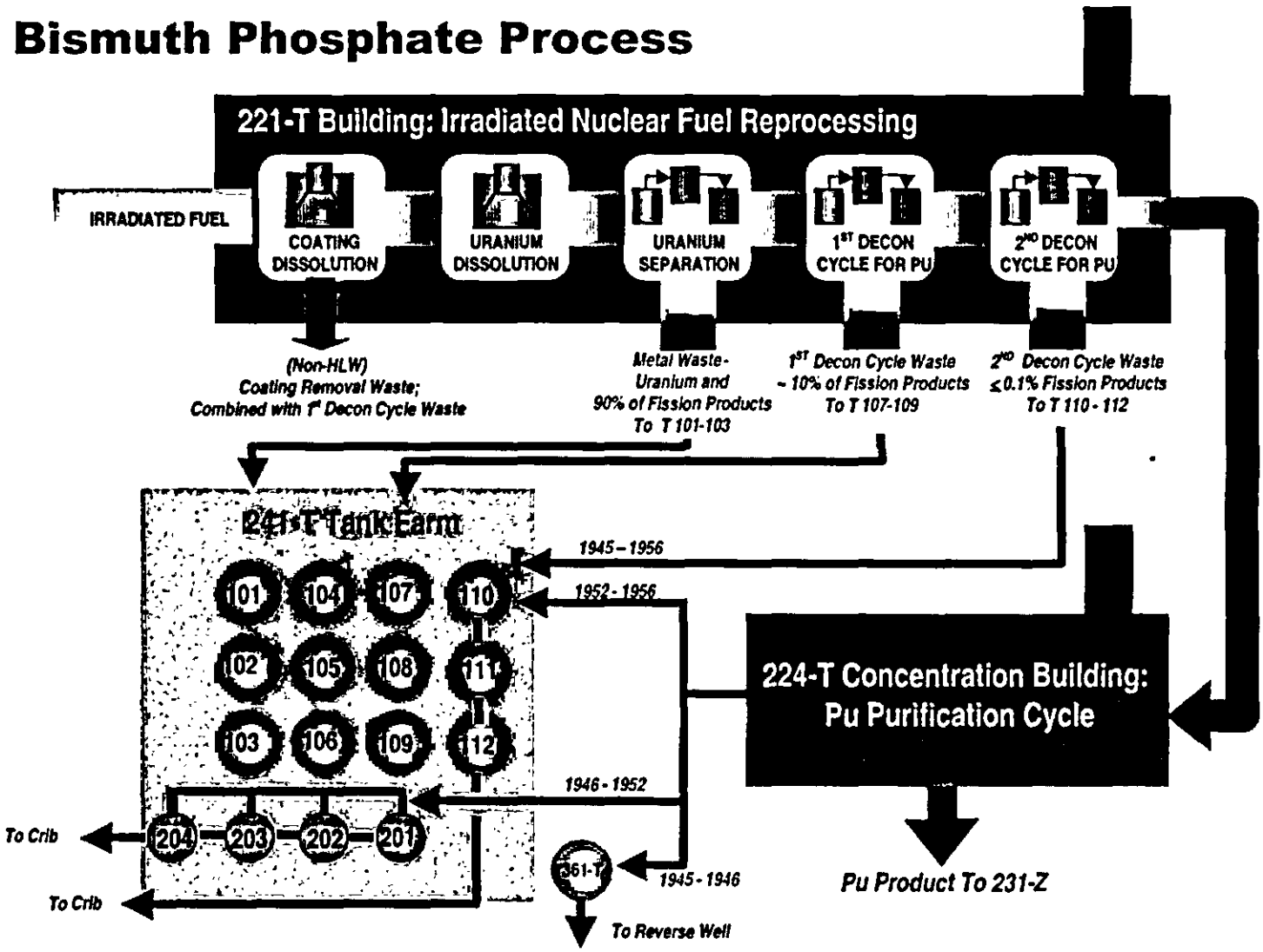

Table 3. Estimated Composition of Bismuth Phosphate Plant Wastes from October 1, 1951 Flowsheet [1].

Analyte [2]	Coating Removal Waste	Metal Waste	First Decontamination Cycle (1C) Waste	Second Decontamination Cycle (2C) Waste
Pu	3.3E-04	2.0E-04	6.0E-07 [4]	1.6E-07 [5]
U	0.15		0.235 [4]	Not reported
Gamma	6.6E+04	1.3E+07	2.3E+06 [4]	1.13E+04 [5]
$NaAlO_2$	95.1			
NaOH	43.6			
$NaNO_3$	61.8			
$NaNO_2$	56.0			
$NaSiO_3$	4.3			
UHN [3]		132		
NO_3		9.7	93.1	61.3
SO_4		24.4	4.73	3.61
PO_4		25.2	26.2	23.0
Na		83.2	47.3	36.7
Bi			2.59	1.31
Ce			0.030	
Zr			0.030	
Fe			1.37	1.82
Cr			0.16	0.06
NH_4			1.98	1.71
SiF_6			4.35	3.67
Volume per Batch (gallons)	795	2,380	2,040	2,090

Notes:

[1] See HW-23043

[2] Analyses are reported in grams per liter, except for gamma activity, which is counts/minute/ml.

[3] HW-23043 page 31 notes that uranium is not actually present in this form, but is probably as $NaUO_2PO_4$ and $Na_4(UO_2)_2CO_3$.

[4] Pu and Gamma concentrations were calculated based on the sum of tanks 13-4 and 14-3 compositions.

[5] Pu and Gamma concentrations were calculated based on the sum of tanks 18-4 and 19-3 compositions.

Table 4. 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	70	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

Notes:

[1] See HW-10728, "Process Waste Data – 200 Areas."

[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 241-B-112 seems to be in error and lacking an exponent in HW-10728.

Table 5. Analyses of First Decontamination Cycle / Coating Waste Supernatant [1].

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
B-107	8-1945	1.7E-02	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						
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 [2]	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

Notes:

[1] See HW-20195, *Radioactive Content of Stored Bismuth Phosphate First Cycle Waste Supernatants.*

[2] Tank 241-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.

3.2 REDOX CONTINUOUS SOLVENT EXTRACTION PROCESSES

The REDOX plant (202-S building) was operated from 1952 through 1966 to reprocess spent nuclear fuels. The bulk of the nuclear fuel elements reprocessed at the REDOX plant were coated with aluminum, which is sometimes referred to as cladding. Some zirconium-clad fuel was also processed in the REDOX plants in 1963 through 1966. A summary of processing activities at the REDOX plant is provided in RHO-CD-505-RD, *Synopsis of REDOX Plant Operations.*

In the REDOX plant, aluminum coated uranium fuel elements that had been irradiated at the Hanford Site reactors was reprocessed to recover uranium and plutonium (HW-38684). The first step in the reprocessing at the REDOX facility was the dissolution of the aluminum coating from the spent nuclear fuel elements. The fuel elements were placed in a dissolver vessel and sodium hydroxide and sodium nitrate solutions were added. The solution was heated to boiling to promote dissolution of the aluminum coating from the uranium fuel elements. The coating removal waste (designated as CW) from the aluminum-clad fuel was inherently alkaline and did not require neutralization before transfer to underground single-shell tanks. The coating waste solution contained approximately 0.03 percent of the uranium and 0.04 percent of the plutonium originally in the spent nuclear fuel element (HW-38684, page 9). Table 6 provides analytical results for a sample of the REDOX coating removal waste, which was reported in March 1953 (DDTS-Generated-607, 1953, "Proposed Cribbing of REDOX Coating Removal Solution").

Table 6. Analysis of REDOX Coating Removal Waste.

Analyte	Concentration
Uranium	0.16 g/L
Plutonium	150 µg/L
Beta emitters	2500 µCi/L
NaOH	3.5%
$NaNO_3$	4.9%
$NaAlO_2$	8.2%
Na_2SiO_3	0.1%
$NaNO_2$	5.2%
H_2O	78.1%
pH	12 to 13

Next, the uranium metal was dissolved in nitric acid. The dissolved uranium metal solution contained approximately 99.97 percent of the uranium and 99.96 percent of the plutonium originally in the spent nuclear fuel element. The uranium metal solution was reacted with an oxidizing chemical (dichromate solution) and then processed through a series of solvent extraction cycles using methyl isobutyl ketone solvent to separate uranium and plutonium from fission products. The fission products and impurities separated during the uranium and

plutonium solvent extraction process were neutralized and transferred to single-shell underground storage tanks, forming supernatant and sludges within the tanks. The plutonium solutions generated at the REDOX plant were transferred to the 234-5Z building (Z-Plant) for further processing. Uranium solutions were transferred to 224-U building (UO_3 Plant) for conversion to an oxide, which was transferred to offsite facilities for re-use in the fabrication of nuclear fuel.

4.0 TRANSURANIC ANALYSES OF WASTE IN U-200 SERIES TANKS

Analyses of specific alpha-emitting transuranic isotopes in the wastes in tanks 241-U-201 through 241-U-204 have not been conducted. Gross alpha analyses have been conducted for these wastes and are summarized in Table 7.

The gross alpha analysis tends to over estimate the sum of alpha-emitting transuranic isotopes, since uranium-238 is included in the gross alpha analysis. The mean and uncertainty estimates for the gross alpha analyses for these wastes were recently evaluated (RPP-10983). The mean gross alpha analyses and upper 95 percent confidence limit for the wastes stored in tanks 241-U-201 through 241-U-203 indicate that the concentration of alpha-emitting transuranic isotopes with half-life greater than 20 years is less than 10 ηCi/g in these wastes. The mean gross alpha analyses and upper 95 percent confidence limit for the waste stored in tank 241-U-204 are less than 62 ηCi/g and 130 ηCi/g. Due to the large uncertainty with the gross alpha analyses for the waste stored in tank 241-U-204, additional analysis of this waste is required to quantify the concentrations of alpha-emitting transuranic isotopes with half-life greater than 20 years.

Table 7. Gross Alpha Analyses for U-200 Series Tanks

Tank	Mean Gross Alpha Concentration (ηCi/gram)	Upper 95% Confidence Limit (ηCi/gram)
241-U-201	< 2	5
241-U-202	< 2	8
241-U-203	< 1	8
241-U-204	< 62	130

5.0 SUMMARY

Single-shell tanks 241-U-201 through 241-U-203 were filled with REDOX coating removal waste in February and March 1956. Single-shell tank 241-U-204 received approximately 35,000 gallons of waste in January 1954 from a flush of a diversion box and this waste was characterized as second decontamination cycle waste from the 221-T Bismuth Phosphate Plant. Tank 241-U-204 was then filled with approximately 18,000-gallons of REDOX coating removal waste in May 1956. The supernatant in all four of these tanks was removed in 1977 and processed through an evaporator, leaving a heel of approximately 4,000 to 5,000-gallons in total of supernatant and sludge in each tank.

The mean gross alpha analyses and upper 95 percent confidence limit for the wastes stored in tanks 241-U-201 through 241-U-203 indicate that the concentration of alpha-emitting transuranic isotopes with half-life greater than 20 years is less than 10 ηCi/g in these wastes. The mean gross alpha analyses and upper 95 percent confidence limit for the waste stored in tank 241-U-204 are less than 62 ηCi/g and 130 ηCi/g.

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