INTEGRATED RADWASTE TREATMENT SYSTEM FINAL REPORT

Topical Report

By
M. N. Baker
H. M. Houston

October, 1997

Work Performed Under Contract No. DE-AC24-81NE44139

Prepared by
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DOE/NE/44139-83
Distribution Category UC-510

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ACKNOWLEDGMENTS

Pretreatment of liquid high-level radioactive waste at the West Valley Demonstration Project represents a very successful operational effort that depended on the skill, competence, and ability of many people. Being able to accurately summarize and depict the events that led to this success has been possible thanks to the graphic skill of Bob Bartkowski, and thoughtful technical reviews done by Bob Fussner, Sam Kelly, Dan Meess, and Carl Ross. Their help has been greatly appreciated in preparing this report.

ABSTRACT

In May 1988, the West Valley Demonstration Project (WVDP) began pretreating liquid high-level radioactive waste (HLW). This HLW was produced during spent nuclear fuel reprocessing operations that took place at the Western New York Nuclear Service Center from 1966 to 1972.

Original reprocessing operations used plutonium/uranium extraction (PUREX) and thorium extraction (THOREX) processes to recover usable isotopes from spent nuclear fuel. The PUREX process produced a nitric acid-based waste stream, which was neutralized by adding sodium hydroxide to it. About two million liters of alkaline liquid HLW produced from PUREX neutralization were stored in an underground carbon steel tank identified as Tank 8D-2. The THOREX process, which was used to reprocess one core of mixed uranium-thorium fuel, resulted in about 31,000 liters of acidic waste. This acidic HLW was stored in an underground stainless steel tank identified as Tank 8D-4.

Pretreatment of the HLW was carried out using the Integrated Radwaste Treatment System (IRTS), from May 1988 until May 1995. This system was designed to decontaminate the liquid HLW, remove salts from it, and encapsulate the resulting waste into a cement waste form that achieved U.S. Nuclear Regulatory Commission (NRC) criteria for low-level waste (LLW) storage and disposal.

A thorough discussion of IRTS operations, including all systems, subsystems, and components, is presented in U.S. Department of Energy (DOE) Topical Report DOE/NE/44139-68, "Integrated Radwaste Treatment System Lessons Learned from 2½ Years of Operation." This document also presents a detailed discussion of lessons learned during the first 2½ years of IRTS operation.

This report provides a general discussion of all phases of IRTS operation, and presents additional lessons learned during seven years of IRTS operation.

EXECUTIVE SUMMARY

In 1980, the United States Congress authorized the U.S. Department of Energy (DOE) to solidify liquid high-level radioactive waste (HLW) at West Valley, New York, into a form suitable for transportation and disposal at a federal repository. Vitrification was the technology selected to solidify the HLW. To efficiently vitrify the HLW, it was necessary to reduce the quantity of wastes that needed to be vitrified. A pretreatment system that uses ion exchange to separate cesium-137 (Cs-137), and other isotopes of concern, from sulfates and nitrates in the waste stream was developed for this purpose. To the maximum extent possible, existing facilities were used to design and construct this pretreatment system.

Several different HLW pretreatment campaigns were conducted to prepare the liquid HLW for vitrification. The primary purpose of these campaigns was to concentrate the HLW that would be processed in the Vitrification Facility, and ultimately, made into borosilicate waste glass.

The bulk of the HLW, approximately two million liters, was formed by the addition of sodium hydroxide to a nitric acid-based waste stream. This waste stream was produced by using the plutonium/uranium extraction (PUREX) process, and stored in an underground carbon steel tank designated as Tank 8D-2. The remainder of the HLW, approximately 31,000 liters, was produced by using the thorium extraction (THOREX) process. This THOREX waste was stored in an underground stainless steel tank designated as Tank 8D-4.

Neutralization caused the PUREX waste in Tank 8D-2 to separate into a liquid salt solution, or supernatant, above semi-solid sludge. The first phase of HLW pretreatment decanted the supernatant and removed Cs-137 from it by processing the supernatant through a series of ion-exchange columns filled with zeolite. The resulting decontaminated liquid was then volume reduced through evaporation, and encapsulated into a cement waste form that achieved NRC criteria for stabilized low-level waste (LLW) storage and disposal.

The second pretreatment campaign eventually involved a series of steps to precipitate plutonium (Pu) and uranium (U) compounds from the liquid and into the sludge, and prevent HLW carry-through to the cement waste form. Sludge was "washed" with a solution of sodium hydroxide and water to increase alkalinity from a pH of 10 to a pH of 12.5. Increasing alkalinity precipitated Pu and U compounds into the sludge. Five 150 horsepower mobilization pumps were used to thoroughly mix tank contents, including interstitial liquids trapped in the sludge. Decontaminated sludge wash liquids were volume reduced through evaporation, and encapsulated into an approved cement waste form. In total, two complete sludge wash operations were performed.

The third and final HLW pretreatment campaign involved transferring THOREX waste from Tank 8D-4 to Tank 8D-2, washing the blended waste, and processing the resulting decontaminated liquids into an approved cement waste form.

1.0 INTRODUCTION

The Integrated Radwaste Treatment System (IRTS) refers to a combined series of pretreatment systems used to decontaminate liquid high-level radioactive waste (HLW), and reduce salts in the HLW. Involving three distinct processing systems, the Supernatant Treatment System (STS), Liquid Waste Treatment System (LWTS), and Cement Solidification System (CSS), the IRTS operated from May 1988 until May 1995. A detailed description of these systems, and the first 2½ years of IRTS operation is given in "Integrated Radwaste Treatment System Lessons Learned from 2½ Years of Operation" [DOE/NE/44139-68].[1] This report provides a general discussion of the IRTS, and presents additional lessons learned during seven years of IRTS operation.

2.0 INTEGRATED RADWASTE TREATMENT SYSTEM OPERATION

2.1 Pretreatment Requirements

The primary purpose of HLW pretreatment was to improve vitrification efficiency by reducing the volume of HLW that would be processed in the Vitrification Facility. This was done by decanting supernatant, removing Cs-137 and other radioisotopes from it through an ion-exchange process, evaporating the resulting decontaminated liquid, and encapsulating the remaining concentrates into a cement waste form that achieved NRC criteria for stabilized low-level waste (LLW) storage and disposal.

2.2 Processing Operations

High-level waste (HLW) pretreatment was accomplished by using an integrated series of systems, subsystems, and components known collectively as the Integrated Radwaste Treatment System (IRTS). The main systems used were the Supernatant Treatment System (STS), Liquid Waste Treatment System (LWTS), and Cement Solidification System (CSS). The STS was designed to decontaminate liquid HLW. Decontaminated liquid was volume-reduced in the LWTS, and resulting concentrates were made into cement drums in the CSS.

Each of the main IRTS systems was used to conduct a series of campaigns involving several distinct tasks: supernatant processing, PUREX sludge washing, and PUREX/THOREX waste blending and washing. These campaigns were run from May 1988 until May 1995.

During supernatant processing, supernatant from Tank 8D-2 was decontaminated by processing it through four zeolite filled ion-exchange columns mounted in Tank 8D-1, which had been designed as the spare for Tank 8D-2 during original reprocessing operations. These ion-exchange columns were arranged in series flow, and designed to remove more than 99.9% of the Cs-137 in the supernatant. After supernatant was decontaminated in the STS, it was volume-reduced through evaporation in the LWTS. Concentrates resulting from the evaporation process were then encapsulated into Portland cement in the CSS and poured into 71-gallon square drums.

The IRTS was used for supernatant processing from May 1988 until November 1990. Over this two-year period, 1,703,385 liters (450,000 gallons) of liquid HLW were decontaminated and evaporated, resulting in concentrates that were encapsulated into 10,393 square drums. About 5.20 million curies of Cs-137 were removed and retained on zeolite for HLW processing.

PUREX sludge washing was done over the course of two separate campaigns. The first campaign was conducted from October 1991 until January 1992. During this period, over 1,550,000 liters (409,479 gallons) of sludge wash solution, containing about 910,000 curies of Cs-137 were processed through the STS, LWTS, and CSS, resulting in 7,279 square drums.

The second sludge wash campaign ran from in May 1994 until June 1994. This campaign was conducted to further reduce the amount of sulfates in the sludge. Approximately 1,350,000 liters (356,643 gallons) of sludge wash solution, containing about 126,000 curies of Cs-137 were processed during this campaign, resulting in 754 square drums.

After PUREX sludge washing was completed, final preparations were made to finish installation of a transfer system linking all HLW tanks with the Vitrification Facility. When the transfer system was in place, acidic THOREX waste in Tank 8D-4 was transferred to Tank 8D-2 while being neutralized. A complete description of THOREX processing operations is given in, "THOREX Processing and Zeolite Transfer for High-Level Waste Stream Processing Blending," [DOE/NE/44139-82]. [2]

THOREX transfer and neutralization operations got underway in January 1995. Once THOREX was transferred and neutralized, it was blended with the PUREX sludge, washed and processed through the IRTS. Approximately 1,190,000 liters (314,374 gallons) of PUREX/THOREX wash solution, containing about 300,000 curies of Cs-137 were processed from January 1995 until May 1995, resulting in 1,451 square drums. A total of 19,877 of square drums were placed in the Drum Cell during the course of IRTS operations.

3.0 IRTS GENERAL LESSONS LEARNED

3.1 IRTS Process Knowledge and Improvements

For the most part, the systems that made up the Integrated Radwaste Treatment System (IRTS), STS, LWTS, and CSS operated as designed. During the earliest phases of supernatant processing, the on-shift engineering group began to focus on statistical process control and product quality. Consequently, the group was designated as Process Control Engineering to reflect its involvement in processing rate improvement.

One area of improvement involved STS dilution. At the beginning of IRTS operations, raw supernatant was withdrawn from Tank 8D-2, and fed undiluted to the ion exchange columns. This maximized the throughput. Beginning with the seventh campaign, which was run in January 1989, supernatant was diluted with two parts of demineralized water to each part of raw supernatant. In later pretreatment campaigns, raw supernatant was diluted with up to three parts demineralized water. Diluting feed to the ion-exchange columns from approximately 23 wt. % total dissolved solids (TDS) to 11 wt.% TDS improved Cs-137 capture on the zeolite by effectively increasing the contact time between the waste liquid and the zeolite. Use of dilution water required evaporation downstream in the LWTS, but improved net throughput and reduced zeolite usage. Using less zeolite ultimately reduced the number of glass logs that would need to be produced in the Vitrification Facility. A summary of campaign cycles is given in Table 3.1.

As process knowledge developed, it became evident that radiation monitors installed on the effluent piping of the ion-exchange columns were not reliable for on-line process control. [See Figure 3.1] High system decontamination factors (DFs), low monitor readings, and awkward conversion from counts per minute on the monitors to microcuries per milliliter in the liquid affected data accuracy. Batch sampling and on-line sampling of the tanks and ion-exchange columns proved to be a better source of primary data for process control.

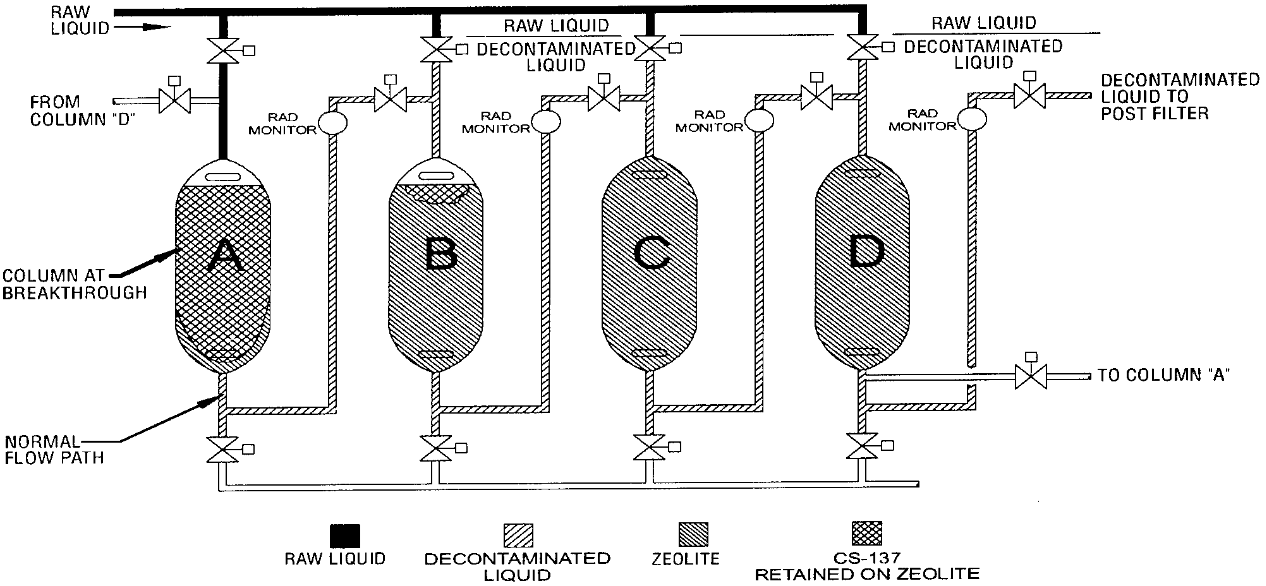

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Figure 3.1 STS Ion-Exchange Columns

There was no particular benefit in filling the ion-exchange columns to the point of "breakthrough" because Cs-137 that was not being retained by the lead column was being captured by the second column. [See Figure 3.1] This meant that a high-volume campaign would be followed by a reduced-volume campaign, that a low-volume campaign would be followed by a high-volume campaign, and so on.

Daily sampling results were received in time to be used for processing decisions. Results showed that the incoming concentration of Cs-137 divided by the outgoing concentration, or decontamination factor (DF), averaged more than 50 times the design minimum of 1,000 DF. As a result, low-dose drums were suitable for shield positions in the Drum Cell, (i.e., the top layer). Since the number of low-dose drums could be predicted, these drums were set aside for shield positions as they were produced. This ultimately resulted in increased Drum Cell capacity because no non-radioactive drums were needed for shield positions.

Table 3.1. Summary of Campaign Cycles

Campaign	Column Sequence	Notes	Gallons	Cs-137 DF
1: 5/88 - 6/88	B-C-D-A	No dilution	26,000	> 1,500
2: 6/88 - 7/88	C-D-A-B	No dilution	15,800	180,000
3: 7/88 - 8/88	D-A-B-C	No dilution Sample jumper not fully closed "D" dump valve failure.	26,400	39,500
4: 8/88 - 9/88	A-B-C	No dilution	21,000	4,000
5: CANCELED	CANCELED	No campaign run report		
6: 11/88 - 12/88	A-B-C-D	No dilution	17,800	11,100
7: 1/89 - 2/89	B-C-D-A	2:1 dilution	35,400	95,500
8: 3/89 - 4/89	C-D-A-B	1.4:1 dilution	76,000	37,500
9: 4/89 - 5/89	D-A-B-C	1.4:1 dilution	53,000	57,200
10: 5/89 - 6/89	A-B-C	1.4:1 dilution	24,100	23,400
11: 7/89 - 8/89	B-C-A	1.4:1 dilution	35,100	26,800
12: 8/89 - 10/89	C-D-A-B	1.4:1 dilution	33,400	67,400
13: 10/89 - 11/89	D-A-B-C	1.4:1 dilution "B" dump valve failure	28,300	24,200
14: 11/89 - 12/89	B-C-A	1.4:1 dilution	33,900	30,400
15: 1/89 - 2/16	D-A-C	2.3:1 dilution	33,300	27,900
16: 3/90 - 4/90	C-A-B	3.6:1 dilution	46,600	34,400
17: 4/30 - 6/90	A-B-C	3.6:1 dilution	45,200	26,900
18: 6/90 - 7/90	B-C-D-A	3:1 dilution; Heel left in D	39,800	33,600
19: 8/90 - 9/90	C-A-B	3:1 dilution	49,200	95,000
20: 9/90 - 11/90	D-A-B-C	3:1 dilution	50,700	62,900
21: 10/90 -1/91	A-B-C-D	3:1 dilution	6,300	Not Reported
22: 4/92 - 9/92	B-C-A	Sludge Wash I starts 1:1 dilution	44,100	38,800

Table 1. Summary of Campaign Cycles (continued)

Campaign	Column Sequence	Notes	Gallons	Cs-137 DF
23: 9/92 - 3/93	C-A-B	1:1 dilution; Heel	53,500	3,100
24: 3/93 - 6/93	A-B-C	1:1 dilution	51,900	49,200
25: 6/93 - 8/93	B-C-A	1:2 dilution; FV-102A leak	49,000	7,000
26: 8/93 -2/94	C-A-B	2:1 dilution	57,600	23,200
27: 2/94 - 3/94	A-B-C	1:2 dilution	50,200	38,000
28: 3/94 - 4/94	B-C-A	1:2 dilution; Low DFs	66,600	3,600
29: 4/94 - 7/94	C-A-B	1:2 dilution	208,800	11,000
30: 8/94 - 2/95	A-B-C	No dilution	240,400	8,500
31: 3/95 - 5/95	B-C-A	No dilution	240,900	40,400

4.0 STS LESSONS LEARNED

4.1 Decontamination Factors

Decontamination factors (DFs) for the IE-96® zeolite used in supernatant processing averaged 50,000 DF, compared to the design minimum of 1,000 DF. This resulted in reduced dose rates downstream in LWTS and CSS.

4.2 STS Zeolite Heels

Normal STS column rotation took the lead column, with capture sites that had been loaded with Cs-137, and dumped its spent zeolite back into Tank 8D-1. After filling this column with fresh zeolite, STS feed valves were reconfigured to place the cleanest column into the final, or "polishing" position. Occasionally, a "heel" of contaminated zeolite would remain, adhering to the sides of the column. This caused radioactivity to appear in the downstream product. If radioactivity was too high, the column would be taken offline, or deliberately fed with raw supernatant to load the capture sites. The column would then be dumped again, and refilled. Using this technique, zeolite was neither wasted nor dumped without first being loaded with Cs-137. This could have posed a problem because zeolite usage had been deliberately minimized to reduce the number of HLW canisters produced during vitrification.

Another processing phenomenon involved the STS piping arrangement, which required crossflow valves to close with liquid feed on one side, and decontaminated liquid on the other side. [See Figure 4.2]

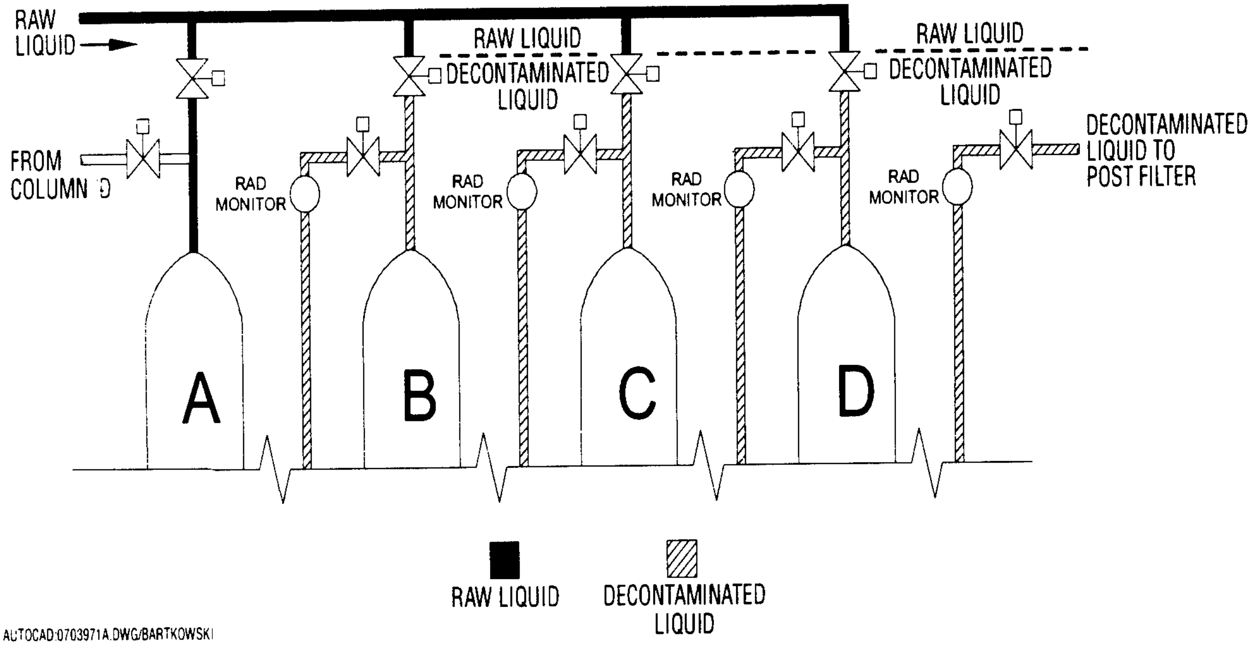

Figure 4.2 STS Piping and Crossflow Valves

On a number of occasions, a leak in one of the closed valves (i.e., ball values), caused untreated liquid to leak through to the "clean," or decontaminated side. The high decontamination factors being achieved significantly reduced the amount of radioactivity in downstream liquid. This greatly improved the ability to detect leaks because the presence of untreated liquid could be easily detected by downstream radiation monitors. In most cases, the process could be stopped and the valve cycled to reseat the valve ball. Some of the troublesome valves were eventually replaced. A full description of valve replacement is discussed in DOE/NE/44139-68.[1]

4.3 Supernatant Removal Pump 50-G-001 Orientation

During Campaign 26, it became apparent that the floating suction of supernatant removal pump 50-G-001 was interfering with the inside wall of Tank 8D-2. Attempts were made to view the floating suction with the in-tank video camera. However, it was too far away from the suction head, and the in-tank camera lighting system did not provide enough light for adequate viewing. In addition, atmospheric conditions in the tank vapor space caused poor visibility. Later, an improved camera and lighting system were installed and used to confirm this interference. The pump and its shield plug were reoriented by unbolting it from its mounting flange and rotating it. By using a containment tent for contamination control, and through the use of mockups and tooling, West Valley Nuclear Services Co. Inc. (WVNS) experience with contaminated work proved to be extremely valuable in this situation. With the pump's floating suction free to move, the liquid level in 8D-2 was reduced to its lowest practical level. This resulted in the elimination of an entire sludge wash, and saved several months on the project schedule.

4.4 Sludge Wash Processing and pH

The decontaminated supernatant was processed at a pH of approximately 10. The sludge washing process required suppression of uranium (U) and plutonium (Pu) by increasing the pH to approximately 12.5. This required the addition of a 50% solution of sodium hydroxide.

5.0 LWTS LESSONS LEARNED

5.1 Plutonium Solubility

Neutralization of nitric-acid based waste in Tank 8D-2 during original PUREX processing operations caused Pu to precipitate as a hydroxide and become part of the sludge layer in the tank. Excess caustic had been added to reduce tank corrosion as a process control. This limited the Pu concentration in the supernatant to minimum levels.

Research shows that the solubility of Pu varies with pH, especially in carbonate containing solutions. Subsequent to supernatant processing, and during sludge washing operations, dilution of "excess caustic" in Tank 8D-2 reduced solution pH, thereby increasing the solubility of Pu in the tank sludge wash solution by a factor of four.

5.2 LWTS Material Balance

Supernatant was sampled and characterized during pretreatment operations to track potential changes in the supernatant. Characterization began with supernatant sampling and analysis. Results were compared to analysis done before IRTS startup in May 1988. Comparison showed an increase in Pu concentration [7.52 E 02 Ci/mL at 39.5% TDS to 11.4 E 02 Ci/mL at 23% TDS]. The IRTS was put in standby and safety evaluations initiated.

Samples were then taken from various locations throughout the IRTS to pinpoint the exact area where fissile material was collecting. [See Figure 5.2] These results showed that material was collecting in the LWTS evaporator. Controlled full-scale testing was done to analyze for Pu before and after evaporation. Actual gross alpha in the concentrate was lower than the calculated expected value [1.35 E 01 Ci/mL compared to 7.83 E 01 Ci/mL]. Lab scale boil-down testing was done to confirm the working theory that fissile material was precipitating out of the evaporator solution. Results supported this speculation. Samples taken from four cement waste drums confirmed that fissile material had not been incorporated into the cement waste form.

After confirming that fissile material was collecting in the LWTS evaporator, it was necessary to determine how much Pu and U had precipitated out of the solution and into the evaporator. Calculating Pu was relatively straightforward. Gross alpha data was available for each batch of evaporator feed and concentrates processed through the LWTS. This data, coupled with the gross alpha-to-plutonium ratio, showed that 359 ± 100 grams of Pu had collected in the evaporator. Calculating U was more difficult because only one data point existed for uranium concentrations before and after evaporation. This data point, with some conservative assumptions, resulted in an estimate of 26 Kg total U collected in the evaporator.

Once concentrations of Pu and U had been calculated, planning began to recover these isotopes from the evaporator in the safest, most efficient way possible. Initial planning focused on gaining access to the evaporator internals. To do this, it was first necessary to remove the evaporator reboiler tube bundle. A radiological survey of the evaporator and surrounding area showed a general area dose rate of 50 to 100 mR/hr, with a one to 2 R/hr hot spot on the reboiler. These results, combined with the knowledge that removing the steam tube bundle would be a labor intensive effort, conflicted with the practice of As Low As Reasonably Achievable (ALARA) philosophy. Therefore, it was necessary to develop an alternative removal method.

Evaporation of two small samples from the evaporator feed tank (7 to 9 wt.% TDS), produced signs of solids, evidenced by cloudiness after being evaporated to 15 to 20 wt.% TDS. Following further evaporation to 40 wt.% TDS (i.e., evaporator target concentration), the liquid concentrate contained only 23 to 24 wt.% TDS of the initial Pu.

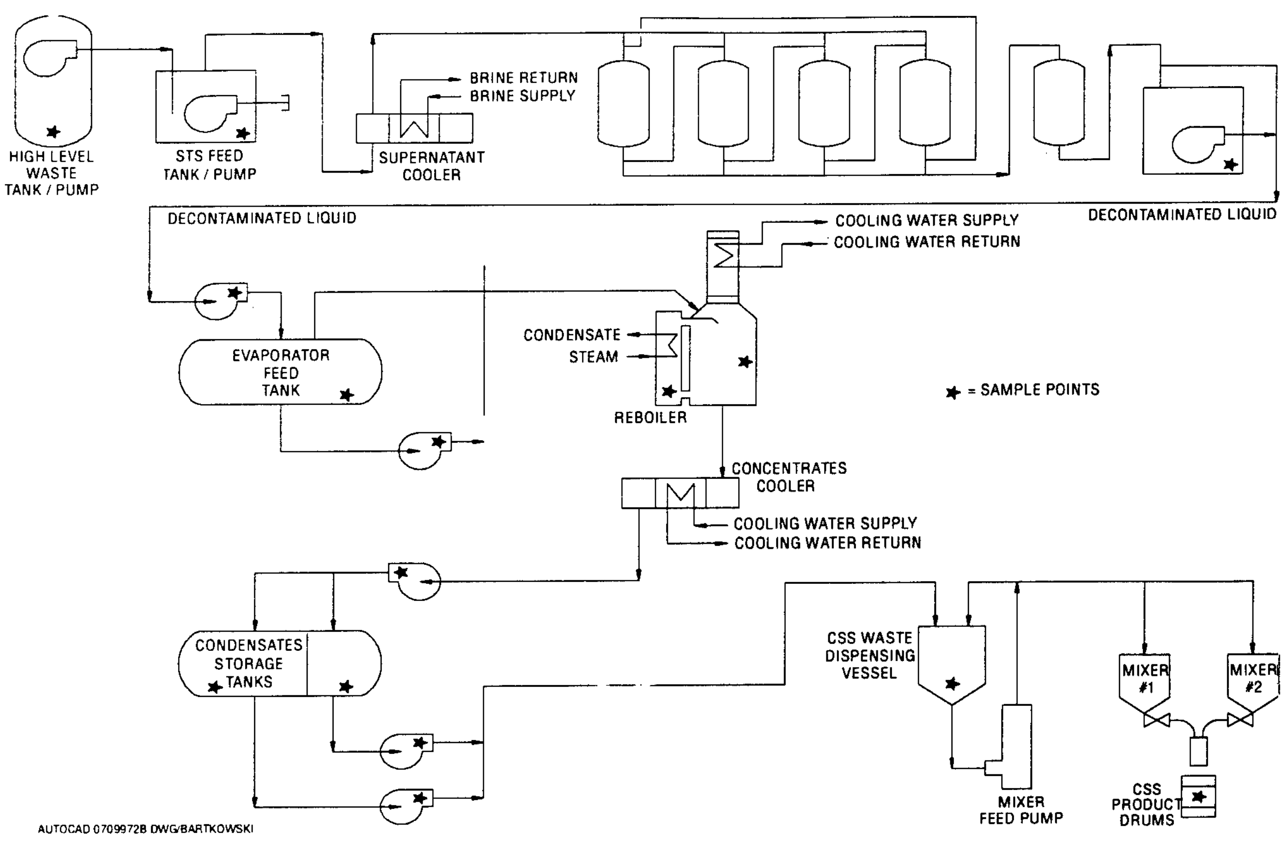

Figure 5.2 IRTS Sampling Points

Further analysis suggested that Pu and U could be removed chemically by using a nitric acid solution. Chemical removal would require no "hands on" work within the evaporator itself, and would make it quite easy to return the isotopes to Tank 8D-2. Outside experts were brought in to help determine the best parameters to use during acid cleaning. Solution with a concentration of one Molar nitric acid was selected.

The evaporator and its associated feed and concentrates tanks were given three separate acid washes with solutions containing one gram per liter of boron as boric acid and up to two Molar nitric acid. Analysis of these acid wash solutions indicated that 310.5 grams of Pu and 12.2 Kg of U were recovered from the evaporator subsystem. Independent analyses conducted by an outside laboratory confirmed these results within 1 percent agreement.

Independent reviewers established that the potential for a criticality event was not possible, that the probability of criticality was zero, and that therefore an unresolved safety question did not exist. Even though the potential for a criticality event was determined to not exist, a new processing safety requirement was put into place when processing resumed to ensure against the potential for accumulating fissile material in processing components. The sampling program was expanded for all samples to include U and Pu, and track the deposition of these isotopes by the Process Control Engineers. The total mass of fissile U and Pu was maintained at less than the agreed-to maximums of 1,500 grams and 450 grams for U and Pu, respectively. Both limits were well below the critical masses for these isotopes, allowing for geometry and other criticality factors.

5.3 Evaporator Gasket Replacement

Not long after the evaporator acid wash, steady increases in the sump level in Extraction Cell 3 (XC-3), the evaporator location, were observed. Observation by camera and personnel entry into the cell showed that there was a slow leak at the top of the evaporator vapor body/condenser joint.

The vessel gasket in place was made of VITON®, a material suitable for use both with acids and bases. However, VITON® was not suitable for use with acid solutions at elevated temperatures. Acid cleaning of the evaporator had been done at elevated temperatures. As a result, the VITON® gasket had deteriorated substantially. Since plans for potential future acid washes were already in place, the first task was to find a suitable gasket replacement material. AFLAS®, a tetrafluoroethylene-propylene copolymer, 1/8-inch thick, was selected because of its superior effectiveness in both acids and bases at high temperatures.

The next task involved developing a suitable method for replacing the gasket. Disassembling a contaminated pressure vessel like the evaporator, without the use of an in-cell crane or hoist, would be an extremely difficult task. Because of nearby piping and vessels, headroom to pull the entire tube bundle was simply unavailable.

Repair techniques that were developed involved welding three brackets, or gussets, onto the condenser and vapor body, and raising the condenser approximately ½", using small hydraulic jacks. Using this approach, the failed gasket could be removed and a new gasket inserted, which had a special cut joint design. The entire vessel joint design was reviewed before repair work began. Based on this review, 12 additional clamps were added between the flange bolts to increase compression on the new gasket.

A number of tools were developed specifically for gasket replacement. Thin sheets of stainless steel were inserted between the flange bolts. Wire hooks were inserted to keep the new gasket from slipping into the reboiler. These hooks could be removed before the flange was tightened.

All equipment was tested on a full size mock-up in the maintenance shop to streamline repair techniques, and gauge the time, and therefore amount of exposure, involved in doing the repair work. [See Figure 5.3]

Locations of the gussets were marked on the vapor body. Interferences were noted, and avoided. The gussets were then clamped into place and welded. A special weld sequence was followed to minimize forces on the vessel shell and flange.

Three gussets were welded on the condenser and vapor body. The condenser then was raised about ½", using the small hydraulic jacks. These jacks were equipped with load indicators. The load on each jack was monitored to guarantee that no unforseen stresses were encountered. A safety lock bolt was furnished for each gusset to mechanically lock the condenser into the raised position. This provided protection against settling if hydraulic pressure was to bleed off any of the jacks.

Figure 5.3a
Evaporator Mock-up

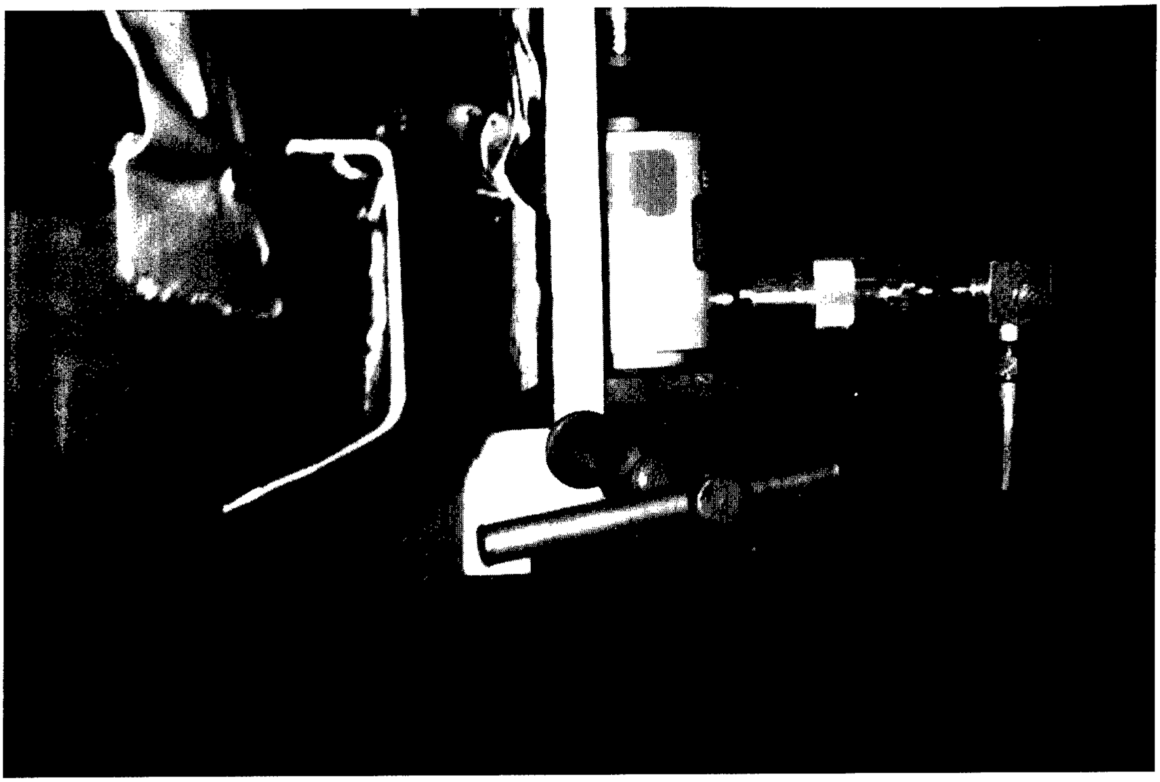

Figure 5.3b Evaporator Mock-up

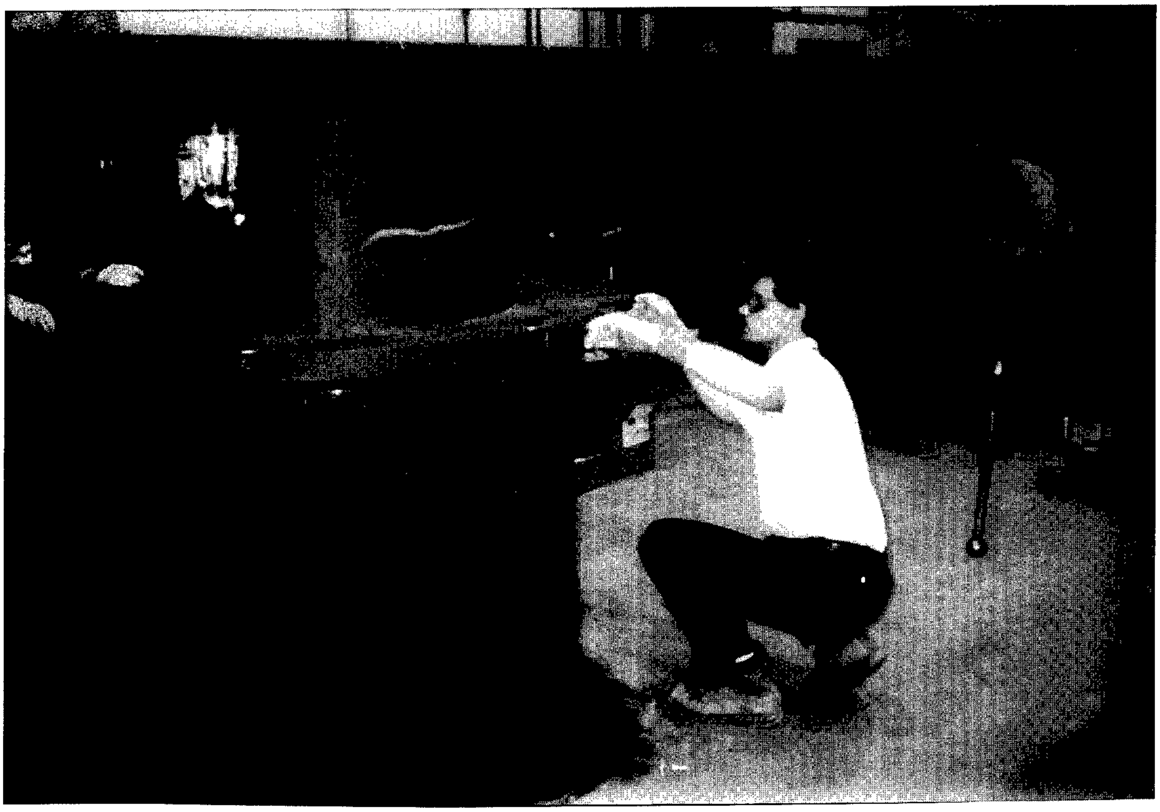

Figure 5.3c Evaporator Mock-up

6.0 CSS LESSONS LEARNED

6.1 Cement Recipe Work

Each waste stream, supernatant, PUREX sludge wash, and THOREX/PUREX sludge wash, required a different recipe, qualified to the U.S. Nuclear Regulatory Commission (NRC) branch technical position on low-level waste forms.

Getting the highest amount of dissolved solids into the waste liquid, and a satisfactory cement mixture (i.e., waste loading) was a consistent objective throughout recipe testing. The NRC branch position required demonstration of performance in the following areas: compressive strength; compressive strength after freeze-thaw cycles; compressive strength after immersion; leachability of radionuclides; leachability of hazardous constituents (heavy metals); biodegradation; and radioactive degradation.

6.2 Supernatant Recipe Work

A recipe qualification program was carried out in 1987-1988, in anticipation of supernatant processing and stabilization. This program tested simulated supernatant at the expected concentration of 39 wt.% TDS. Observations revealed a product with entrained air, low density, and lower-than-expected compressive strength.

Laboratory work confirmed that addition of a silicon-based anti-foamer could suppress foaming, thereby improving waste form durability and strength. An extensive laboratory study was done to optimize anti-foam concentration and mix time to produce a product with high density, low porosity, and high compressive strength. Results of all laboratory testing compared favorably with the previous recipe qualification work.

During testing, WVNS used 2" x 2" x 2" cement waste cubes made from non-radioactive waste simulant prepared according to ASTM method C-109. This standard method reduced the test period significantly. A sample of supernatant was removed from Tank 8D-2, and decontaminated using IE-96® zeolite, simulating the STS process. Solidification of part of this sample was unsuccessful. The resulting mixture underwent a phase separation, with the cement settling out of solution. Bleed water was observed as cement particles settled out of the slurry. It was obvious that set retardants were present in the actual supernatant that had not been present in the simulant.

The presence of organics, which are known set retardants, was suspected based on observation of the mix. Plant records of additions to Tank 8D-2 were reviewed to identify additives that could produce a set retarding effect. It was found that, during plant decontamination efforts in the 1970's, relatively large amounts of citric, oxalate, and tartrate (a total order of magnitude of 30,000 pounds) were flushed to Tank 8D-2 as sodium salts.

A comprehensive study was conducted to select set-enhancing additives that would overcome the set retarding effect produced by the presence of these organics. The objective was to produce a formulation that did not exhibit bleed water; had high compressive strength; and had the same leaching and other performance characteristics as the original, qualified Portland Type I cement/waste simulant recipe. A modified formulation was developed by extensive testing with simulated supernatant adjusted with organic acid salts, calcium nitrate, and sodium silicate additives. The target formulation was tested using the remaining sample of real supernatant. It produced a waste stream that showed improved processibility. Modifying the formulation with additives changed constituents of the original recipe by less than 2%.

Supplementary qualification testing confirmed that admixtures enhanced product performance in compressive strength, compressive strength following immersion, compressive strength following thermal cycling, and leach resistance.

6.3 Sludge Wash Recipe Work

Based on waste stream evaluation, a decision was made to use the existing recipe for PUREX sludge wash. This was possible because there was only a slight difference between the supernatant and PUREX sludge wash waste streams. However, the NRC Branch Technical Position had been revised and updated since the start of supernatant processing.[4] This revision incorporated experience gained during supernatant processing. Requirements for baseline compressive strengths had been increased, and the use of 2" x 2" x 2" cubes per ASTM method C-109 recommended.

The first phase of testing used simulated PUREX sludge wash liquid to run a statistically designed screening test (i.e., Plackett-Burman structure) with 13 variables to determine potential effects on gel time, compressive strength, and bleed water. Key variables included mix time, mixer speed, water to cement ratio, percentage of solids, additives, etc. A Box-Behnken test was then done to establish approximate, satisfactory ranges for the key variables, which became the basis for the Process Control Plan.

The decision was made to begin the decontamination phase of PUREX sludge washing in 8D-2 before recipe qualification work was completed because recipe qualification required nearly six months for processing, coring, compressive strength testing, immersion, and post-immersion testing. Consequently, the full-scale recipe qualification program started with actual decontaminated PUREX supernatant liquid adjusted to create a sludge wash surrogate liquid in the CSS.

A nominal recipe of 20 wt.% TDS was selected, with recipes at 24% and 27% TDS, to determine if waste loading could be increased later. To provide an operating margin for potential changes in plant conditions, the sulfate concentration of the decontaminated sludge wash liquid was increased by a nominal 25%. This was done by adding sodium sulfate to the actual decontaminated PUREX sludge wash liquid while it was in the CSS Waste Dispensing Vessel. Eleven test drums were processed, cured, and cored for the required testing. These cores provided excellent compressive strengths, thermal cycling resistance, and Toxicity Characteristic Leaching Procedure (TCLP) metals retention.

The cores, however, did not pass the 90-day immersion test. After 30 days immersion, noticeable deterioration was observed. Cores were compression tested after 42, 65, and 90 days of immersion and all resulted in low strengths. The failure of the immersion test dictated a review of the entire test program. It was decided to obtain cores from actual CSS production drums. They were expected to provide a better representation of the solidified sludge wash recipe than the test drums for the following reasons: the pH of the actual sludge wash waste solution was 10.4, instead of the test solution pH of 12.0 to 12.6; variations in sulfate concentrations were expected; and a "heel" of decontaminated supernatant may have been left in one of the LWTS tanks or the Waste Dispensing Vessel and piping.

Other tests indicated that sulfate attack was taking place in the form of crystal formation. The decision was made to begin waste form qualification using Portland Type V cement, which is known to have sulfate-resisting properties, and would even allow higher waste loadings (higher dissolved solids in the waste liquid).

The recipe qualification program was repeated, including screening tests. Eleven test drums were processed in CSS using actual decontaminated PUREX sludge wash liquid. The sulfate concentration was increased further, to provide an increased margin for potential operating conditions during the second and third sludge washes. Waste loadings at up to 33 wt.% TDS were tested. All test drums passed all tests by a wide margin.

6.4 THOREX-PUREX Wash Recipe Work

Waste form qualification testing was done before the start of THOREX/PUREX wash processing. No attempts were made to qualify a recipe using Portland Type I cement. Only the previously successful Type V cement blend was used. Although taking cores from actual, full-scale cement waste drums seems awkward and expensive, it proved to be more effective than producing scaled-down specimens in a laboratory mixer, and simulating drum curing in an oven. Scaled-down production and simulated curing could have introduced questions and concerns about scaling factors, mixing mechanics, and curing environments into the test process by using laboratory-produced specimens, and additional test variables. Instead, controls were added to the Process Control Plan, including: limiting and sampling Type V Portland Cement to confirm less than 5.0% tricalcium aluminate content; controlling cement blending; and sampling Tank 8D-2 on a monthly basis to ensure that major chemical and key radionuclide concentrations remained uniform, and within tested ranges.

As with the supernatant waste form, a program of core drilling and testing drums at regular intervals was instituted for both PUREX wash and THOREX/PUREX wash waste streams to test long-term compressive strength. In addition, an extra immersion test was added to PUREX wash and THOREX/PUREX wash long-term test plans to indicate immersion resistance, which is the most rigorous test criterion.

6.5 Cement Mixer Build up

Build up of residual cement at the end of each batch processing cycle was troublesome throughout CSS operations. Each of the waste streams processed through the CSS (i.e., supernatant, PUREX sludge wash liquid, and THOREX/PUREX sludge wash liquid), exhibited different gel times and characteristics. Cleaning the mixer with a high pressure water wash (30,000 psi) between campaigns proved to be very effective when combined with regular flushing done during processing.

As waste form work indicated, improper flushing or excessive flush water could lead to storage of hazardous wastes in the IRTS drain Tank (7D-13), or downstream in the low-level water treatment system. Flush volumes and flush frequency were modified to prevent this. Flush water was mixed with cement in special flush drums.

7.0 CONCLUSION

The Integrated Radwaste Treatment System (IRTS) was used to pretreat over two million liters of liquid high-level radioactive waste (HLW) that had been produced during spent nuclear fuel reprocessing operations that took place at the Western New York Nuclear Service Center.

IRTS pretreatment operations resulted in the production of 19,877 drums of cement encapsulated waste that achieved NRC criteria for stabilized LLW storage and disposal. Over 6.6 million curies of Cs-137 and Sr-90 were retained on zeolite loaded, ion-exchange columns mounted in Tank 8D-1. This zeolite was subsequently transferred to Tank 8D-2 for vitrification processing.

The IRTS operated in excess of its design rate, with a decontamination factor (DF) approximately 50 times better than the design minimum. The resulting lower radiation levels and exposure rates allowed many repairs and improvements to be made to downstream equipment that otherwise might not have been possible without complex remote systems and tooling.

During seven years of IRTS operation, three different HLW liquids were treated, supernatant, PUREX sludge wash liquid, and PUREX/THOREX sludge wash liquid. Design criteria used for the IRTS had built in flexibility that anticipated changing tank chemistry. The imaginative, forward-looking approach taken to IRTS design and operation contributed to the success of HLW pretreatment.

There were many challenges during IRTS operations, including failure of several pieces of inaccessible equipment. These items were repaired using remote tooling techniques that were developed by West Valley engineers, and the principles of the As Low As Reasonably Achievable (ALARA) philosophy. Combined engineering innovation, and ALARA philosophy, ensured safe production of a high quality product.

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ACRONYMS

ASTM	American Society of Testing Material
ALARA	As Low As Reasonably Achievable
Cs-137	Cesium-137
CSS	Cement Solidification System
DOE	United States Department of Energy
DF	Decontamination Factor
HLW	High-level Waste
IRTS	Integrated Radwaste Treatment System
LLW	Low-level Waste
LWTS	Liquid Waste Treatment System
NRC	United States Nuclear Regulatory Commission
Pu	Plutonium
PUREX	Plutonium/Uranium Extratction Process
Sr-90	Strontium-90
STS	Supernatant Treatment System
TCLP	Toxicity Characteristic Leaching Procedure
TDS	Total Dissolved Solids
THOREX	Thorium Extraction Process
U	Uranium
WVNS	West Valley Nuclear Services Company, Inc.
XC	Extraction Cell

M98002842

Report Number (14) DOE/NE/44139- -83

Publ. Date (11) 199710

Sponsor Code (18) DOE/NE, XF

UC Category (19) UC-510, DOE/ER

DOE