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**HIGH-LEVEL RADIOACTIVE WASTE MANAGEMENT
PROGRAM AT THE NATIONAL REACTOR TESTING STATION**

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I. INTRODUCTION

Most of the high-level radioactive waste at the National Reactor Testing Station (NRTS) is generated at the Idaho Chemical Processing Plant (ICPP) during the reprocessing of spent nuclear fuel. The fuel comes from nuclear reactors using highly-enriched ^{235}U fuel. The high-level radioactive waste is primarily the first-cycle raffinate from the solvent extraction of dissolved fuel solutions. This waste is self-heating in both the liquid and solid forms. Although the second- and third-cycle solvent extraction wastes are not self-heating, they will be treated as high-level waste. While solid plant waste and trash (contaminated with radionuclides) might be inferred by the title of this paper, such waste will not be discussed extensively here, because the quantity of radionuclides involved is very small compared to those from the first-cycle raffinate.

The principal concern in the management of high-level waste at the ICPP is to store the liquid waste safely within a period not to exceed five years, calcine the liquid waste to a dry solid, and store the solid safely in engineered storage bins where it will be retrievable at all times^[1]. The dry storage bins are designed for a lifetime of hundreds of years although removal of the radioactive waste for further treatment or transport to a Federal repository might be accomplished whenever desired.

II. SOURCES OF HIGH-LEVEL RADWASTES AT THE ICPP

The first-cycle raffinate wastes produced at the ICPP are the acid aluminum waste from various test reactor fuels, fluoride-bearing waste from zirconium-matrix fuel, a small amount of stainless steel sulfate waste from fuel from developmental reactors such as the Organic Moderated Reactor Experiment (OMRE), acid stainless steel nitrate waste from the electrolytic dissolution of Experimental Breeder Reactor-II (EBR-II) reactor fuel, and an acid waste from the recovery of uranium from nuclear rocket fuel^[2,3]. The second- and third-cycle raffinates for all of these fuels have a common composition. Typical chemical compositions of the liquid wastes are given in Table 1. Radiochemical compositions of the content of three of the first-cycle zirconium-bearing waste tanks are given in Table 2 and of three tanks containing second- and third-cycle waste in Table 3^[4]. Based on the isotopic content for Pu and U and cooling times at the ICPP, the concentrations of ²⁴¹Am, ²⁴³Am, ²⁴²Cm and ²⁴⁴Cm have been calculated for the average high-level zirconium-bearing waste to be 3.8×10^{-5} , 1.1×10^{-6} , 9.9×10^{-10} and 6.6×10^{-8} g/l, respectively. Most of the first-cycle raffinates are self-heating, while the second- and third-cycle wastes and waste-evaporator bottoms are low-heat liquid wastes. Intermediate level wastes are evaporated, the overhead is cleaned by ion exchange columns to less than RCG limits before discharge to the water table, and the bottoms processed as high-level waste. Individual storage tanks generally contain components of several types of waste.

To date the ICPP has produced approximately 4.6 million gallons of high-level liquid waste of which 2.0 million gallons are in the liquid form as shown in Table 4 and 2.6 million have been calcined to a solid. High-level wastes began accumulating at the ICPP in 1953 and the rate of production has steadily increased^[3]. New wastes are anticipated from new fuels and will be discussed in Section VIII.

TABLE 1

**TYPICAL CHEMICAL COMPOSITIONS OF THE LIQUID WASTES IN INTERIM STORAGE AT ICPP
(Molarity, M)**

<u>Component</u>	<u>Acid Aluminum</u>	<u>Fluorid- Bearing</u>	<u>Stainless Steel Sulfate</u>	<u>Electrolytic</u>	<u>Graphite^(a)</u>	<u>Second- and Third-Cycle</u>
H ⁺	1.0	1.5-2.0	3.2	2.2	2.3	0.1-1.6
Al ⁺³	1.16	0.7	—	0.2	0.3	0.5
Zr ⁺⁴	—	0.45	—	—	—	T
Cd ⁺²	—	0-0.1	—	—	—	—
Nb ⁺⁵	—	< 0.01	—	—	0.046	—
Na ⁺	0.065	0.003	—	0.004	—	1.6-2.3
Fe ⁺³	0.005	0.004	0.069	0.09	—	0.02
Cr ⁺³	—	T	0.017	0.03	—	—
Ni ⁺²	—	T	0.01	0.01	—	0.0004-0.003
NH ₄ ⁺	—	—	—	0.1	0.06	0.04
Gd ⁺³	—	—	—	0.0025	—	—
Fissium ^(b)	—	—	—	0.004	—	—
B ⁺³	—	0.2	—	—	0.065	0.005-0.2
Hg ⁺²	0.018	—	—	—	—	0.0007-0.005
K ⁺	—	—	—	—	—	0.2
NO ₃ ⁻	4.6	2.4	2.6	3.2	2.0	4.6-5.0
SO ₄ ⁻²	0.01	—	0.6	—	—	0.04-0.06
PO ₄ ⁻³	—	—	—	—	—	0.02
F ⁻	—	2.5-3.2	—	—	1.2	0.0007-0.007
Cl ⁻	—	0.001	—	—	—	0.03

(a) Design estimates.

(b) Fissium is composed of 48 w/o molybdenum, 40 w/o ruthenium, 4 w/o rhodium, 4 w/o palladium, and 2 w/o zirconium.

T = Trace, less than 0.1 w/o.

TABLE 2

RADIOISOTOPE ANALYSIS OF FIRST CYCLE WASTE (ICPP)

Isotope	Tank No. WM-185	Ci/t WM-187	WM-188
^{60}Co	1.5×10^{-3}	9.0×10^{-4}	1.5×10^{-3}
^{95}Zr	4.3×10^{-3}	8.7×10^{-3}	$< 1.6 \times 10^{-3}$
^{95}Nb	4.9×10^{-3}	1.4×10^{-2}	$< 8.9 \times 10^{-4}$
^{106}Ru	1.1×10^{-1}	9.0×10^{-2}	2.5×10^{-2}
^{125}Sb	3.3×10^{-2}	3.0×10^{-2}	1.8×10^{-2}
^{134}Cs	2.1×10^{-1}	2.1×10^{-1}	1.2×10^{-1}
^{135}Cs	1.0	7.0×10^{-1}	8.0×10^{-1}
^{144}Ce	1.0	9.0×10^{-1}	2.0×10^{-1}
^{154}Eu	1.5×10^{-2}	1.1×10^{-2}	1.3×10^{-2}
Gross β	6.5	4.7	3.2
^{90}Sr	9.4×10^{-1}	1.9×10^{-1}	2.9×10^{-1}
^3H	8.1×10^{-4}	1.7×10^{-4}	1.2×10^{-4}

PLUTONIUM ANALYSES

^{239}Pu , wt%	72.2	67.1	70.4
^{240}Pu , wt%	17.6	20.5	18.9
^{241}Pu , wt%	7.8	9.2	8.2
^{242}Pu , wt%	2.5	3.4	2.7
Total Pu, g/t*	1.93×10^{-3}	1.19×10^{-3}	1.54×10^{-3}

* ^{238}Pu constitutes an additional 5% to the g Pu/t

URANIUM ANALYSES

^{234}U , wt%	1.7	1.2	1.3
^{235}U , wt%	56.1	72.6	30.8
^{236}U , wt%	5.9	6.3	3.6
^{238}U , wt%	36.3	19.9	64.3
Total U, g/t	4.7×10^{-4}	9.3×10^{-4}	2.8×10^{-3}

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Np, g/t	1.19×10^{-5}	1.57×10^{-5}	9.74×10^{-6}
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TABLE 3

RADIOISOTOPE ANALYSIS OF SECOND- AND THIRD-CYCLE WASTE (ICPP)

Isotope	Tank No	Ci/t		
		WM-186	WM-184	WM-180
^{95}Zr		6.2×10^{-5}	1.0×10^{-5}	3.3×10^{-4}
^{95}Nb		3.3×10^{-5}	4.2×10^{-6}	1.0×10^{-4}
^{106}Ru		8.6×10^{-3}	1.5×10^{-4}	3.3×10^{-3}
^{125}Sb		6.7×10^{-4}	2.4×10^{-4}	3.0×10^{-3}
^{134}Cs		1.0×10^{-3}	2.6×10^{-5}	1.0×10^{-3}
^{137}Cs		5.6×10^{-2}	1.2×10^{-2}	2.4×10^{-1}
^{144}Ce		2.8×10^{-3}	4.0×10^{-5}	7.6×10^{-4}
^{154}Eu		2.6×10^{-4}	2.9×10^{-5}	7.0×10^{-4}
^{60}Co		—	1.0×10^{-5}	—
^{90}Sr		4.7×10^{-2}	1.2×10^{-2}	2.1×10^{-1}
^3H		6.1×10^{-5}	1.0×10^{-4}	3.4×10^{-5}
Gross β		2.1×10^{-1}	4.4×10^{-2}	7.7×10^{-1}

PLUTONIUM ANALYSES

^{239}Pu , wt%	—	88.3	86.1
^{240}Pu , wt%	—	10.3	8.6
^{241}Pu , wt%	—	1.1	4.9
^{242}Pu , wt%	—	0.4	0.4
Total Pu, g/t ^(b)	(a)	1.893×10^{-3}	0.938×10^{-3}

(a) WM-186 contained a constituent that interfered with the analysis.

(b) ^{238}Pu constitutes an additional 5% to the total Pu.

URANIUM ANALYSIS

^{234}U , wt%	0.09	0.95	0.65
^{235}U , wt%	5.86	64.39	43.70
^{236}U , wt%	0.52	3.07	2.08
^{238}U , wt%	93.53	31.59	53.57
Total U, g/t	0.191	2.01×10^{-2}	2.31×10^{-2}

TABLE 4

SUMMARY OF ICPP TANK FARM LIQUID WASTE INVENTORY
AS OF FEBRUARY 26, 1974

Tank No.	Type of Waste	Volume (gal)
WM-180[a]	Second- and Third-Cycle, and PEW Evaporator Bottoms	265,000
WM-181[b]	Spare (for Service Waste Diversion)	22,000
WM-182[a]	Fluoride	139,000
WM-183[a]	Nonfluoride	112,800
WM-184[b]	Second- and Third-Cycle, and PEW Bottoms	283,000
WM-185[a]	Fluoride	278,000
WM-186[b]	Second- and Third-Cycle, and PEW Bottoms	283,000
WM-187[a]	Fluoride	277,000
WM-188[a]	Fluoride	14,000
WM-189[a]	Fluoride	144,400
WM-190[a]	Spare	0
WM-103[c]	Stainless Steel Sulfate	16,000
WM-104[c]	Spare	0
WM-105[c]	Stainless Steel Sulfate	1,800
WM-106[c]	Stainless Steel Sulfate	15,000
TOTAL		1,839,000

[a] 300,000-gal tank with cooling coils.

[b] 300,000-gal tank without cooling coils.

[c] 30,000-gal tank with cooling coils.

III. INTERIM STORAGE OF HIGH LEVEL RADIOACTIVE LIQUID WASTES

The high-level radioactive liquid wastes are stored in stainless steel 300,000-gallon tanks located in underground concrete vaults pending their solidification in the Waste Calcining Facility (WCF). At present the liquid waste is stored for a period under four years and is dependent in part on the rate of calcination and rate of waste generation. The tank farm consists of eleven 300,000-gallon tanks and four special tanks of 30,000 gallons each.

A schematic of one of the 300,000-gallon tanks is shown in Figure 1[5,6].

The 300,000-gallon tanks are 50 ft in diameter with a 21-ft high wall and a 32-ft height from the floor to the top of the hemispherical roof. The floor is flat and the sides are vertical. The vessel floors and lower 8 ft of the sides are made from 5/16-in.-thick stainless steel plate, and the hemispherical roof is 3/16-in.-thick stainless steel plate. The tanks are designed for a liquid with a specific gravity of 1.4, for a temperature of 200°F, and a pressure of 3.9 in. water vacuum.

The 300,000-gallon waste tanks and the associated concrete secondary containment vaults were analyzed for their dynamic response to a hypothetical earthquake occurring at the NRTS, using the STRAP-D (Structural Analysis Package - Dynamic) computer code^[7]. Ground acceleration histories from four of the largest West Coast earthquakes were used as forcing functions for the code; viz, the 1940 El Centro, 1949 Olympia, 1952 Taft, and 1966 Parkfield.

Calculations show that the waste storage tanks and vaults will withstand the severe ground shaking accompanying very strong earthquakes^[6]. Even if subjected to the 1940 El Centro earthquake record scaled to a peak ground acceleration of 0.5 g, the waste tanks were stressed to 21,300 psi which is 59.1% of the 36,000 psi required to cause tank failure. Using the unscaled 1940 El Centro earthquake record which had a peak ground acceleration of 0.33 g, 50.4% of yield stress in the tanks was produced. An acceleration of 0.33 g corresponds to the acceleration expected at ICPP from a maximum hypothetical earthquake of Richter magnitude 7-3/4 located on the Arco fault at a point 15 miles from ICPP. It does not take into account the appreciable energy attenuation which likely would occur because of the particular geological structure of the Snake River Plain.

Under the same conditions, analysis of the reinforced concrete vaults housing the storage tanks showed that a peak ground acceleration of 0.5 g would cause a peak stress of 87.7% of yield stress while the unscaled 1940 El Centro record would impose 79% of yield stress of the vaults.

It is emphasized that an earthquake producing a 0.33-g peak ground acceleration at ICPP is considered a purely hypothetical upper-limit event which has not been exceeded in magnitude by any earthquake in recent geological times. The Arco fault has not been seismically active in historic time.

Fig. 1. Schematic diagram of typical 300,000-gal liquid waste tank at ICPP.

The liquid level in the 300,000-gallon tanks that contain highly radioactive waste is continuously monitored. A level indicator in each tank is set to alarm when the volume reaches 285,000 gallons. This alarm sounds in both the control house and the main process building. A second leak detection system is a liquid-level indicator in the sump of the vault surrounding the tanks.

Internal tank pressure is also measured and transmitted to the control panel. If the relief valve is activated, the pressure is released to a line that vents directly to the stack. The normal tank venting system is connected to the process vessel off-gas system in the main plant, which is a filtered system.

Specific gravity of the liquid waste is measured, and the value is transmitted to a recorder which has an alarm in both the control house and the main process building that activates at a specific gravity of 1.35.

Multiple temperature points in the tanks provide individual temperature measurements for each 2 ft of vertical wall height and for the tank bottom. In addition, a complete set of thermocouples is provided on the opposite side of each tank. A total of 24 points is available for each of the cooled tanks (10 for each uncooled tank). The temperature of each tank is controlled below 50°C for corrosion control and is continuously recorded on multiple point recorders. A high-temperature alarm is installed for each tank, with alarms at the control house and at the main process building. Normally the wastes are well below the high-temperature alarm point.

Eight of the eleven 300,000-gallon tanks contain a closed-loop cooling system consisting of coils of stainless steel pipe around the walls and on the floor of the tank. The water pressure in the cooling coils is greater than the pressure in the waste tanks so that normally if a leak occurred the cooling water would leak into the tank. However, a radiation monitor is installed on the return coolant line from the tank and will alarm in the event the cooling water becomes contaminated. By closing the individual valves on the 30 cooling coils in each tank, and then opening these valves one at a time and observing the radiation monitor, the leaking coil could be located and isolated. Contaminated water can be transferred to the waste evaporator system for processing, and fresh water can be added to the cooling coils. The tanks with cooling coils have been in operation since 1953, and no leak has occurred in any of the tanks or cooling systems to date.

Each of the 300,000-gallon storage tanks is contained in an individual concrete vault which serves as a secondary containment barrier if waste leaks from the tank. The vault is constructed of reinforced concrete, approximately 2 ft thick. The vaults are either octagonal or square and are about 60 ft across and 33 ft tall.

The tanks are also connected to reflux condensers, which will condense the vapors if the waste boils. These condensers are stainless steel shell-and-tube condensers contained in concrete housings in the tank farm area. The condenser pits have sumps equipped with jets to transfer leakage back to one of the waste tanks; however, the condensers are not normally used.

Each of the tanks within the vaults rests on a thin layer of sand on top of a concrete pad at the bottom of the vault; the bottom of the vault rests on bedrock. The vaults generally have 1- to 3-ft-deep by 1- or 2-ft-square sumps to collect liquid. An increase in the level in the sump indicates that the tank contents are leaking or groundwater from outside the vault has leaked in. Groundwater has been observed to enter past the cover of the vault which is not sealed. Any liquid in the sumps can be returned either to the tank or to the main plant for processing. Each sump has a measuring device which indicates the liquid level in the sump. The measuring unit is connected to a multipoint recorder that records the liquid level in all of the sumps continuously. The readings for the sumps are recorded in a logbook once each 8-hr shift. The system is also connected to an alarm which sounds in the control house and in the main plant if the level exceeds a predetermined value (15 in.). Water in the vault sumps can be sampled manually by submerging a bottle attached to a handline; remote samplers are being installed.

One of the tanks containing cooling coils always remains empty to serve as a spare; thus, if a leak develops, the contents of the leaking tank can be transferred to the spare. A freeboard volume equivalent to 300,000 gallons is also retained in the sum of the other seven cooled tanks so that the liquid from a leaking tank can be distributed among seven tanks instead of all of it going to the spare tank. In effect, two empty tanks are available for any emergency in the tank farm.

Three of the 300,000-gallon tanks that do not contain cooling coils are used to store radioactive liquid wastes that do not generate any appreciable heat because the concentration of fission products is too low. One of these three tanks is retained as a spare in the event that failure of a process vessel containing radioactive solution contaminates normally nonradioactive cooling water. If this occurs, the contaminated cooling water is diverted to the spare tank and later processed in the main plant waste system to remove the radionuclides before the water is released to the discharge well.

The four 30,000-gallon stainless steel tanks are buried in a horizontal position. They are 12 ft in diameter and have a 38-ft-long straight side. The shell and head of these tanks are 11/16 and 9/16 in. thick, respectively. They also are equipped with cooling coils and are connected to a shell-and-tube condenser to remove heat produced by the decay of radionuclides. These tanks rest on a concrete pad but are not surrounded by a vault. A curb surrounds the concrete pad, and a sump equipped with a level alarm collects any leakage from the tanks. A 24-in.-diameter pipe extends from the surface of the ground to the sump so that a portable unit can be used to empty the sump if necessary.

The four 30,000-gallon tanks were used to store wastes from early processes used at the ICPP. One of these tanks has been emptied, one is nearly empty, and the other two are about one-half full but will be emptied in the near future. When emptied, none of these tanks are planned for the routine storage of radioactive liquids, as they do not meet the present secondary containment criterion for storage of high-level radioactive waste.

All underground waste lines (totalling 9,000 ft) in the tank farm are stainless steel. All lines that carry radioactive waste have either: (a) a stainless steel-lined concrete encasement, (b) a secondary steel pipe with centering spacers, or (c) a tile pipe with spacers encased in concrete^[8]. This construction provides two safety features as follows:

- (1) It prevents contact of the primary waste line with the soil to avoid underground corrosion.
- (2) It provides very rapid detection of leaks in the primary line by the use of monitors.

The lines carrying radioactive waste go through junction boxes constructed of concrete with a stainless steel liner. If radioactive waste leaks into a junction box or the secondary pipe encasement, it drains to a sump in a 300,000-gallon tank vault. There is no evidence, direct or indirect, of any leak in the waste lines to date. The jet system on each tank is equipped with a siphon break to avoid creating a siphoning action. During periods of surface runoff resulting from melting snow or heavy rains, 5 of the 11 vaults containing 300,000-gallon waste storage tanks collect water from in-leakage through the top of the vault. This water is periodically transferred into the tank or to the waste evaporator for processing. The path to the vault of the surface water has recently been traced and means to eliminate it are underway.

IV. SOLIDIFICATION OF HIGH-LEVEL LIQUID WASTES IN THE WASTE CALCINING FACILITY (WCF)

The policy at the ICPP is to solidify the high-level radioactive liquid wastes and to store on-site the resulting granular solids in engineered bins where the waste will be in safe interim storage and fully retrievable. In the solid form the waste is less mobile than as a liquid; the volume of the waste is reduced by a factor of 9:1; the cost of solidification and storage is less than building more tanks for liquid wastes; and the operation meets the basic philosophy of waste management of the USAEC.

The WCF at the ICPP has been solidifying high-level wastes in a fluidized-bed calciner since December 1963^[9,10]. As of January 1974, over 1,510,000 gallons of aluminum-type waste and 1,089,000 gallons of zirconium-type waste had been calcined to give a total of 42,500 cubic feet of solids. The original process was developed for solidifying high-level waste from the processing of aluminum-clad test reactor fuel. The process has been modified to calcine fluoride-bearing zirconium waste, stainless steel sulfate waste, and ammonium nitrate-containing waste. The WCF is a vital unit in an integrated program of spent fuel storage, fuel reprocessing, and waste management at the ICPP. An aerial view of the WCF and three sets of solid storage bins is shown in Figure 2.

A schematic flowsheet of the WCF is shown in Figure 3. Solidification takes place in the 4-ft-diameter by 13-ft-high fluidized-bed calciner. Most of the plant equipment is devoted to cleaning up the entrained radioactive particles in the calciner off-gas. The off-gas passes into a cyclone to remove most of the solids, a quench tank to cool the gas, a venturi scrubber-separator-demister, a silica gel adsorber for the removal of volatile ruthenium, another cyclone, and finally a high-efficiency HEPA filter to remove submicron particles before discharging to the 250-ft plant stack.

In the calciner, the liquid waste is pneumatically atomized from three nozzles at 85-140 gph into the heated 6-ft-deep fluidized bed of solidified granular waste at 400-500°C. The process is endothermic; therefore the bed is heated by in-bed combustion of an oxygen-atomized stream of kerosene. The process is started with a bed of granular material such as dolomite, which is replaced by calcine as the operation continues. By preheating the bed with externally-heated fluidizing air to a bed temperature of 360 to 400°C, the atomized kerosene will ignite in the presence of a nitrate waste.

Nitric acid and water are condensed in the off-gas scrubbing system to dissolve the entrained calcine. The off-gas scrubber solution is recycled at 15 to 30 gph by returning it to the calciner feed; the recycle solution is 20-30% of the total feed to the calciner. Based on the cross-sectional area of the empty calciner vessel, the superficial inlet fluidizing velocity is 0.6 to 1.2 ft/sec. The volumetric ratio of atomizing air to liquid feed is varied as necessary to cause sufficient breakup of bed particles by impingement and thereby control the average bed particle size at a desired value. The value of this volumetric ratio varies from 500 to 600 for alumina-type waste to a value of 700-800 for zirconium-type waste. Bed height is controlled by the rate of removal of calcined solids.

Fig. 2. Aerial view of WCF.

ACC-A-0008

Fig. 3. Schematic flowsheet (WCF).

The originally built calciner was heated by a heat exchanger using liquid sodium-potassium alloy as a transfer medium which was heated externally in an oil-fired furnace. Although 35,000 hours of satisfactory services were obtained with NaK, an in-bed combustion process was developed and installed to obtain a greater processing rate; other changes noted were lower vessel wall temperatures, lower ruthenium volatility, and increased reliability.

Typical physical, chemical, and radiochemical properties of calcined solids from acid aluminum waste and fluoride-bearing zirconium waste are given in Table 5. Other components in the calcine, e.g., actinide elements, can be calculated on the basis of the analyses of the liquid waste.

TABLE 5
TYPICAL PROPERTIES OF CALCINER PRODUCT

Physical properties:	Aluminum Waste	Zirconium Waste
Mass median particle diameter, mm	0.56 to 0.70	0.6 to 0.8
Bulk density, g/cc	1.0 to 1.2	1.7
Composition (wt%):		
Zirconium as ZrO_2	—	21.4
Calcium as CaF_2	—	54.2
Aluminum as Al_2O_3	88.2 to 89.1	21.9
Sodium as Na_2O	1.3 to 2.0	—
Nitrogen as N_2O_5	3.9 to 4.1	1.9
Mercury as HgO	2.9	—
Water	2.0	0.6
Gross fission product oxides	0.6	—
Radioactive properties:		
Heat generation, Btu/hr-lb	0.3 to 0.8	0.1 to 0.2
Principal radioisotopes, Ci/lb		
Strontium-90	4.9 to 7.2	0.8 to 2.5
Cesium-134	0.2 to 0.4	—
Cesium-137	5.2 to 8.0	1.0 to 3.0
Cerium-144	1.3 to 17.3	—
Ruthenium-106	0.04 to 0.5	0.01 to 0.03
Zirconium/niobium-95	0 to 0.5	—
Promethium-147	3.3 to 10.6	—

Typically current performance of the individual units in the WCF off-gas cleaning system is summarized in Figure 4^[9].

Fig. 4. Typical decontamination factors achieved in WCF off-gas system.

Five campaigns have been completed in the WCF. Overall off-gas decontamination factors achieved, when indirect heating was used for the calciner, varied from 0.8×10^7 to 0.9×10^8 for particulates. In campaign 4, in-bed combustion was used and the overall decontamination factor for particulates was 1.4×10^7 , and for ruthenium-106 it was 0.8×10^6 .

In Table 6 are summarized typical operating conditions for the WCF as applied to the two major liquid wastes at the ICPP. The conditions for the calciner vessel are based on indirect heating with NaK; in-bed heating gives similar results except that the bed might be subject to short periods of exposure to a high temperature in the immediate vicinity of the burning kerosene. Similarly, the off-gas composition leaving the calciner will depend on the method of heating.

TABLE 6
TYPICAL CALCINER OPERATING CONDITIONS

Feed Rates:	Aluminum Waste	Zirconium Waste
Gross to calciner, gph	85 - 100	85 - 100
Recycle to calciner, gph	15 - 30	25 - 30
Calcium nitrate, gph ^(a)	~	20 - 30
Net to calciner, gph	60 - 80	45 - 55
Product Rate:		
To solids storage, lb/hr	50 - 55	80 - 90
Calciner Conditions:		
Superficial fluidizing velocity, ft/sec	1.0	1.3
Volume ratio of nozzle air to feed rate	510 - 660	650 - 800
Bed temperature, °C	400	400
NaK inlet temperature, °C	690 - 735	690 - 735
Bed height, in.	73	73
Mass median bed particle diameter, mm	0.5 - 0.8	0.6 - 0.8
Scrubbing System:		
Solids entering scrubbing system, lb/hr	9 - 13	10 - 20
Aluminum, M	0.9 - 1.1	0.2 - 0.3
Acid, M	2 - 4	2 - 5
Undissolved solids, g/l	2	1 - 4
Scrubbing solution rate to quench tower, gpm	60 - 62	60 - 62
Pressure drop across quench tower, in. H ₂ O	3	3
Scrubbing solution rate to venturi scrubber, gpm	11 - 13	11 - 13
Pressure drop across venturi scrubber in. H ₂ O	55 - 70	55 - 70
Ruthenium Adsorption and Filtering:		
Total gas flow, scfm	900 - 1600	900 - 1600
Dew point of gas, °C	59 - 64	55 - 65
Adsorber operating temperature, °C	66 - 74	65 - 70
Pressure drop across adsorbers, in. H ₂ O	6 - 9	6 - 9
Final filter operating temperature, °C	80 - 85	80 - 85
Pressure drop across final filters and adjacent piping, in. H ₂ O	5 - 10	5 - 10

(a) To precipitate fluoride in waste.

V. STORAGE OF HIGH-LEVEL WASTE CALCINE

The calcined product from the WCF is spherical and granular in nature and readily transported pneumatically to the calcine storage bins (one hundred yards distant) where it will be safe and readily retrievable.

The WCF calcined product is stored in vented stainless steel enclosed bins located in underground concrete vaults^[11]. The original waste storage facility, consisting of four of the annular bins shown in Figure 5a, has been filled with approximately 7,500 cubic feet of calcined solids. The second solids storage facility (shown in Figure 5b) consists of seven 12-ft-diameter by 42-ft-high cylindrical bins nested in a 46-ft-diameter concrete vault. The bins are designed for a centerline temperature of 700°C. This facility, having a combined storage capacity of approximately 30,000 cubic feet, was filled by February 1972. Thus, construction of a third set of bins (similar to those of Figure 5b) was started during 1969 and filling of the new bins started shortly after their completion in 1972.

During filling of the bins, a cyclone separator above the bins removes the solids from the transport air stream, the solids drop into a vertical standpipe (inside the solids storage vault), and the transport air returns to the calciner for decontamination. Solids collected in the vertical standpipe are directed to one of seven stainless steel bins by inclined lateral lines and a solids flow diverter valve. The lateral lines, five in number, are attached to the standpipe at different elevations, and the corresponding bins fill sequentially as the level of solids rises in the standpipe. A diverter valve located at the base of the standpipe is used to direct the solids, by means of air purges, to either of the two remaining bins. This valve normally blocks the flow of granular solids by use of a horizontal offset which is sealed by the angle of repose of the solid material. When flow into the bin is desired, a small flow of air admitted to the horizontal section disturbs the angle of repose and permits the solids to flow.

Natural-circulation cooling air passes through the vault surrounding the bin and out a stack, thus removing a considerable portion of the heat liberated from the stored solids. Direct radiation from the bins to the surrounding vault wall, and thence conduction into the ground, removes the balance of the heat liberated. The cooling air leaving the bin vault is continuously monitored for radiation (which, if present, would indicate a possible bin failure) and entrainment of particles. Should radiation be detected significantly above background, the monitor sounds an alarm and closes a valve in the cooling air duct. Closing the valve will halt the cooling air flow and prevent contamination of the environment. In a test with a full set of bins with the cooling duct closed, heat was removed through the vault wall with only an increase of 50°C in the centerline temperature in the bins. Each vault contains a sump, sample lines, and jet with liquid-level instrumentation for detecting the entry of groundwater, rainwater, or floodwater into the vault. If liquid is detected, it can be transferred to the ICPP liquid waste evaporator system. No liquid has been detected in the vaults since initial operation of the WCF.

Fig. 5a.

Fig. 5b.

Special features of the storage facility include installation of corrosion coupons and provisions for solids removal. Some 160 sample coupons of various alloy metals—ASTM Types 405, 304, and 304L stainless steels, and AISI Type 1025 carbon steel—have been attached to cables and inserted into the bins. At various periods extending up to 80 years, the samples will be withdrawn, examined, and measured for corrosion. The results should assist in determining the best construction materials for such long-term solids storage. A set of corrosion samples was removed in 1973 from a bin of alumina waste and another set from zirconia waste. No particle agglomeration was evident, nothing was coated on the metal specimens, and the corrosion rate was minimal^[12]. Provision for a solids removal system have been made, since it is possible that advancing technology or integral failure of the bins in the facility may make it desirable or necessary to remove the calcined solids. A 6-in.-diameter pipe extends from the top of each bin upward through the vault roof. If required, the pipe can be cut off and a pneumatic solids removal system inserted down the pipe for retrieval of the solids from the bin.

Laboratory-scale tests, performed on radioactive solids during the first WCF processing campaign, established the feasibility of using a 700°C maximum temperature for design of the second set of storage bins. These initial tests have now been extended to determine high temperature effects on radioactive solids for periods lasting at least one year. The significant conclusions of this work are: (1) essentially no cesium volatilizes at temperatures up to 700°C, but significant amounts volatilize at temperatures of 800°C or higher; (2) no strontium or cerium volatilize at temperatures of up to 1200°C; (3) the volatilization of appreciable quantities of ruthenium and mercury is highly time-dependent, but does occur at temperatures as low as 525°C (ruthenium volatilizes very slowly, while mercury volatilization is rapid); (4) the originally amorphous alumina formed in the WCF will convert to either gamma or alpha crystalline alumina during storage, depending on temperature (only gamma alumina forms below 800°C; alpha alumina predominates above this temperature); and (5) volatile constituents condensed in cooler regions and, in absence of gas purges, were completely retained within the solids. On the basis of these results, it is believed that no radionuclides can escape from stored solids at high temperatures; instead, movement of the heat sources due to radionuclide migration may result in a stable situation in which the maximum temperature of the solids is lower than that without migration.

The solids storage facilities for WCF product have been designed and constructed to withstand any foreseeable natural or man-made occurrence without extensive damage or release of radioactivity to the environment. The storage vaults and bins were designed to withstand an earthquake of magnitude 7 with an epicenter as close as 20 miles. In addition, the stainless steel bins have sufficient corrosion allowance that their integrity can be maintained for at least 500 years, even if the storage area were totally flooded because of some unexpected change in present climatic conditions.

VI. POST CALCINATION TREATMENT OF RADIOACTIVE CALCINE

Although WCF calcine is stored at the ICPP in a safe condition and fully retrievable, no decision has been made at this time to ship it to a Federal repository. In the event that the calcine were to be moved, criteria for the shipping container or of the form of the solid waste have not been fully established. It is possible that one might be able to ship and store the calcine as it exists. On the other hand, one might prefer to change some of the characteristics of the granular calcine in order to reduce dispersability or leachability.

A development program is underway at the ICPP to examine methods of treating WCF calcine; some methods retain the granular nature of the WCF product while others convert it to a more massive form. Methods of treatment include: (1) conversion to a cermet by the incorporation of the calcine in a metal matrix such as aluminum or lead-tin alloy; (2) conversion to a ceramic by the addition of a ceramic material such as clay and sintering; (3) incorporation into a concrete by the addition of binders or cement; and (4) coating of the calcine particles with pyrolytic carbon, glazes, metals, etc.

Of course, the conversion of WCF calcine to a borosilicate glass has been demonstrated as a possibility since 1963, but this phase of the study is not being performed by Allied Chemical since extensive work on incorporation of waste in glass has been completed at Battelle's Pacific Northwest Laboratories.

The experimental program is in the early stages and a comparison of the products of various methods of treatment is only preliminary. A listing of advantages and disadvantages depends on the criteria to be imposed on the product as well as on little-known variables such as cost. However, a tabulation of properties of typical WCF calcine incorporated by various post treatment methods is given in Table 7^[13]. Also shown are the properties of the untreated WCF calcine. In some instances the numerical values of the property vary by several orders of magnitude, while in others only qualitative terms are used. The term retrievability is very important and can mean either a simple recovery (e.g., as from the WCF storage bins) in preparation for packaging and shipping to a repository, or recovery and use or conversion to another form. For example, if one converts a calcined waste to a glass or ceramic block, the unit may be easily retrievable for shipment but may be very difficult to process to another form or to recover specified fission products. On the other hand, untreated WCF calcine has had a minimum of treatment, can be converted to any other form, can be readily processed for desired components, and is fully retrievable.

TABLE 7
QUALITATIVE EVALUATION OF THE POST TREATMENT OF WCF CALCINE

PROPERTY	WCF CALCINE	WCF CALCINE POST TREATMENT METHOD				GLASSES
		CEMENT	CERMET	CERAMIC	GRAPHITE	
Thermal Conductivity cal/sec/cm ² / °C/cm	~.0007	.0007 to .005	.05 to .5	.0007 to .005	.005 to .01	.002 - .004
F. P. Leach Rate g/cm ² /day	50-95% Cs 30% Sr in 7 weeks	~10 ⁻⁴	10 ⁻⁵ - 10 ⁻⁷	10 ⁻⁵ - 10 ⁻⁸	10 ⁻⁴ - 10 ⁻⁷	10 ⁻⁵ - 10 ⁻⁷
Temperature Stability °C	~700°	200°	300° to 1200°	600° - 1500°	600° - 1500°	500° - 900°
Radiation Stability (R) Gross α, β, γ dose	>10 ¹²	>10 ¹²	~10 ¹⁴	~10 ¹⁴	>10 ¹² >	>10 ¹² May Devitrify
Ruggedness	Fair	Good	Very good	Good	Fair - Good	Good
Volume Increase Over Calcine		20 - 60%	20 to 100%	20 - 80%	30 - 150%	20 - 50%
Retrievability for future Improvement	Very good	Poor	Fair - Good	Poor - Fair	Poor - Fair	Poor
Uncontrolled Storage Safety	Fair	Fair	Good	Good	Fair	Fair

VII. ENVIRONMENTAL IMPACT OF WASTE SOLIDIFICATION AT THE ICPP

The direct impact of waste management at the ICPP on a segment of the general population is minimized by siting the plant at the NRTS, a remote government reservation of 900 sq mi. Releases of minor quantities of radionuclides under routine operating conditions are monitored by continuous air and water sampling and measuring instruments. Potential releases of radionuclides under postulated accident conditions and their environmental impact have been calculated and reported in various safety review and environmental impact documents^[7,10,11,14].

1. Routine Releases

The high-level waste operations under consideration here are the storage of liquid waste, waste calcination, and storage of calcine in bins. In the 20 years of storing high-level liquid waste, there have been no instances of releases to the ground, nor even of leaks of waste from the tanks to their surrounding vaults. Similarly, there have been no releases of waste from the solid calcine storage bins and vaults. Decay heat from the liquid storage tanks and the calcine bins is discharged to the ground and atmosphere. The temperature rise of the ground in the immediate vicinity of these vaults is a few degrees and has no significant impact on man's environment.

The WCF calciner generates a significant quantity of fine calcined particles which are removed from the gas stream in the air cleaning equipment with a decontamination factor of approximately 1×10^6 . Volatile compounds of radoruthenium and tritium are also formed; the ruthenium is removed to low levels in adsorbers and the tritium is released to the environment. In Table 8 are given the average annual releases to the atmosphere of tritium, ^{106}Ru , ^{90}Sr , ^{137}Cs , and total alpha from the WCF at an operating rate of 2,100/gal/day for 150 days/yr; the average concentration at the NRTS boundary for these radionuclides assuming the nearest downwind boundary and a dispersion factor, X/Q of $3.8 \times 10^{-8} \text{ s/m}^3$; and the Radiological Concentration Guide values for these isotopes in air for a 168-hr week in an uncontrolled area^[14].

TABLE 8

RELEASES OF RADIONUCLIDES TO THE ATMOSPHERE FROM THE NORMAL OPERATION OF THE WCF

<u>Nuclide</u>	<u>Ci/yr to Atmosphere</u>	<u>Ave Conc. ($\mu\text{Ci/ml}$) at NRTS Boundary</u>	<u>RCC_a $\mu\text{Ci/ml}$</u>
Tritium	1200	1.4×10^{-12}	2×10^{-7}
^{106}Ru	1	1.2×10^{-15}	2×10^{-10}
^{90}Sr	1	1.2×10^{-15}	3×10^{-11}
^{137}Cs	1	1.2×10^{-15}	5×10^{-10}
Total Alpha	4×10^{-3}	4.8×10^{-18}	6×10^{-14}

From a comparison of the RCG_a with NRTS boundary concentrations, the atmospheric releases have had a very small impact on the environment. It should be pointed out that the daily release rates vary by as much as a factor of 20 and the values in the table are annual integrated releases.

Liquid wastes are generated in the decontamination of the WCF during down times for maintenance. The PEW wastes are evaporated, the bottoms recycled, and overhead further cleaned in an ion exchange column. The final effluent is discharged to the service well at 50,000 gal/yr of low-level liquid waste as condensate. These wastes are also discharged to the service waste system without significant effect on the environment.

In addition to gaseous and liquid waste, solid waste contaminated with radionuclides are produced by the WCF. Routine wastes of this nature include contaminated rags, blotting paper, plastic wrap, gloves, shoe covers, wood, glass, floor sweepings, scrap metal, HEPA filters and prefilters, spent ion exchange resin, and used equipment. Most of these wastes are packed in cardboard boxes and disposed of in the NRTS Burial Ground. The HEPA filters from the WCF off-gas system are buried at the Burial Ground in their stainless steel housings. In the future it is planned to leach the great majority of radionuclides from the spent HEPA filters and recycle the waste to the calciner. The annual rate of generation of the solid waste is 500 ft^3 with a content of approximately 40-100 curies of gross fission product activity.

Nonradioactive wastes are released during calcination; the major components are nitrogen oxides, carbon monoxide, and particulates. Estimated releases are shown in Table 9 for WCF operation at 2,100 gal/day for 150 days/yr and an atmospheric dispersion factor (X/Q) of $3.8 \times 10^{-8} \text{ s/m}^3$. Air quality standards shown in this table are from 42CFR410.

TABLE 9

ATMOSPHERIC RELEASES OF CHEMICAL POLLUTANTS FROM THE WCF

<u>Chemical</u>	<u>Release Rate, T/d</u>	<u>Ave. Conc. at NRTS Boundary, $\mu\text{g}/\text{m}^3$</u>	<u>Air Quality Std, $\mu\text{g}/\text{m}^3$</u>
NO_x	1.6	0.26	100
CO	1	0.2	10,000
Particulates	1.5×10^{-9}	2.5×10^{-10}	75

2. Actual and Postulated Abnormal Releases

A few, relatively minor incidents, resulting in releases of radionuclides, have occurred in the 10 years of operation of the WCF. As already stated, there have been no releases from the liquid storage tanks or the calcine bins.

Incidents are described briefly:

- a. In January 1972, approximately 1 Ci of particulate activity—principally ^{106}Ru —was released from the ICPP during WCF operation. The activity was released as very fine particles, but the cause was never found.
- b. In May 1972, an open decontamination line in the calcine transport air line resulted in the release to the ground surface of approximately 10 Ci of long-lived fission product activity. The majority of the release was packaged with accompanying soil and sent to the NRTS Burial Ground; the remainder was covered with clean soil and left *in situ*.

An analysis of land commitment, water resource commitment, impact on biota and impact on human activity has been made on the basis of the general radiation levels at the NRTS and vicinity in 1972^[14]. The contribution of the WCF to the total activity at the NRTS indicates negligible environmental impact to the general public^[15,17].

The main concern in the management of high-level waste is the environmental impact of a postulated maximum credible accident. The major environmental effect from an accident would be from the storage of high-level liquid waste; accidents to the WCF or to the solid storage bins will have an appreciably lesser effect. However, the entire high-level waste system is designed to resist earthquakes, tornadoes, variations in temperature, and floods of maximum credible magnitude.

VIII. SOLIDIFICATION OF OTHER WASTES AT ICPP^[16]

Aluminum nitrate and zirconium fluoride containing high-level liquid waste have been routinely calcined at the ICPP. Wastes containing ammonium nitrate and also stainless steel sulfate wastes have been satisfactorily calcined in the WCF. Calcination process flowsheets are being developed for new wastes from fuels to be processed at the ICPP such as from graphite fuels and the High Temperature Gas-Cooled Reactor (HTGR). A flowsheet remains to be demonstrated for the million gallons of 2nd and 3rd cycle raffinates and PEW evaporator concentrate which has accumulated (see Table 4). The latter is a high-level but low-heat producing waste. The major problem in calcining this waste is its high Na^+ content which results in agglomeration of the bed by NaNO_3 . The same problem is experienced in calcining neutralized Purex waste. Additives to form high-melting sodium compounds show promise in maintaining fluidization of the bed. The stainless steel nitrate waste from electrolytic dissolution of EBR fuel is blended with aluminum waste for liquid storage, and based on pilot-plant studies the blend can be readily calcined in the WCF.

Pilot-plant work has been performed for the Savannah River Laboratory and for Battelle's PNL to develop fluidized-bed process flowsheets for solidifying neutralized AEC waste and for acid Purex waste. A Purex waste, partially neutralized and containing 2M Na^+ (type PW-6), will require an additive to prevent agglomeration in the fluidized bed, a problem already referred to in the calcination of ICPP second- and third-cycle waste, and one which is encountered with neutralized AEC waste containing 6M Na^+ . A number of additives have been tested in the laboratory and pilot plant, but a satisfactory flowsheet has not been demonstrated. The acid Purex waste studies have direct application to waste from commercial fuel reprocessing plants. In fact, results on simulated acid Purex waste (type PW-4b) indicate satisfactory operation in a 6-in.-diameter pilot-plant fluidized-bed calciner. The properties of the solid calcine are summarized in Table 10.

TABLE 10.
PROPERTIES OF CALCINED SIMULATED PW - 4b WASTE

Volume reduction factor under continuous operation	30
Bulk density of bed	2.2 gms/cc
Bulk density of fines	1.2 gms/cc
Product-to-fines ratio	2.5
Mass median particle diameter	0.30 mm
Attrition resistance index	20-30
Structure (X-ray diffraction)	Amorphous

In spite of the excellent production record of the WCF, it is showing evidence of wear, and replacement is inevitable. Furthermore, the anticipated fuel reprocessing rates, combined with a limited capacity to store the resulting high-level liquid waste, will require greater processing rates than presently available from the WCF. The corrosion of equipment—due largely to fluorides for which the WCF was not designed—has lead to greater deposition of radionuclides, particularly ^{106}Ru , which cannot be sufficiently decontaminated to avoid radiation exposures to maintenance personnel commensurate with current safety regulations. Consequently, a New Waste Calcining Facility has been proposed for the ICPP for installation by the end of 1978. The net throughput will be approximately 3,000 gal/day. The plant will embody the major features of the WCF with upgrading of materials of construction, increased remote maintenance capability, and improved cleanup of the process off-gas.

IX. CONCLUSIONS

High-level radioactive waste is produced at the ICPP during the recovery of spent highly-enriched nuclear fuels. The management of the high-level waste is performed in accordance with the latest requirements and regulations for such waste. Liquid waste is stored safely in doubly-contained tanks made of stainless steel. The liquid waste is calcined to a solid and stored safely in a retrievable form in doubly-contained underground bins. The calcine can be treated further or left untreated in anticipation of ultimate storage. Fluidized-bed calcination has been applied to many kinds of high-level waste. The environmental impact of high-level waste management at the ICPP has been negligible and should continue to be negligible.

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