

**TECHNICAL DATA SUMMARY
SMALL DISSOLVER FOR THE
MULTIPURPOSE PROCESSING FACILITY**

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October 1976

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SUMMARY

Conversion of the existing recycle precipitator (EP 10-19-5) in the multipurpose processing facility (MPPF) to dissolver service has been demonstrated in full-scale prototype equipment at TNX. This system would allow processing of ^{252}Cf in the MPPF without expenditure of the capital cost (~\$250,000) required to complete the installation of the larger dissolver (17.3). One californium target tube can be charged into the modified 10-19-5 precipitator and dissolved at a slow rate to easily meet the current demand for ^{252}Cf . The dissolver processing rate is limited by subsequent process steps of: solids settling, supernate decantation, filtration, and washing of the solids in the dissolver rather than by the chemical solution step. The predicted processing capacity of this equipment is two californium target tubes per month (about 40 mg of ^{252}Cf per month) at 50% attainment.

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TECHNICAL DATA SUMMARY

SMALL DISSOLVER FOR THE MULTIPURPOSE PROCESSING FACILITY

INTRODUCTION

Several alternatives were studied for modifying existing MPPF equipment for dissolving one californium target tube per month. This rate would exceed the current demand for ^{252}Cf without expenditure of the capital cost required to complete the installation of the larger MPPF 17.3 dissolver.

The recycle precipitator 10-19-5 can easily be converted to dissolver service by removing the existing agitator and installing a charging column on the agitator flange. No other MPPF vessel of sufficient capacity has an existing opening large enough to accept the charging column. The recycle precipitator is also most convenient for dissolver service because of availability, location, capacity, existing services, existing spare nozzles, and existing piping arrangement.

Reference 1 describes the recommended design for the larger 17.3 dissolver which is capable of dissolving up to four californium targets per batch; a scale-down and revision of that flowsheet to one californium target per batch was used for the demonstration of this small dissolver.

ALTERNATIVE MODES OF DISSOLUTION

The dissolution of aluminum in sodium hydroxide-sodium nitrate solution is exothermic, and ammonia is evolved. The dissolution rate is very rapid at the boiling temperature and must be controlled or limited so that the heat of reaction can be removed and so that the ammonia can be diluted below the explosive limit with air purge. Two alternative modes of controlling or limiting the dissolution rate are boiling and nonboiling dissolution.

Boiling Dissolution

The dissolution rate was controlled by limiting the addition rate of sodium hydroxide into the dissolver while boiling the dissolver solution. The dissolution proceeds after initiation

at approximately the same rate as the caustic addition. An internal reflux condenser is required to remove the heat of reaction, to condense the vapor from the boiling solution, and to cool the noncondensable offgases (mostly air purge) to less than 50°C temperature.

Nonboiling Dissolution

The dissolution rate was also controlled by cooling the dissolver solution well below the boiling point during dissolution to maintain an acceptably low dissolution rate. The existing cooling jacket on 10-19-5 can remove the heat of reaction and maintain a 40.5°C maximum dissolution temperature with all of the sodium hydroxide in the dissolver. At this temperature, one target tube can be dissolved in about 50 hr. The existing air purge to 10-19-5 is more than sufficient to dilute the ammonia at this slower dissolution rate. The equipment modifications are greatly simplified because a reflux condenser is not required, and thus the OGE line does not need to be modified. Another simplification is that the sodium hydroxide solution can be added batchwise into the dissolver without any need for limiting the addition rate. The nonboiling dissolution is the recommended mode of dissolution.

REQUIRED EQUIPMENT MODIFICATIONS

The following modifications are required for either mode of dissolution:

- Remove the existing agitator from 10-19-5, and replace it with a new charging column.
- Provide 1.0-scfm air sparge to the existing spare dip-leg in 10-19-5 for mixing.
- Shorten the existing supernate dip-leg by 2 in. to leave a 15-liter heel instead of the 1.5-liter heel in 10-19-5 when transferring out through the supernate dip-leg.

The above equipment modifications are adequate for non-boiling dissolution, but the following additional modifications are required for boiling dissolution:

- Provide a downdraft reflux condenser of sufficient capacity to condense the vapors from the boiling solution and to cool the noncondensable offgases (mostly air purge) to less than 50°C temperature; the condenser can be installed around the charging column.
- Blank off the existing offgas-exhaust (OGE) outlet from 10-19-5.

- Provide OGE jumper with a small air jet from the OGE outlet of condenser to the OGE header.
- Blank off the existing overflow line from 10-19-5.
- Provide cooling water supply and return jumpers from existing spare nozzles on Rack 5 to the new condenser.
- Provide 1.75-scfm air purge to existing spare nozzles on 10-19-5.
- Provide means of limiting the 23% NaOH addition rate to less than 0.46 lb/min.

TEST PROCEDURES AND RESULTS

General

A prototype recycle precipitator EP 10-19-5 was mocked up to full scale with a prototype filter EP 10-19-7, a prototype filtrate waste transfer tank EP 10-19-11, a prototype dissolved cake tank EP 10-19-9, and all associated air jets, valves, and piping. A prototype charging column with a downdraft reflux condenser was designed, fabricated, and installed on the agitator flange in place of the agitator. The prototype equipment mockup is shown in Figure 1.

The equipment performance was characterized by determining pressure drops, heat removal rates, heat transfer coefficients, and air inleakage rates using water, steam, and/or air. The measured data were fitted to equations by the least squares technique; statistical deviations of the data from the fitted equations were also determined. Simulated full-scale chemical tests demonstrated caustic dissolution of aluminum targets, solids settling, supernate decantation, filtration, solids washing, decontamination from impurities, and nitric acid dissolution of solids. The flowsheet used for the simulated chemical demonstrations is shown in Figure 2; the material balance for this flowsheet is shown in Table 1. The conceptual time cycle presented in Table 2 shows that a total of 180 hr is required per californium target dissolved; with this time cycle the processing capacity is two californium targets per month (about 40 mg of ^{252}Cf per month) at 50% attainment.

Equipment Characterization

Reflux Condenser - Charging Column

A prototype reflux condenser - charging column with a 5-in.-ID by 7-ft-long charging column was designed to bolt onto the 6-3/4-in.-ID agitator flange. The column will accept one Mark 18

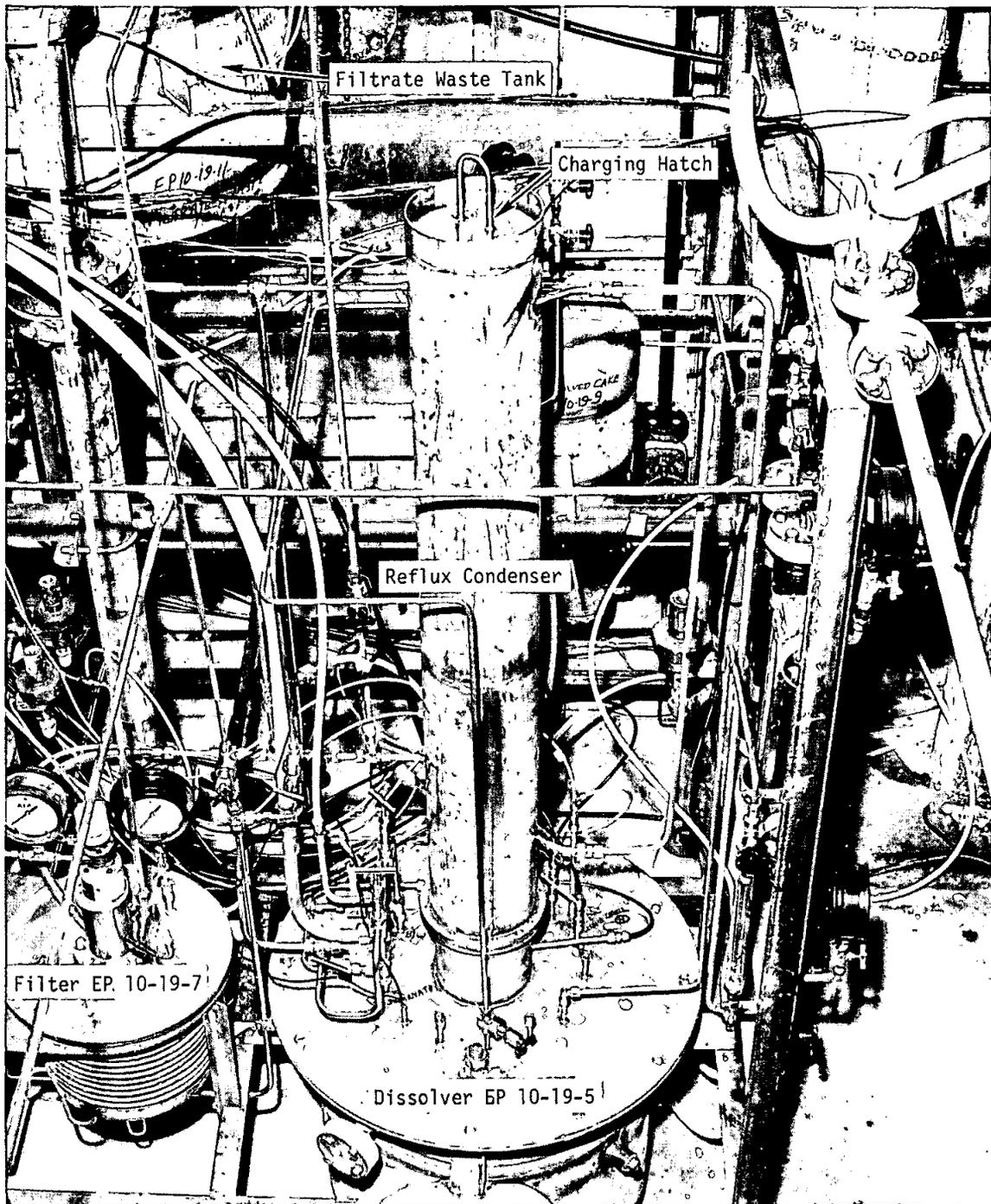
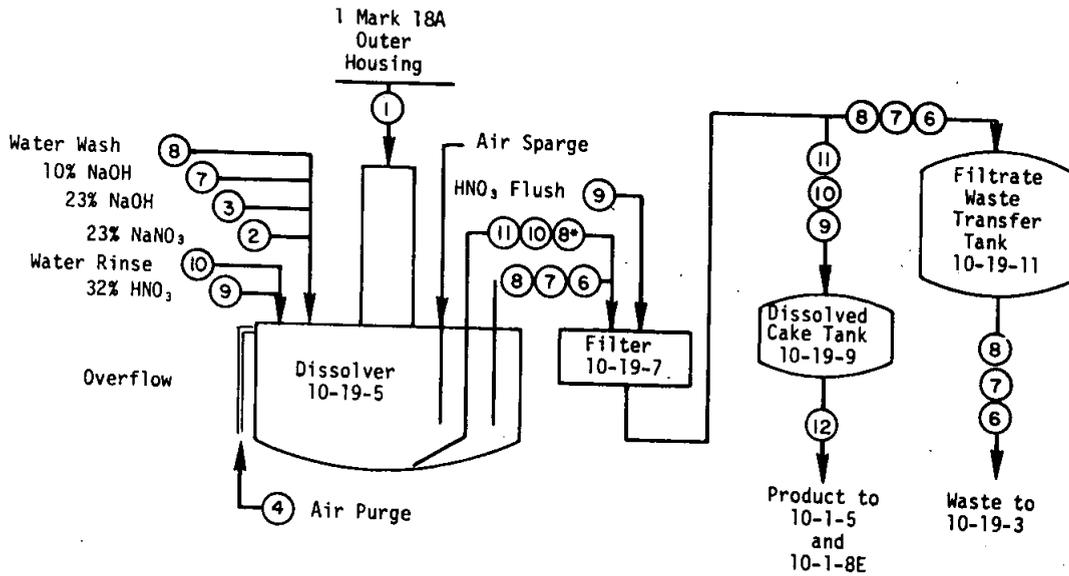


FIGURE 1. Prototype Dissolver and Filter Equipment



* 15-liter heel remaining after third water wash removed through long dip-leg.

FIGURE 2. Small MPPF Dissolver Flowsheet

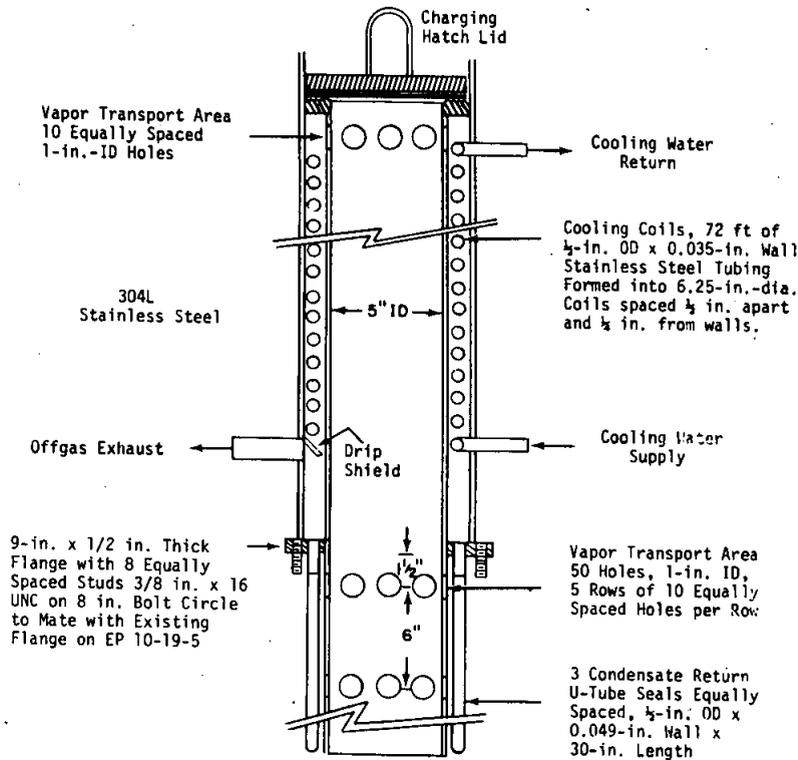


FIGURE 3. Reflux Condenser - Charging Column Design

TABLE 1

Material Balance for Small MPPF Dissolver Flowsheet

Line No.	1	2	3	4	5	6	7	8	9	10	11	12
Process Material	Mark 18A Outer Housing	23% NaNO ₃	23% NaOH	Air Purge	Aluminum Dissolution Offgas	Caustic Aluminum Waste	10% NaOH Washes ^a	Water Washes ^d	32% HNO ₃ Filter Flush	32% HNO ₃ Dissolution	Water Rinse	Composite Product Solution
Erbium, g ^a	126	-	-	-	-	-	-	-	-	-	-	266 ^e
Dysprosium, g ^a	126	-	-	-	-	-	-	-	-	-	-	270 ^e
Aluminum, lb	11.5	-	-	-	-	-	-	-	-	-	-	-
Silicon, lb ^b	0.046	-	-	-	-	0.046	-	-	-	-	-	-
Iron, lb ^b	0.023	-	-	-	-	-	-	-	-	-	-	0.077 ^e
Magnesium, lb ^b	0.053	-	-	-	-	-	-	-	-	-	-	0.32 ^b
NaNO ₃ , lb	-	38.6	-	-	-	17.6	-	-	-	-	-	-
NaOH, lb	-	-	40.3	-	-	28.4	58.6	-	-	-	-	-
NaNO ₂ , lb	-	-	-	-	-	8.2	-	-	-	-	-	-
NaAlO ₂ , lb	-	-	-	-	-	34.9	-	-	-	-	-	-
NH ₃ , lb	-	-	-	-	2.18	-	-	-	-	-	-	-
H ₂ , lb	-	-	-	-	0.0077	-	-	-	-	-	-	-
Air Purge, scfm	-	-	-	>1.5	>1.5	-	-	-	-	-	-	-
HNO ₃ , lb	-	-	-	-	-	-	-	-	3.4	12.6	-	15.0
H ₂ O, lb	-	129.1	134.9	-	>2.0	262	527	792	7.1	26.8	33.0	66.9
Total, lb	12.2	167.7	175.2	>23	>27.2	351.1	585.6 ^c	792 ^d	10.5	39.4	33.0	83.5
Solids, %	100	23	23	-	-	25	10	0	0	0	0	1.9
Specific Gravity	-	1.167	1.252	-	-	1.258	1.109	1.000	1.193	1.193	1.000	1.116
Volume, liters	-	65.3	63.6	-	-	127	240 ^c	360 ^d	4	15	15	34

a. Erbium and dysprosium added to Tests 3 and 4 to simulate actinide-lanthanide fraction.

b. Silicon, iron, and magnesium are major impurities in 6063A aluminum targets.

c. 10% NaOH washes divided into two 120-liter washes.

d. Water washes divided into three 120-liter washes.

e. Weights of nitrate salt of element.

TABLE 2

Conceptual Time Cycle for Flowsheet

<i>Description of Operation</i>	<i>Time, hr</i>
Charge target and chemicals	2
Caustic dissolution of aluminum	50
Settle solids	24
Filter supernate	12
Flush filter	1
First caustic wash of solids	6
Settle solids	12
Filter first caustic wash supernate	4
Flush filter	1
Second caustic wash of solids	6
Settle solids	12
Filter second caustic wash supernate	4
Flush filter	1
First water wash of solids	1
Settle solids	8
Filter first water wash supernate	2
Flush filter	1
Second water wash of solids	1
Settle solids	8
Filter second water wash supernate	2
Flush filter	1
Third water wash of solids	1
Settle solids	8
Filter third water wash supernate	2
Flush filter	1
Nitric acid dissolution of solids	6
Filter	1
Rinse dissolver	1
Filter dissolver rinse	1
Total	180

californium target tube (4.22-in. OD) up to 7 ft in length. The reflux condenser - charging column design is shown in Figure 3.

The reflux condenser coil contains 72 ft of 1/2-in. OD x 0.035-in. wall stainless steel tubing. The flow rate of cooling water through the condenser coil as a function of pressure drop at constant temperature is given by the following equation for a pressure drop range of 25 to 37.5 psi and a temperature range of 33.7 to 40.15°C with a deviation of ±0.0165 gal/min at 95% confidence from the measured data:

$$\text{Flow (in gal/min)} = 0.3092 (\Delta P^{0.5409}) (T^{0.084})$$

where ΔP = cooling water supply pressure less discharge pressure, psi

T = cooling water supply temperature, °C

The weight of the charging hatch lid (12.27 lb) provides adequate force for sealing against air inleakage and also provides pressurization relief above 0.62 psi. A neoprene gasket is attached to the lid to seal against multiple sharp-edge concentric grooves (1/32-in. by 45° angle) in the mating flange of the hatch. The air inleakage around the charging lid was measured at various OGE vacuums in the dissolver and is given by the following equation for a range of 0.5 to 50 in. of water dissolver vacuum with a deviation of ±0.038 scfm at 95% confidence from the measured data:

$$\text{scfm} = 0.1799 (\Delta P)^{0.4957}$$

where ΔP = pressure outside dissolver less pressure inside dissolver, in. of H₂O

The heat removal rate of the reflux condenser was tested by metering saturated steam into the dissolver through a limiting flow orifice at various regulated pressures. Various rates of air purge were also added to the dissolver during these tests. The cooling water inlet temperature to the condenser was maintained constant at 34.6 ±1.0°C during these tests. The maximum heat removal by the condenser was 134,300 Btu/hr. The maximum overall heat transfer coefficient was 272 Btu/(hr-ft²-°F) and the OGE temperature was 64.7°C when 1.5-scfm of air purge was added.

The overall heat transfer coefficient is given by the following equation for a heat removal range of 10.1 to 134.3 thousands of Btu/hr with a deviation of ±8.17 Btu/(hr-ft²-°F) from the measured data:

$$U_o = 55.86 Q^{0.01292} - 49.91$$

where U_o = overall heat transfer coefficient, Btu/(hr-ft²-°F)

Q = heat removal rate, thousands of Btu/hr

The vapor transport area between the 5-in.-ID vapor outlet column (19.6 in.² open area) and the column condensing coils is 7.85 in.² (ten 1-in.-ID holes). The vapor transport area between the dissolver pot and the vapor outlet column is a minimum of 7.85 in.² (ten 1-in.-ID holes) when the dissolver liquid level is less than about 27 in. of water. The area increases by 7.85 in.² for each interval of 6 in. lower liquid level for four additional rows of ten 1-in.-ID holes.

The pressure drop across the reflux condenser column is less than 0.1 in. of water for 3.0-scfm air purge rate and 70,000 Btu/hr heat removal rate. The column pressure drop as a function of heat removal rate at constant air purge rate is shown in Figure 4. The pressure drop across the reflux condenser for various air purge rates at ambient temperature is given by the following equation for a range of 0.1 to 13.7 scfm with a deviation of ±0.0467 in. of water ΔP from the measured data:

$$\Delta P \text{ (in in. of H}_2\text{O)} = 0.002696 (\text{scfm})^{2.403} + 0.0157$$

The OGE outlet of the condenser was connected to the 3-in. OGE header with 3 ft of 1-in.-OD x 0.065-in.-wall tubing. The tubing had two 90° smooth bends of 4-in. radius, one *Swagelok* (Registered tradename of Crawford Fitting Co.) tee fitting used as an elbow, and an entrance with a sudden enlargement into the 3-in. header. The total tubing length is about 12 equivalent ft. The pressure drop across this line from the condenser outlet into the 3-in. OGE header for various air purge rates at ambient temperature is given by the following equation for a range for 0.1 to 13.7 scfm with a deviation of ±0.127 in. of water ΔP from the measured data:

$$\Delta P \text{ (in in. of H}_2\text{O)} = 0.02636 (\text{scfm})^{1.381}$$

The OGE temperature at the outlet of the condenser was less than 45°C for heat removal rates of 133,600 Btu/hr when no air purge was added, but it increased to 50°C at heat removal rates as low as 11,500 Btu/hr where 1.5 scfm of air purge was added. The variation of OGE temperature with various air purge rates was very erratic (Figure 5). If the condenser heat load is maintained less than 30,000 Btu/hr and the air purge rate is maintained less than 3 scfm, the OGE temperature should be less than or near 50°C.

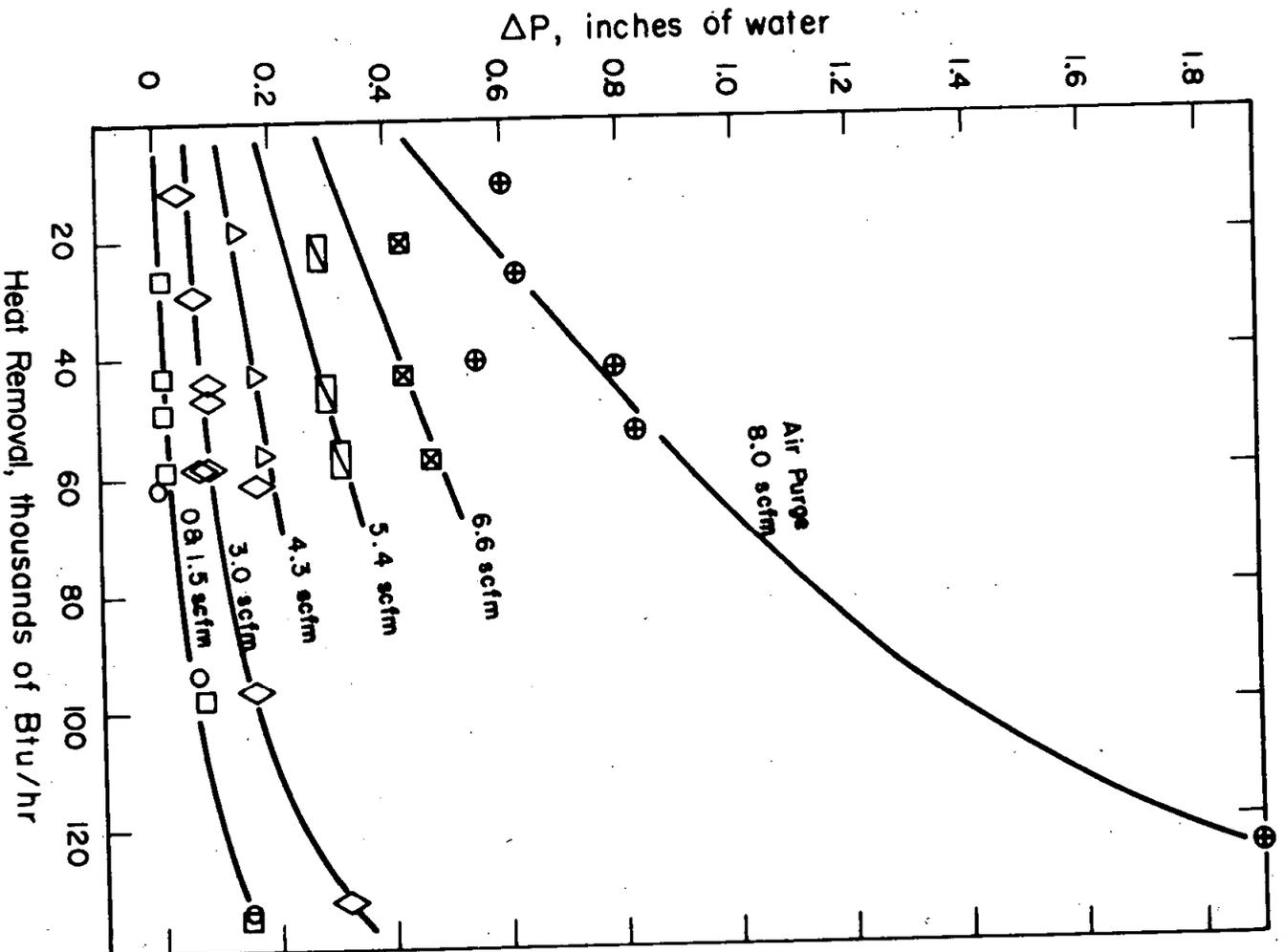


FIGURE 4. Reflux Condenser Pressure Drop

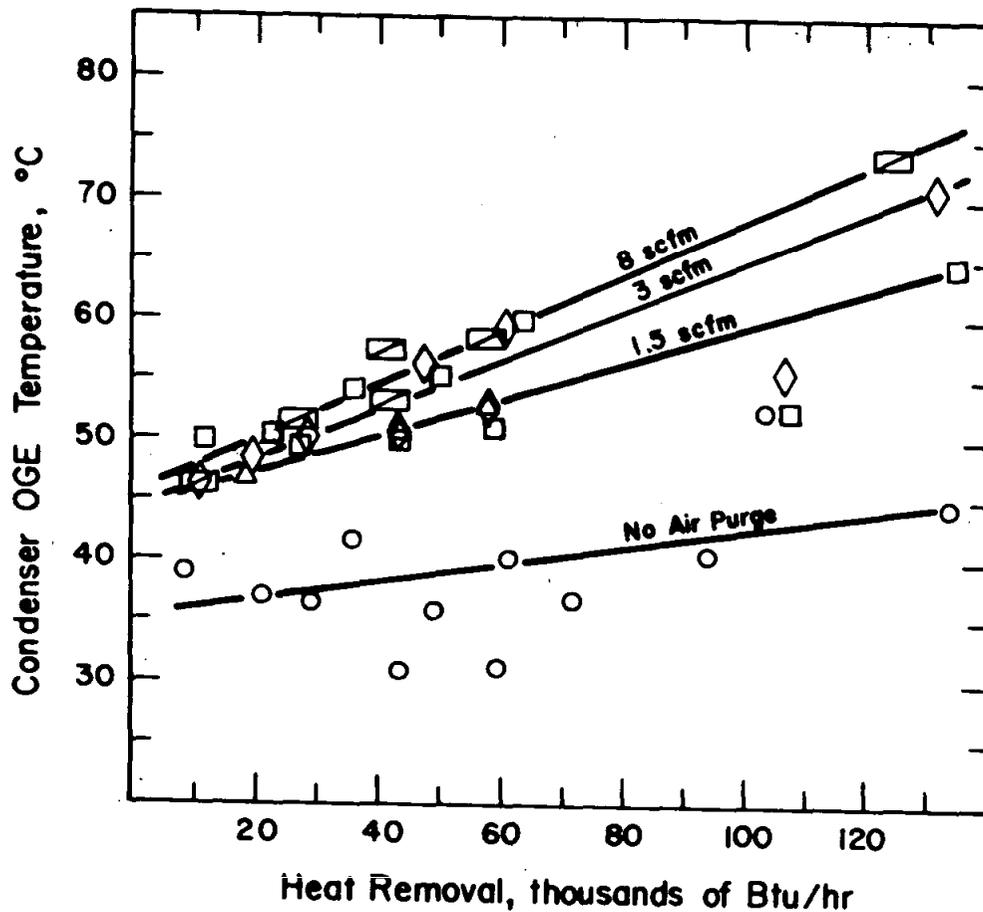


FIGURE 5. Reflux Condenser Performance

The air purge leaving the reflux condenser is saturated at the exit temperature with condensate vapor. This vapor will condense in the OGE system after cooling unless it is diluted below saturation with sufficient dry air. The condensable water vapor was calculated as 1.5 to 2.4 ml/scfm of air for air that is saturated at 50°C and cooled to 100 and 70°F, respectively. These values increase to 3.8 to 4.7 ml/scfm of air for air that is saturated at 60°C. The air purge rate and OGE exit temperature should be minimized to minimize this condensation unless sufficient dilution with dry air is possible. The volume ratio of dilution air at 70°F and 50% relative humidity required to dilute the air purge saturated at 50°C is 5.7 when cooled to 100°F, and 9.2 when cooled to 70°F.

Cooling Jacket

The maximum heat removal rate measured for the cooling jacket was 1480 ± 83 Btu/min when the pot temperature was $96.6 \pm 2.9^\circ\text{C}$ and the jacket cooling water supply was 4.5 gal/min flow at $35.6 \pm 0.6^\circ\text{C}$. The pot was agitated during this test with 1.0-scfm air sparge. The overall heat transfer coefficient calculated from the above test was 162 Btu/(hr-ft²-°F).

The heat removal rate for the cooling jacket decreases as the temperature of the pot contents decreases. The rate of temperature decrease was measured while the pot contents were cooling during a water demonstration and during the first three simulated chemical tests. The heat removal rate was calculated at various pot temperatures by assuming the specific heat of the pot contents to be 1.0 Btu/(lb-°F) divided by the specific gravity and then subtracting the heat loss. The heat removal rate of the cooling jacket is given by the following equation for a temperature range of 38 to 98°C with a deviation from the measured data of ± 124 Btu/min at 95% confidence.

$$\text{Btu/min} = 179.7 e^{0.0243T} - 19.04 e^{0.0253T} - 385.14$$

where T = temperature of pot contents, °C

The cooling water supply temperature averaged $35.3 \pm 0.6^\circ\text{C}$ during the above tests, and the flow rate averaged 4.3 ± 0.6 gal/min. The pot was also agitated during these tests with 1.0-scfm air sparge.

Heat Loss

The heat loss to the ambient air surrounding the dissolver pot is given by the following equation for a range of 16.9 to 74.8 Btu/min with a deviation from the measured data of ± 14.2 Btu/min at 95% confidence:

$$\text{Btu/min} = 19.04 e^{0.0253T} - 34.06$$

where T = temperature of pot contents, °C

The ambient air temperature surrounding the pot averaged $22.9 \pm 6.3^\circ\text{C}$ during the above test. The pot was agitated with 1.0-scfm air sparge during the above test.

Reaction Rate

Aluminum dissolution in sodium hydroxide generates a very large quantity of heat (11,644 Btu/lb) which must be removed. This heat of reaction is removed from boiling solution by the reflux condenser or from nonboiling solution by the cooling jacket. The dissolution rate, and thus the heat generation rate, increases with temperature up to the boiling point and also increases with caustic concentration. In boiling solution, the reaction rate is so rapid that a controlled caustic addition rate must be used to limit the reaction rate. The reaction rate at temperatures less than 70°C is much slower and can be controlled by cooling the dissolver solution with the cooling jacket to maintain the temperature less than 70°C during dissolution.

The heat evolution rate was determined during the fifth test by measuring the rate of temperature increase; the rate was calculated at various temperatures assuming the specific heat of the pot contents to be 1.0 Btu/(lb-°F) divided by specific gravity and then adding the heat loss. The heat evolution rate is given by the following equation for a temperature range of 31.4 to 68°C with a deviation of ±6.8 Btu/min at 95% confidence:

$$\text{Btu/min} = 5.81T - 164$$

where T = temperature of pot contents, °C

Above 70°C, the heat evolution rate increases rapidly to about 1600 Btu/min at 85°C.

A steady-state dissolution temperature of 40.4°C is predicted by setting the equation for rate of heat removal by the cooling jacket equal to the equation for the rate of heat generation by the reaction; the actual temperature measured during run 4 was 40.5°C.

Simulated Chemical Demonstrations

General

A total of five simulated full-scale chemical demonstrations were made by dissolving one dummy Mark 18 californium target tube each test. The dummy tubes were 7 ft long and contained only reactor grade 6063A aluminum. These tubes were dissolved in a full-scale prototype EP 10-19-5 recycle precipitator which had been modified by installing a charging column - reflux condenser on the agitator flange in place of the agitator. In addition, full-scale prototypes of the recycle filters EP 10-19-7, the filtrate waste tank EP 10-19-11, the dissolved cake tank EP 10-19-9, and all associated air jets, valves, and piping were mocked up with 10-19-5.

Solids settling, supernate decantation, filtration, washing of solids, and dissolution of solids were demonstrated for each test.

Caustic Dissolution of Aluminum

The first three dissolving demonstrations were made at or near boiling temperature by initially heating 65.3 liters of 23% sodium nitrate solution in the dissolver with hot water supply (85 to 91°C) to the jacket before beginning the controlled addition of 63.6 liters of 23% sodium hydroxide. The dissolution solution was heated above about 97°C by the heat of reaction, and then about 3000 to 7000 Btu/hr of the heat of reaction was removed by the hot water jacket when 85 to 90°C water was supplied. Further heat of reaction was removed through the reflux condenser cooling water. The sodium hydroxide addition rate was controlled in an attempt to limit the maximum reaction rate and thus limit the maximum heat removal rate and OGE exit temperature required of the reflux condenser.

The heat removed by the reflux condenser, the dissolution solution temperature, and the OGE exit temperature from the reflux condenser are shown in Figures 6, 7, and 8 for the first three dissolution demonstrations. The OGE exit temperature from the reflux condenser was above the 50°C guideline for about 0.3, 1.3, and 0.6 hr and reached maximum temperatures of 54, 59.5, and 60.1°C, respectively, in the three runs as can be seen in the above figures. These results agree with the condenser performance tests (Figure 4), which indicated that the condenser heat removal rate must be maintained less than 30,000 Btu/hr and that the air purge must be maintained less than 3 scfm to maintain the OGE temperature less than or near 50°C.

The addition rates of 23% sodium hydroxide used in these three demonstrations (0.21, 0.37, and 0.14 l/min) were relatively ineffective in limiting the maximum reaction rate and heat load. This is attributed to the inability to initially heat the dissolver solution to boiling before starting addition to the sodium hydroxide solution. The dissolver solution was heated to 83 to 87°C with hot water supply temperatures of 86 to 91°C to the dissolver jacket. The addition rate of sodium hydroxide solution exceeds the reaction rate initially while the temperature is less than about 90°C. These conditions allow the excess sodium hydroxide concentration to increase in the dissolver.

The heat of reaction increases the dissolver solution temperature to the initiation temperature, and the reaction rate then increases exponentially until the temperature either reaches boiling or steady state near boiling. The reaction rate is then

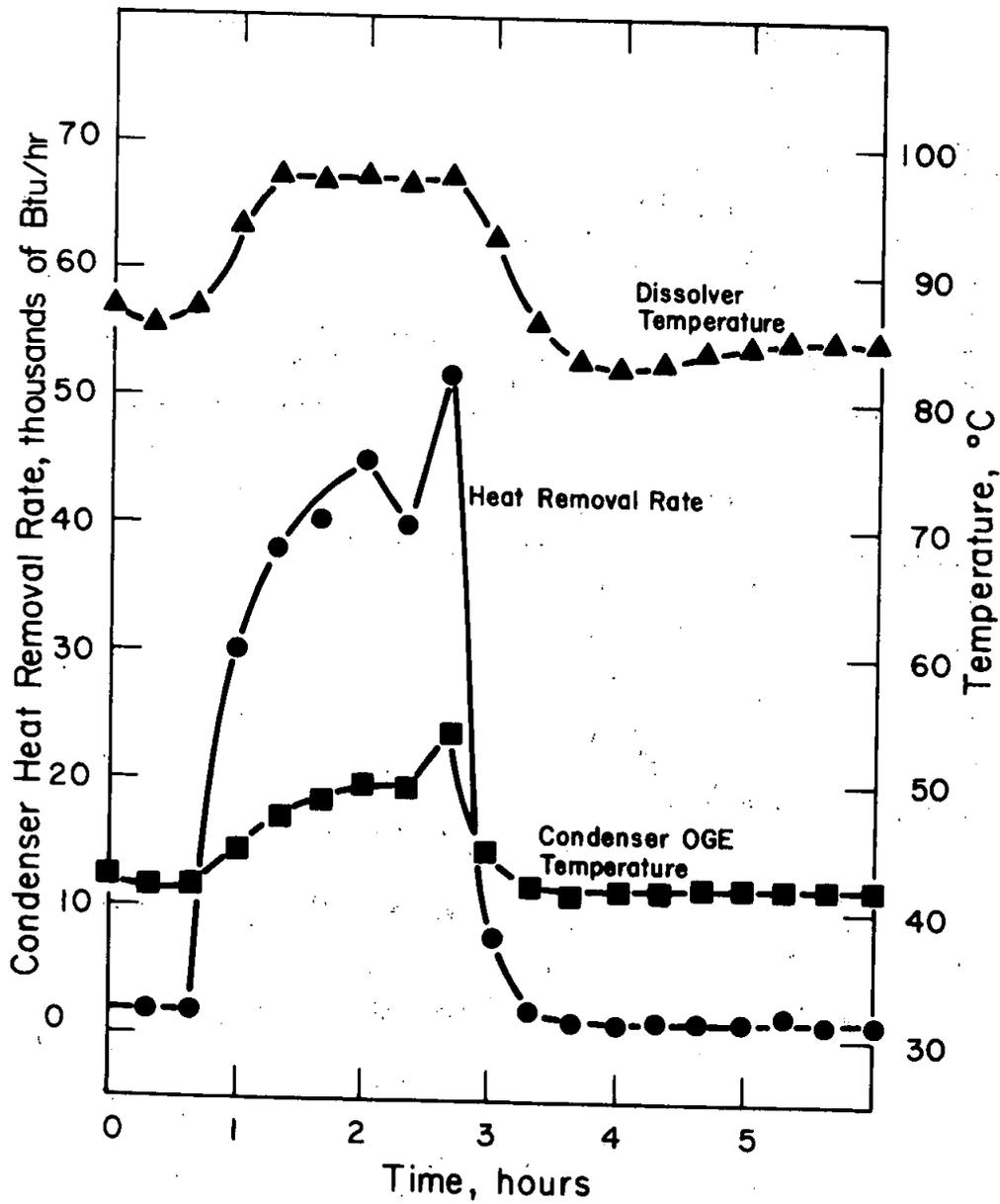


FIGURE 6. Test 1 Dissolution

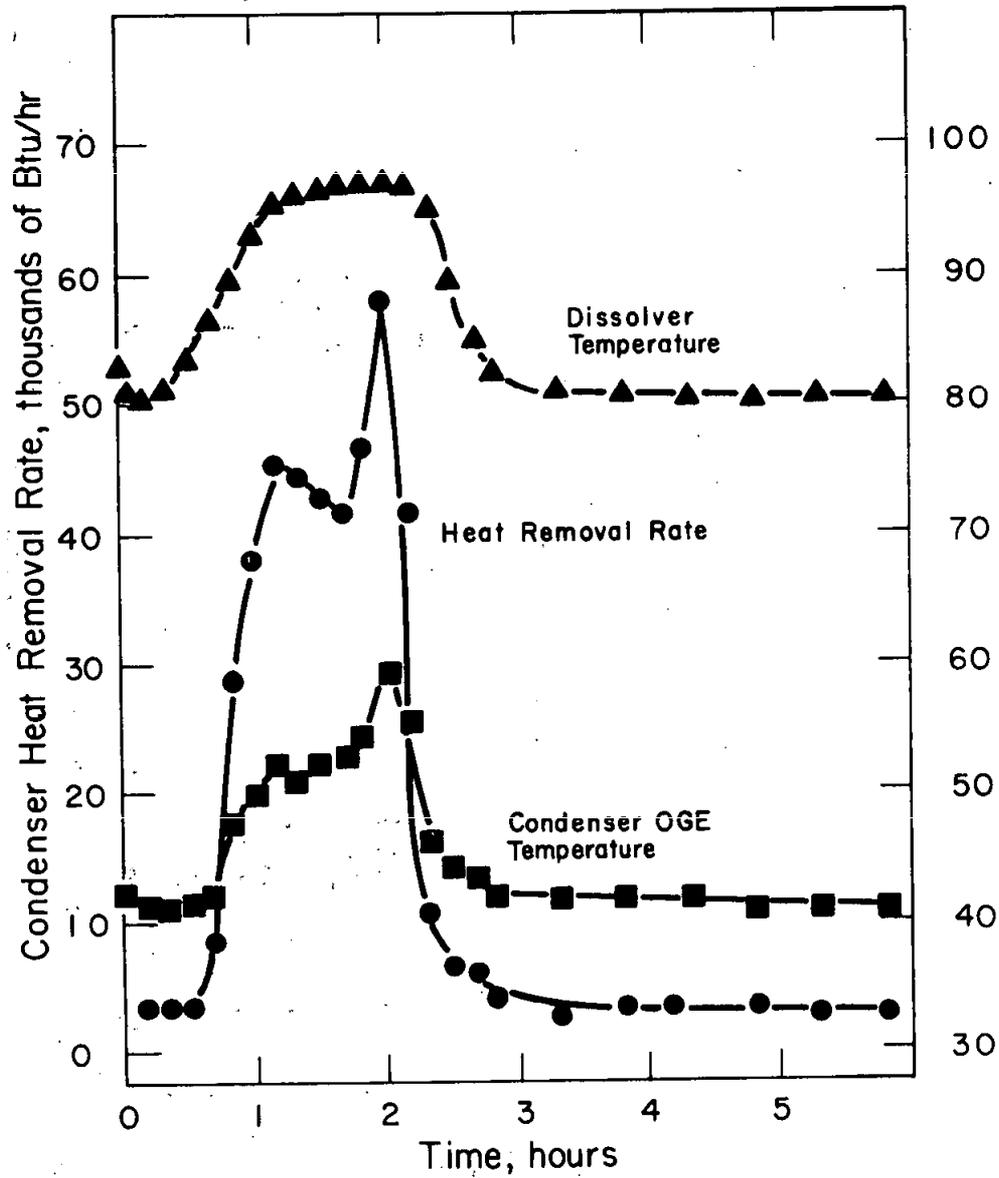


FIGURE 7. Test 2 Dissolution

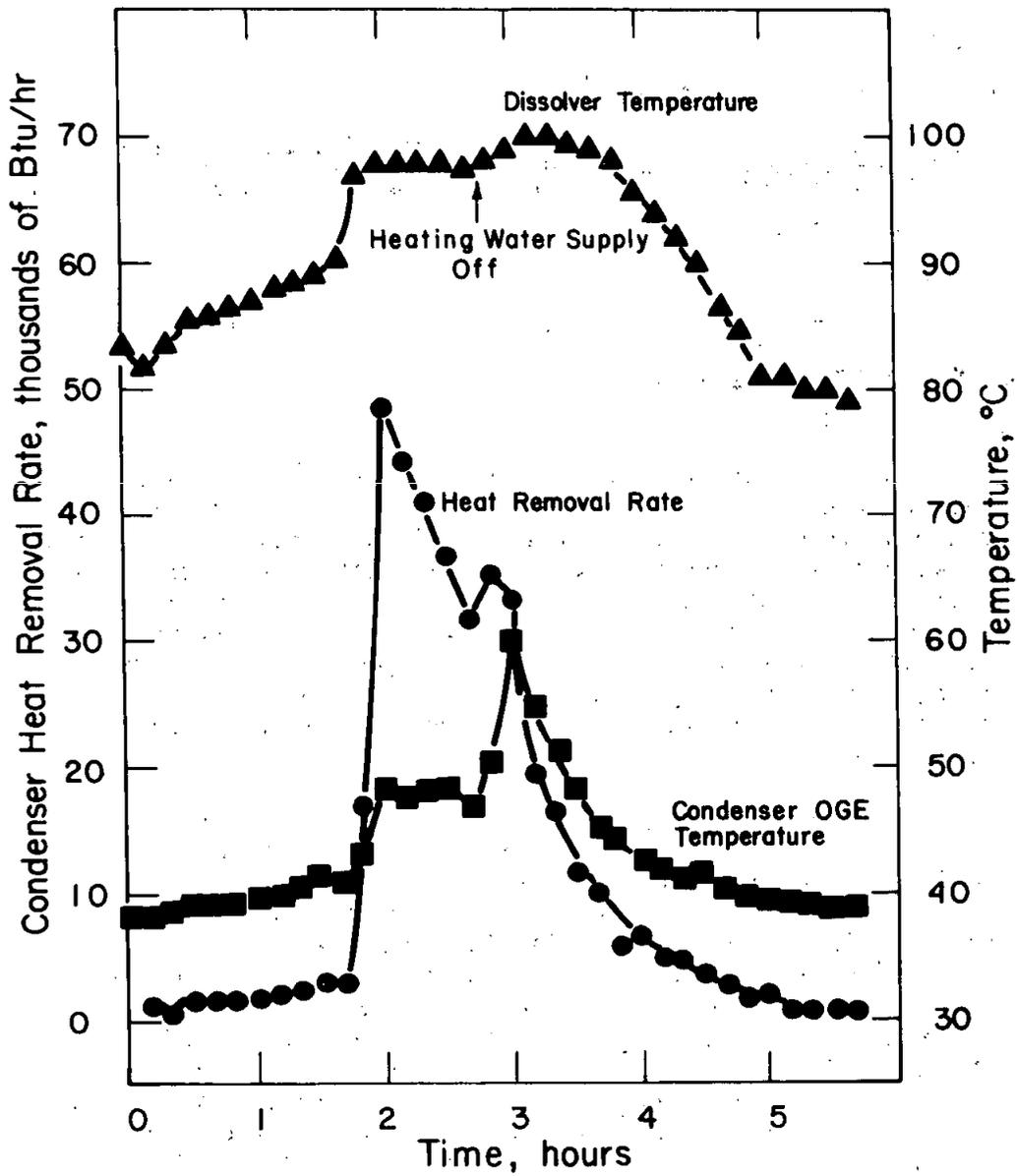


Figure 8. Test 3 Dissolution

controlled by the excess sodium hydroxide concentration buildup in the dissolver rather than by the addition rate. The data before the first three demonstrations are summarized in Table 3.

TABLE 3

Conditions Before Tests 1 Through 3

Test	Temperature, °C			Initiation Time, hr	23% NaOH Added Before Initiation		
	Starting	Minimum	Initiation		Total Volume, liters	Average Rate, liters/min	Excess NaOH, mol/liter
1	87	85.5	87	0.65	8.4	0.22	<0.8
2	83	81.0	83.5	0.5	12.0	0.40	<1.1
3	83.3	81.7	90.3	1.65	20.4	0.16 ^a	<1.7

a. First 5 liters added in 5 min.

The reaction rate can be better controlled by either maintaining the minimum dissolver temperature above the initiation temperature during the controlled sodium hydroxide addition or by keeping the dissolver temperature constant throughout dissolution. The hot water temperature would have to be increased to greater than or equal to 95°C, which is probably undesirable, to maintain the dissolver at greater than or equal to 90°C. If the dissolver temperature is controlled constant below 70°C throughout dissolution, the sodium hydroxide rate need not be controlled.

The fourth dissolving demonstration was made without boiling the solution. About half of the aluminum target tube was dissolved in 20 hr by cooling the dissolver solution to a maximum temperature of 40.5°C with the cooling jacket. The maximum OGE exit temperature was 30.7°C during the dissolution. The cooling water supply to the dissolver cooling jacket was then turned off to allow the heat of reaction to increase the dissolver solution temperature while the remaining portion of the aluminum target tube was dissolving. The dissolver solution increased slowly to a maximum of 74°C in 6.3 hr and then began to decrease, indicating that dissolution of the aluminum was almost complete. The maximum OGE exit temperature was 37.8°C during the above dissolving process. Controlling the reaction rate by controlling the dissolver solution temperature is far superior to controlling the sodium hydroxide addition rate for slow rates of dissolution.

The fifth dissolution demonstration was made to determine adverse effects from loss of cooling water supply during dissolution. 63.6 liters of 23% sodium hydroxide solution was added batchwise to the demonstration solution; the addition time was

3 min. The dissolution temperature was immediately increased 4°C by the addition of the sodium hydroxide because of the heat of dilution. The cooling water supply was purposely left off, and the dissolver solution was allowed to be self-heated by the heat of reaction caused by the dissolution of aluminum. The dissolver solution was agitated with 1 scfm of air sparge, and air purge was pulled into the dissolver through the overflow line by an OGE header vacuum of 0.35 in. of water. Vapor began coming out of the dissolver overflow after 3.7 hr of dissolving without cooling water supply; the dissolver had just exceeded 70°C at this time. The dissolver temperature increased rapidly from 70 to 100°C; at this time, the dissolver pressure became sufficient (0.62 psi) to lift the dissolver charging hatch lid and expel vapors from the charging hatch. The cooling water supply to the dissolver jacket was started after the dissolver temperature exceeded 100°C; the dissolver solution was cooled to 90°C in twenty minutes and then the cooling water supply was turned off again. The temperature did not further increase in the dissolution solution; thus, the dissolution was about complete. The cooling capacity of the dissolver cooling jacket, however, is insufficient to reduce the dissolver temperature if the reaction temperature is allowed to exceed 70°C when sufficient undissolved aluminum is present in the dissolver. Above 70°C, the heat evolution rate of the dissolution reaction increases rapidly and exceeds the cooling capacity of the jacket; for example, the heat evolution rate at 85°C is about 1600 Btu/min while the jacket cooling capacity is only 870 Btu/min.

A total of 2.1 liters of dissolver solution was collected from the dissolver overflow during this demonstration, and 4.9 liters of condensate was collected from the OGE system. Analysis of the condensate for aluminum and nitrate indicated less than 0.5% entrainment of dissolver solution into the condensate during the demonstration.

No adverse effects result from loss of cooling water supply during dissolving if the cooling water is restarted before the dissolver solution temperature exceeds 70°C or if more than one-half of the aluminum has been dissolved when the cooling water is stopped. Otherwise, the adverse effects are overflow of a small amount of dissolver solution into the sump and entrance of a small amount of condensate into the OGE system.

The dissolving data for the five demonstrations are summarized in Table 4.

Impurities in Aluminum Target Tubes

Samples of the aluminum target tubes dissolved in Tests 1 and 3 were analyzed for impurities to verify that the aluminum

TABLE 4

Summary of Dissolving Data

	Test 1	Test 2	Test 3	Test 4	Test 5
Aluminum Weight, g	5221	5226	5199	5235	5221
Erbium Weight, g	-	-	126	126	-
Dysprosium Weight, g	-	-	126	126	-
Air Sparge Rate, scfm	1	1	1	1	1
Air Purge Rate, scfm	3	4	1½	4	3
Total Air Rate, scfm	4	5	2½	5	4
Avg. NaOH Addition Rate, l/min	0.21	0.37	0.14	0.36	21.2
Total NaOH Addition Time, hr	4.0	2.9	5.7	2.9	0.05
Dissolving Time, hr	~2.7	~2.3	~3.0	~26	~4.2
Digestion Time, hr	4	4	2	-	-
Pot-to-Column ΔP, in H ₂ O	0.10-0.23	0.23-0.55	0.04-0.60	0.21-0.35	0.1>1.0
Column-to-OGE ΔP, in H ₂ O	0.15-0.27	0.28-0.60	0.10-0.20	0.20-0.43	0.1-0.4
Avg. OGE Temp., °C	43.4	45.0	42.9	30.7	33.3
Max. OGE Temp., °C	54	59.5	60.1	37.8	86.9 ^a
Max. Reflux Condenser Cold Water Return Temp., °C	55	60	53.7	35	-
Reflux Condenser Cold Water Supply Rate, gpm	2.9	2.7	2.8	2.8	-
Humidity in OGE, liters	1.01	1.69	0.6	1.42	4.9
Humidity in OGE, l/scfm air	0.25	0.34	0.24	0.28	-
Jacket Hot Water Supply Rate, gpm	4.3	4.2	4.2	-	-
Jacket Hot Water Supply Temp., °C	91	85	90	-	-
Time to Heat from 30 to 80°C, min.	34	48	44	-	-
Jacket Cold Water Supply Rate, gpm	4.5	4.5	4.5	4.5	0 ^a
Jacket Cold Water Supply Temp., °C	35	35	35.5	35.5	34
Time to Cool from 80 to 45°C, min.	55	53	72	-	-
Time to Cool from 45 to 40°C, min.	30	33	57	-	-

a. Cooling water supply to the dissolver jacket was started after the dissolver temperature exceeded 100°C and was continued for 20 min until dissolver temperature was cooled to 90°C.

was reactor grade 6063A (Table 5). The maximum expected error in these analyses is +100% to -50%. The aluminum target tubes dissolved in Tests 1 through 4 were all fabricated from the same batch of aluminum.

The impurities, copper, iron, and magnesium, are present in sufficient quantities to form a copious volume of insoluble solids which follow the actinide-lanthanide solids and must be settled, decanted, washed, filtered, and finally dissolved in nitric acid. These impurities will have to be separated later in the MPPF process from the actinide product fraction.

TABLE 5

Impurities in Aluminum

<i>Element</i>	<i>Test 1</i>	<i>Test 3</i>
Ag	<1 ppm ^a	<1 ppm ^a
B	<2 ppm ^a	<2 ppm
Be	<1 ppm ^a	<1 ppm ^a
Bi	<20 ppm ^a	<20 ppm ^a
Cd	<1 ppm ^a	<1 ppm ^a
Co	<5 ppm ^a	<5 ppm ^a
Cr	<20 ppm	<20 ppm
Cu	0.03%	<0.02%
Fe	0.22%	0.17%
Mg	0.38%	0.54%
Mn	35 ppm	20 ppm
Mo	<5 ppm ^a	<5 ppm ^a
Nb	<10 ppm ^a	<10 ppm ^a
Ni	10 ppm	75 ppm
Pb	<1 ppm	2 ppm
Sb	<10 ppm ^a	<10 ppm ^a
Si	0.43%	0.36%
Sn	<1 ppm ^a	<1 ppm
Ti	50 ppm	100 ppm
V	<20 ppm	100 ppm
W	<50 ppm ^a	<50 ppm ^a
Zn	<10 ppm	10 ppm
Zr	<10 ppm	<10 ppm ^a

a. Element not detected.

The silica impurity is slightly soluble in hot sodium hydroxide solution and can be partially removed by sufficient washes. Although no difficulties were encountered because of silica solids in these demonstrations, filtration of full-level irradiated targets will probably require additional washes because silica grows in by transmutation reaction $^{27}\text{Al}(n,\gamma)^{28}\text{Si}$ during irradiation and will be several times more (up to ~2.8% Si) than present in the unirradiated aluminum.

Settled Solids

The settled volume of solids was measured at various times for many samples taken from the full-scale demonstrations (Tests 3 and 4) and also for two laboratory-scale one-liter size dissolvings (Figures 9, 10, and 11). The settled volume of solids was generally 5 to 10% of the solution volume after 0.1-day settling time with the exception of one full-scale sample and one laboratory-scale dissolving which required 1 to 2 days time to settle to 10%.

The volume of settled solids continued to decrease very slowly as an exponential function of time after an initial settling time of about 0.1 day. The percent volume of settled solids after the initial settling time can be expressed as bt^n where the value of the exponent n is -0.06 for dissolver solution, -0.14 for 10% sodium hydroxide washes, and -0.08 for water washes. The value of the constant b is lower for Test 4 than for Test 3 indicating a lower bulk volume of solids and thus denser, more-compact settled solids. The values of the constant b are shown below for the two tests:

	<i>Test 3</i>	<i>Test 4</i>
Dissolver solution	7.1	6.0
10% NaOH washer	$8.4 < b < 11.2$	$4.5 < b < 5.1$
Water washes	$5.3 < b < 8.9$	$4.3 < b < 6.3$

The denser, more-compact volume of settled solids for Test 4 was also verified by comparing the ratio of the gravity settled volume of solids to centrifuged volume of solids in a sample. The ratio of settled volume to centrifuged volume was lower for Test 4 than for Test 3 as shown below:

	<i>Test 3</i>	<i>Test 4</i>
Minimum ratio	2.8	2.6
Average ratio	4.0	3.3
Maximum ratio	5.7	4.0

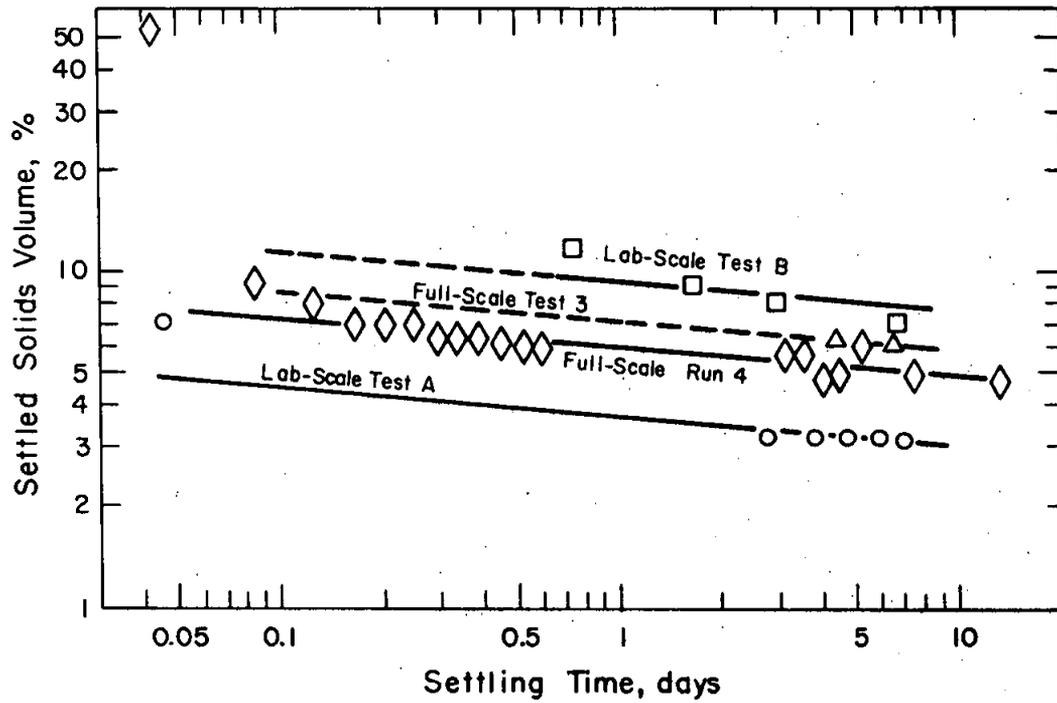


FIGURE 9. Solids Settling in Dissolver Solution

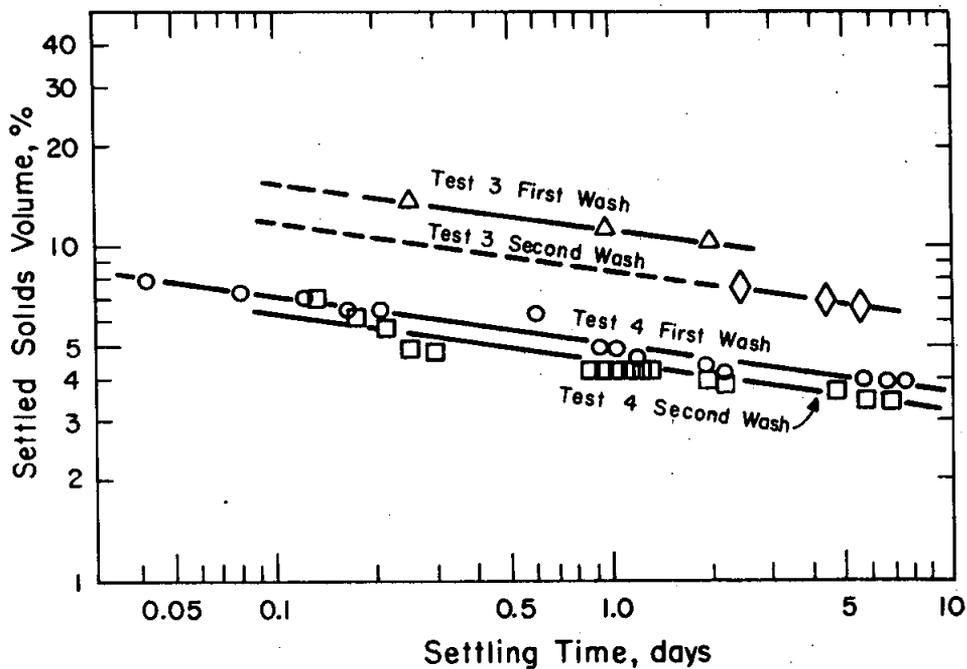


FIGURE 10. Solids Settling in 10% NaOH Washes

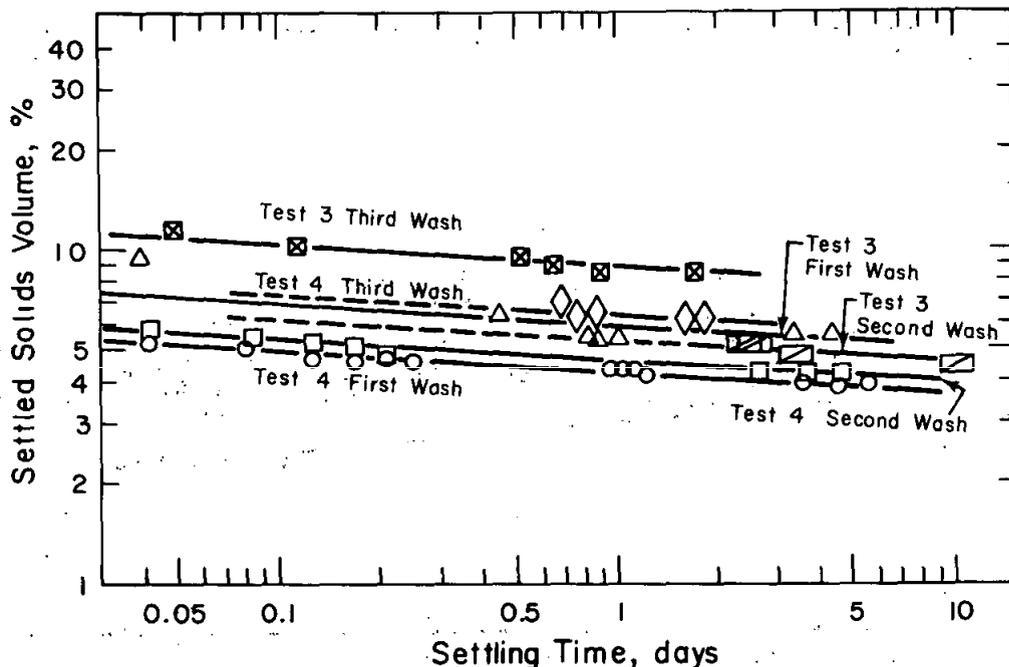


FIGURE 11. Solids Settling in Water Washes

Air flow to the liquid level and to the specific gravity bubblers in 10-19-5 must be turned off during settling, decanting, and filtration to prevent stirring the solids.

Decanting

The bottom of the existing supernate dip-tube is positioned about $1\frac{1}{2}$ in. from the bottom of EP 10-19-5; this leaves a level of about 1.3 in. or a volume heel of about 3.0 liters when transferring out through the supernate dip-tube. In Tests 1 and 2, the supernate dip-tube was at that position and excessively long filtrations occurred that required repeated flushings of the filter. Because the volume of settled solids is generally about 6.5 to 13 liters, no decanting of supernate was obtained for Tests 1 and 2 because about 54 to 77% of the settled solids volume was above the bottom of the supernate dip-tube.

The supernate dip-tube was shortened to about 3.6 in. off the bottom; this leaves a level of about 3.4 in. or a heel volume of about 15.3 liters when transferring out through the supernate dip-leg. In Tests 3 and 4, the shortened supernate dip-tube greatly improved the filtrations and reduced the frequency of filter flushings required.

Filtration

The solids were allowed to settle generally for one or more days, and then the supernate was decanted through the supernate dip-tube and through the prototype filter EP 10-19-7 by pulling >25 in. Hg vacuum on the prototype filtrate waste transfer tank EP 10-19-11. The vacuum was generated on the filtrate waste transfer tank on 10-19-11 by an AV-116 air jet EP 10-100-2.² The liquid level bubbler in Dissolver 10-19-5 was turned off after the initial filtration on Test 1 to prevent agitating the settled solids. The filtration rate could not be monitored after that time. An average filtration rate was obtained periodically by stopping the filtration long enough to drain and measure the volume of filtrate collected over a given period of time. This collection period of time was infrequent on Test 1 and hourly on Tests 2, 3, and 4. The completion of filtration is generally signaled by a vacuum break unless the filter is almost totally plugged. The vacuum break is a rather sudden decrease in vacuum caused by entrance of air into vacuum source after filtration of all liquid is complete; all other conditions remained constant. The magnitude and rate of vacuum break is used as an indication of filter pluggage. The filter agitator was run at about 200 rpm during all filtrations. The completion of filtration was generally signaled also by an increase in agitator speed as the liquid level in the filter decreased below the agitator blade.

The accumulated volume filtered as a function of time in the initial part of Test 1 can be represented by the following equation:

$$V = 31.61 t^{0.1343}$$

where V = volume, liters for $0 < V < 43.8$

t = time, hr for $0 < t < 13.67$

The instantaneous filtration rate can be represented as a function of time for the initial part of Test 1, as shown below:

$$\frac{dV}{dt} = 4.245 t^{-0.8657}$$

where $\frac{dV}{dt}$ = instantaneous filtration rate, liters/hr

t = time, hr for $0 < t < 13.67$

The above filtration was terminated by valving off flow from 10-19-5 and venting the filter; about 0.6 liter more filtrate was obtained from the filter after an extended period of evacuation.

Physical examination of the filter cake showed about 1/16-in.-thick dark brown cake. The filter cake was rinsed twice with one liter of 10% sodium hydroxide solution. The first liter of rinse filtered through the cake in 52 seconds for an average rate of 1.2 l/min. The second liter of rinse filtered through in 20 sec after scraping some of the cake from the filter surface. The above rinses were followed by a one-liter water rinse which filtered through at an average rate of 5.2 l/min. The filter was flushed with 2 liters of 70% nitric acid for 2 hr at 84.5°C to dissolve the filter cake, then cooled to 50°C, and filtered through in 70 sec time; the volume of acid flush filtrate collected was 2.85 liters. The above flush was followed first by a 4-liter rinse of prefiltered deionized water which filtered through at an average rate of 2.3 l/min and then by second rinse of 17 liters of prefiltered deionized water which filtered through at an average rate of 3.2 l/min. The filter agitator was run at 200 rpm during all flushing, rinsing, and filtration.

The air flow to the 10-19-5 liquid level and specific gravity bubblers was valved off and the solids allowed to settle for several days before completing the filtration. The remaining 73.5 liters was then filtered in 6 hr for an average filtration rate of 12.25 l/hr. The heel of solids and liquid remaining in the dissolver was about 4 liters. The filter was flushed with 2.0 liters of 70% nitric acid for 2 hr to dissolve the filter cake then filtered through the filter in 46 min. The filter flush was followed by two 2-liter rinses of prefiltered deionized water which filtered through at an average rate of 0.96 and 1.5 l/min. The filter was flushed with 4 liters of 50% sodium hydroxide solution for 2 hr at >50°C to dissolve any silica pluggage which may have accumulated on the filter from the previous filtration. The 50% sodium hydroxide flush filtered through at an average rate of 1.4 l/min and was followed by one 4-liter and one 16-liter prefiltered deionized water rinse which filtered through at an average rate of 6 l/min and 4.7 l/min, respectively.

The 10% sodium hydroxide wash of the solid-liquid heel remaining in 10-19-5 was allowed to settle for several days then was filtered at an average rate greater than 5.8 l/hr. There was no vacuum break across filter at completion of filtration to indicate the time of completion. The filter was so plugged that a water rinse could not be filtered through, so the water rinse was removed and the filter was unplugged with a one-liter flush of 70% nitric acid for 5 min followed by a 6-liter prefiltered deionized water rinse. The remaining test program for Test 1 was terminated.

The dissolver, filter, and other equipment were thoroughly flushed after the first test to remove all traces of solids.

before starting the second test. The final flush of 120 liters of prefiltered deionized water was filtered in 30 min 56 sec at an average rate of 3.8 l/min, and was followed by a vacuum break to 11 in. Hg in 1 min 52 sec.

Another aluminum target tube was dissolved for the second test, then the solids were allowed to settle undisturbed for 4.6 days before starting the filtration. An hourly average filtration rate was obtained by stopping the filtration long enough to drain and measure the volume of filtrate collected after each hour of filtration (Table 6).

TABLE 6

Test 2 Filtration of Dissolver Solution
(First Part)

<i>Time, hr</i>	<i>Filtration Rate, l/hr</i>
1	20.5
2	5.0
3	0.04
4	0.06
5	0.02
6	0.01

The above data show that the filter was severely plugged after the first 2 hr of filtration and remained plugged through the next 4 hr. The filter was unplugged by flushing with 4 liters of 60% nitric acid at 33°C temperature for 15 min. The acid flush was filtered through at 2.8 l/min; a vacuum break to 6-in. Hg in 1.7 min followed. Then 12 liters of prefiltered deionized water was filtered through at 4.7 l/min; a vacuum break to 6-in. Hg in 0.6 min followed.

The filtration of dissolver solution from the second test was continued (Table 7).

TABLE 7

Test 2 Filtration of Dissolver Solution
(Second Part)

<i>Time, hr</i>	<i>Filtration Rate, l/hr</i>
1	79.0
2	3.5
3	1.2
4	0.04
5	0.00
6	0.02

The above data show that the filter was severely plugged after 3 hr of filtration and remained plugged through the next 3 hr. The filter was unplugged by flushing with 4 liters of 60% nitric acid at 30°C temperature for 15 min. The acid flush was filtered through at 2.3 l/min; a vacuum break to 7-in. Hg in 0.7 min followed. Then 12 liters of prefiltered deionized water was filtered through at 4.8 l/min; a vacuum break to 6-in. Hg in 0.7 min followed.

The filtration of dissolver solution from the second test was continued (Table 8).

TABLE 8

Test 2 Filtration of Dissolver Solution
(Third Part)

<i>Time, hr</i>	<i>Filtration Rate, l/hr</i>
1	2.5
2	2.9
3	1.05
4	0.35
5	0.09

The above data show that the dissolver was severely plugged after 4 hr of filtration. The filter was unplugged by flushing with 4 liters of 60% nitric acid for 15 min followed by 12 liters of prefiltered deionized water which filtered at 4.6 l/min; a vacuum break to 6-in. Hg in 0.7 min followed.

The filtration of the dissolver solution from the second test was completed with the collection of the final 0.13 liter of filtrate followed by a vacuum break to 11-in. Hg in 7.7 min. The filter was examined visually and found to have ~1/16-in.-thick dark brown cake of solids. The filter was flushed alternately with nitric acid, sodium hydroxide, and prefiltered deionized water to remove all traces of solids and to verify that the filter was clean before starting the next filtration (Table 9).

The filter pluggage during the filtering of the prefiltered deionized water flush after the first 10% sodium hydroxide flush is attributed to the reprecipitation of silica solids from the residual silica dissolved by the preceding sodium hydroxide flush. An additional sodium hydroxide flush to reduce and dilute the residual silica left in the filter before the water flush possibly could have prevented this pluggage. Further flushing did not increase the filtration rate to the previous rate, the vacuum did not break to as low a level, and the time required for vacuum break was longer.

TABLE 9

Test 2 Flushes and Flow Tests After
Filtration of Dissolver Solution

<i>Flush Volume, liters</i>	<i>Composition, %</i>	<i>Time, min.</i>	<i>Temp., °C</i>	<i>Filtration Rate, l/min</i>	<i>Vacuum Break</i>	
					<i>Vacuum, in. Hg</i>	<i>Time, min.</i>
4	60% HNO ₃	15	29	2.2	7	0.8
12	Deionized Water	5	29	4.6	7	0.5
12	10% NaOH	15	31	3.4	7	0.7
12	Deionized Water	5	30	~1	No Break	>15
4	60% HNO ₃	15	33	2.0	11	2.9
12	Deionized Water	5	32	2.4	14	2.7
4	60% HNO ₃	15	30	1.9	12	3.6
12	Deionized Water	5	29	3.0	13	2.5
12	10% NaOH	15	29	2.4	13	2.3
12	Deionized Water	5	30	3.1	12	3.5

The dissolver heel of liquid and solids (3.7 liters) was washed with 120 liters of 10% NaOH for 4 hr at 86°C. The solids were allowed to settle for 2.6 days before starting filtration of the wash solution. The hourly average filtration rate of the wash is shown for each hour of filtration in Table 10.

TABLE 10

Test 2 Filtration of 10% NaOH Wash

<i>Time, hr</i>	<i>Filtration Rate, l/hr</i>
1	104.5
2	11.8
3	0.23
4	0.03
5	0.01
6	0.00
7	0.00

The above data show that the filter was severely plugged after 3 hr and remained plugged. The filter was unplugged by flushing with 4 liters of 70% nitric acid at 29°C for 15 min.

The acid flush filtered through at 3.4 l/min; a vacuum break to 6-in. Hg in 0.7 min followed. The acid flush was followed by 12 liters of prefiltered deionized water which filtered through at 4.8 l/min; a vacuum break to 5-in. Hg in 1.5 min followed. The filtration of the 10% sodium hydroxide wash was completed with the collection of the final 0.2 liter of filtrate followed by a vacuum break to 10-in. Hg in 3.6 min. The filter was flushed with 4 liters of 70% nitric acid which filtered at 3.4 l/min; a vacuum break to 6-in. Hg in 0.7 min followed. A 12-liter prefiltered deionized water flush filtered at 4.8 l/min; a vacuum break to 5-in. Hg in 1.6 min followed.

The dissolver heel of liquid and solids was washed with three deionized water washes. The settling time, filtration rate, and vacuum break data for each wash is shown in Table 11.

TABLE 11

Filtration of Deionized Water Washes

Water Wash	Settling Time, days	Filtration Rate, l/hr	Vacuum Break	
			Vacuum, in. Hg	Time, min.
First	0.44	199	20	1.3
Second	0.63	208	20	9.7
Third	None ^a	116	No Break	>180

a. Filtered through long dip-leg with liquid level and specific gravity bubblers on.

The filter was flushed after filtering each of the water washes; the data are shown in Table 12.

TABLE 12

Test 2 Flushes and Flow Tests After Filtration of Deionized Water Washes

Flush After Water Wash	Volume, liters	Composition, %	Time, min.	Temp., °C	Filtration Rate, l/min	Vacuum Break	
						Vacuum, in. Hg	Time, min.
1	4	70% HNO ₃	15	32	2.4	6	0.7
1	12	Deionized Water	5	31	4.8	6	0.6
2	4	70% HNO ₃	15	32	3.4	5	1.5
2	12	Deionized Water	5	32	4.8	5	0.9
3	4	70% HNO ₃	15	31	2.6	8	3.6
3	12	Deionized Water	5	32	3.9	14	2.3
3	4	70% HNO ₃	15	32	4.8	7	2.2
3	12	Deionized Water	5	33	3.8	8	0.8

The dissolver pot was washed three times with 20 liters of 32% nitric acid at 85°C temperature for 4 hr to dissolve solids remaining in the dissolver. These solutions were cooled and filtered through the long dip-leg (Table 13).

TABLE 13

Test 2 Filtration of Nitric Acid Washes

Nitric Acid Wash	Temp., °C	Filtration Rate, l/min	Vacuum Break	
			Vacuum, in. Hg	Time, min.
First	34	2.16	13	1.5
Second	26	-	10	1.1
Third	34	3.1	10	2.3

Several flushes and flow tests of the filter were made after the first and second nitric acid washes (Table 14).

TABLE 14

Test 2 Flushes and Flow Tests After Filtration of Nitric Acid Washes

Flush After Wash	Volume, liters	Composition, %	Time, min.	Temp., °C	Filtration Rate, l/min	Vacuum Break	
						Vacuum, in. Hg	Time, min.
1	4	Deionized Water	15	33	>2.0	13	>1.8
1	12	Deionized Water	5	33	2.9	18	1.8
1	4	70% HNO ₃	15	33	3.1	6	0.7
1	12	Deionized Water	5	32	4.2	9	0.7
2	4	Deionized Water	15	30	>2.6	10	>1
2	12	Deionized Water	5	31	4.4	12	2.3
2	4	70% HNO ₃	15	32	2.5	6	1.0
2	12	Deionized Water	5	31	4.5	7	0.5

The supernate dip-leg was shortened to about 3.6 in. off the bottom of Dissolver 10-19-5 before Tests 3 and 4; this leaves a level of about 3.4 in. or a heel volume of about 15.3 liters when transferring out through the supernate dip-leg. The shortened dip-leg allowed decanting of the supernate from above the level of settled solids and greatly improved the filtrations and reduced the frequency of filter flushing required.

Nonradioactive erbium and dysprosium oxide solids were added into the dissolver along with the aluminum target tube on both Tests 3 and 4 to simulate the actinide-lanthanide solids which would be present in irradiated targets.

The filtration of dissolver solution from third run was started after allowing the solids to settle for 0.57 day (Table 15).

TABLE 15

Test 3 Dissolver Solution Filtration
(First Part)

<i>Time, hr</i>	<i>Filtration Rate, l/hr</i>
1	35.0
2	19.0
3	8.4
4	5.9
5	2.6
6	0.0

The above data show that the filter was plugged after 5 hr of filtration. The filter was unplugged and flow tested with the following flushes before continuing the filtration of dissolver solution (Table 16).

TABLE 16

Test 3 Flushes and Flow Tests After Filtration
of First Part of Dissolver Solution

<i>Flush</i>		<i>Filtration</i>		<i>Vacuum Break</i>		
<i>Volume, liters</i>	<i>Composition, %</i>	<i>Time, min.</i>	<i>Temp., °C</i>	<i>Rate, l/min</i>	<i>Vacuum, in. Hg</i>	<i>Time, min.</i>
4	70% HNO ₃	15	29	-	7	1.2
12	Deionized Water	5	27	-	15	1.2
4	70% HNO ₃	15	26	2.3	7	0.8
12	Deionized Water	5	26	4.5	7	0.9

The filtration of the remaining Test 3 dissolver solution was completed in 0.41 hr at an average filtration rate of 92.5 l/hr after allowing the solids to settle for 3.2 days. The vacuum break following the filtration was to 17-in. Hg in 3.1 min. Even though the filter was not plugged, it was flushed and flow tested to verify that it was completely clean before starting the next filtration of the 10% sodium hydroxide wash of the dissolver solid-liquid heel. The flush and flow test data are given in Table 17.

TABLE 17

Test 3 Flush and Flow Tests After Filtration
of Second Part of Dissolver Solution

<i>Flush</i>				<i>Filtration</i>		<i>Vacuum Break</i>	
<i>Volume,</i> <i>liters</i>	<i>Composition,</i> <i>%</i>	<i>Time,</i> <i>min.</i>	<i>Temp.,</i> <i>°C</i>	<i>Temp.,</i> <i>°C</i>	<i>Rate,</i> <i>l/min</i>	<i>Vacuum,</i> <i>in. Hg</i>	<i>Time,</i> <i>min.</i>
4	32% HNO ₃	60	80	36	3.5	5	1.2
12	Deionized Water	5	36	36	4.8	7	0.5
4	70% HNO ₃	15	34	34	2.9	7	0.6
12	Deionized Water	5	33	33	4.7	7	0.5

The filtration of the 10% sodium hydroxide wash of Test 3 was started after allowing the solids to settle for 2.6 days; the data are shown in Table 18.

TABLE 18

Test 3 Filtration of First 10% NaOH Wash

<i>Time, hr</i>	<i>Filtration Rate, l/hr</i>
1	64.0
2	52.0
3	0.06

No vacuum break was detected at the end of the above filtration, thus indicating the filter was plugged. The filter was flushed and flow tested as shown in Table 19.

TABLE 19

Test 3 Flush and Flow Tests After
Filtration of First 10% NaOH Wash

<i>Flush</i>				<i>Filtration</i>		<i>Vacuum Break</i>	
<i>Volume,</i> <i>liters</i>	<i>Composition,</i> <i>%</i>	<i>Time,</i> <i>min.</i>	<i>Temp.,</i> <i>°C</i>	<i>Temp.,</i> <i>°C</i>	<i>Rate,</i> <i>l/min</i>	<i>Vacuum,</i> <i>in. Hg</i>	<i>Time,</i> <i>min.</i>
4	32% HNO ₃	60	80	39	4.2	6	0.8
12	Deionized Water	5	32	32	4.8	5	0.5
4	70% HNO ₃	15	31	31	3.4	6	0.7
12	Deionized Water	5	29	29	4.8	5	0.7

The dissolver solid-liquid heel was washed with a second 10% sodium hydroxide wash and the solids allowed to settle for 2.0 days before starting filtration as shown in Table 20.

TABLE 20

Test 3 Filtration of Second 10% NaOH Wash

<i>Time, hr</i>	<i>Filtration Rate, l/hr</i>
1	79.0
2	36.0
3	0.06

No vacuum break was detected at the end of the above filtration indicating the filter was plugged. The filter was flushed and flow tested as shown in Table 21.

TABLE 21

Test 3 Flush and Flow Tests After Filtration of Second 10% NaOH Wash

<i>Flush</i>				<i>Filtration</i>		<i>Vacuum Break</i>	
<i>Volume, liters</i>	<i>Composition, %</i>	<i>Time, min.</i>	<i>Temp., °C</i>	<i>Temp., °C</i>	<i>Rate, l/min</i>	<i>Vacuum, in. Hg</i>	<i>Time, min.</i>
4	32% HNO ₃	60	82	39	4.0	6	0.9
12	Deionized Water	5	34	34	4.8	5	0.9
4	70% HNO ₃	15	32	32	2.8	6	0.8
12	Deionized Water	5	30	30	4.8	5	0.8

The dissolver heel of liquid and solids was washed with three deionized water washes. The settling time, filtration rate, and vacuum break for each wash is shown in Table 22.

TABLE 22

Test 3 Filtration of Deionized Water Washes

<i>Water Wash</i>	<i>Settling Time, days</i>	<i>Filtration Rate, l/hr</i>	<i>Vacuum Break</i>	
			<i>Vacuum, in. Hg</i>	<i>Time, min.</i>
First	0.46	183	20	4.5
Second	2.46	195	16	2.0
Third	0.52	225	18	3.1
Heel ^a	None	13.5	No Break	60

a. Solids and liquid heel remaining after third wash were mixed with air sparge then filtered through long dip-leg.

The filter was flushed after filtering the first and second deionized water washes and then after filtration of the heel of liquid and solids remaining after the third deionized water wash (Table 23).

TABLE 23

Test 3 Flush and Flow Tests After Filtration of Deionized Water Washes and Solids Heel

<u>Flush After Wash</u>	<u>Flush</u>				<u>Filtration</u>		<u>Vacuum Break</u>	
	<u>Volume, liters</u>	<u>Composition, %</u>	<u>Time, min.</u>	<u>Temp., °C</u>	<u>Temp., °C</u>	<u>Rate, l/min</u>	<u>Vacuum, in. Hg</u>	<u>Time, min.</u>
1	4	32% HNO ₃	60	80	30	4.1	5	1.0
1	12	Deionized Water	5	29	29	4.7	6	0.6
1	4	70% HNO ₃	15	30	30	2.6	6	0.7
1	12	Deionized Water	5	29	29	4.7	6	0.6
2	4	32% HNO ₃	60	81	33	3.7	4	-
2	12	Deionized Water	5	36	36	4.8	6	0.6
2	4	70% HNO ₃	15	37	37	2.9	6	1.9
2	12	Deionized Water	5	31	31	4.8	6	0.6
3+Heel	4	32% HNO ₃	60	84	37	2.7	12	1.1
3+Heel	12	Deionized Water	5	32	32	2.8	-	-
3+Heel	4	32% HNO ₃	60	81	38	2.4	11	1.1
3+Heel	12	Deionized Water	5	32	32	3.1	16	1.3
3+Heel	4	70% HNO ₃	15	24	24	1.4	10	1.0
3+Heel	12	Deionized Water	5	24	24	2.6	16	5.2

The solids remaining in the dissolver were dissolved and removed with the following nitric acid and water washes (Table 24).

TABLE 24

Test 3 Filtration of Solids Dissolution

<u>Dissolver Wash</u>				<u>Filtration</u>		<u>Vacuum Break</u>	
<u>Volume, liters</u>	<u>Composition, %</u>	<u>Time, hr</u>	<u>Temp., °C</u>	<u>Temp., °C</u>	<u>Rate, l/min</u>	<u>Vacuum, in. Hg</u>	<u>Time, min.</u>
20	40% HNO ₃	4	81	24	1.4	23	3.5
30	Deionized Water	-	28	28	3.4	9	1.1
20	32% HNO ₃	4	81	30	2.8	8	1.1

The filter was flushed after the 40% nitric acid wash and after the 32% nitric acid (Table 25).

TABLE 25

Test 3 Filter Flush and Flow Test After Filtration
of 40% HNO₃ Wash and 32% HNO₃ Wash

Volume, liters	Composition, %	Time, min.	Temp., °C	Filtration		Vacuum Break	
				Temp., °C	Rate, l/min	Vacuum, in. Hg	Time, min.
4 ^a	32% HNO ₃	60	81	37	3.8	10	1.1
12	Deionized Water	5	35	35	4.5	13	3.0
4	70% HNO ₃	60	77	38	3.4	9	1.6
12	Deionized Water	5	30	30	4.6	7	0.5
4 ^b	32% HNO ₃	60	79	39	3.3	7	0.6
12	Deionized Water	5	33	33	3.7	8	0.8
4	70% HNO ₃	15	34	34	3.2	7	0.6
12	Deionized Water	5	30	30	4.6	6	0.7

a. After 40% HNO₃ wash

b. After 32% HNO₃ wash

The aluminum target tube was dissolved for Test 4 using the nonboiling mode previously described; the maximum dissolver temperature reached during dissolution was 74°C. The solids were allowed to settle for 10 days before starting filtration. The temperature of the dissolver solution during filtration was 20°C. The filtration data are shown in Table 26.

TABLE 26

Test 4 Filtration of Dissolver Solution

Time, hr	Hourly Average Filtration Rate, l/hr
1	40.0
2	16.5
3	8.5
4	9.8
5	9.8
6	11.4
7	6.0
8	0.17
9	0.18

The accumulated volume filtered in the above filtration can be represented as a function of time:

$$V = 40t^{0.4709}$$

where V = Volume, liters for $0 < V < 102$

t = time, hr for $0 < t < 7$

The instantaneous filtration rate for the above filtration can then be represented as a function of time:

$$\frac{dV}{dt} = 18.84t^{-0.5291}$$

where $\frac{dV}{dt}$ = instantaneous filtration rate, l/hr

t = time, hr for $0 < t < 7$

There was no vacuum break after the above filtration, and a heel of 22.1 liters of liquid and solids remained in the dissolver. The filter was flushed with 4 liters of 32% nitric acid at 76°C for 1 hr and was followed by 12 liters of prefiltered, deionized water. The filtration rate of the acid flush was 3.2 l/min and was followed by a vacuum break to 6-in. Hg in 1 min. The water flush filtered at 3.7 l/min and was followed by a vacuum break to 6-in. Hg in 1 min.

Two 10% sodium hydroxide washes were made of the dissolver heel of solids and liquid as shown in Table 27.

TABLE 27

Test 4 Filtration of 10% NaOH Washes

Wash	Settling Time, days	Temp., °C	Filtration Rate, Liters/hr		Dissolver Heel Volume, liters	Vacuum Break	
			First	Second		in. Hg	Time, min.
First	0.63	25	80	52.5	15.5	21	8.3
Second	0.64	25	59.5	71.4	15.2	22	1.4

No filter flushes were made after either of the 10% sodium hydroxide washes.

Three deionized water washes were made of the dissolver heel of solids and liquid as shown in Table 28.

TABLE 28

Test 4 Filtration of Deionized Water Washes

Wash	Settling Time, days	Temp., °C	Filtration Rate, liters/hr		Vacuum Break	
			First	Second	Vacuum, in. Hg	Time, min.
First	0.69	23	62	75	22	4.5
Second	2.6	22	63	50.5	22	-
Third	0.63	22	63	74.3	21	4.3
Heel	None	20	77.1	-	22	4.6

No filter flushes were made between the above washes. After the filtration of the dissolver heel of solids and liquid, the cake was dissolved from the filter with 4 liters of 32% nitric acid at 77°C for 4 hr. The dissolved cake solution filtered at 4.7 l/min and was followed by a vacuum break to 8-in. Hg in 1.9 min. The cake dissolution was followed by a 16-liter pre-filtered deionized water rinse which filtered at 2.4 l/min and was followed by a vacuum break to 19-in. Hg in 15.3 min.

The solids remaining in the dissolver were dissolved with 20 liters of 32% nitric acid at 80°C for 4 hr. The nitric acid solution filtered at 2.6 l/min and was followed by a vacuum break to 15-in. Hg in 9.9 min.

Washing of Solids

The purpose of washing the solids is to remove impurities which are soluble in sodium hydroxide solution such as aluminum and silica and to remove the excess sodium hydroxide also. Impurities which are insoluble in the sodium hydroxide solution, such as iron, magnesium, and copper, will remain with the actinide-lanthanide solids and will have to be separated later in other MPPF processes.

Aluminum is soluble in sodium hydroxide solution if sufficient excess hydroxide is present, but it may precipitate if there is insufficient excess hydroxide. Precipitated aluminum may be difficult to redissolve; for this reason, the first wash should contain at least 10% sodium hydroxide. Higher concentrations can be used, but filtration rates may be slower because of increased viscosity.

Silica may be present in the dissolver solution as both soluble and as very fine solids which are at least partially removed by filtration; this was confirmed by analysis of several samples of Test 2 dissolver solution and filtrate shown in Table 29.

TABLE 29

Analysis of Test 2 Dissolver Solution and Filtrate

<u>Dissolver Solution</u>		<u>Filtrate</u>	
<i>Sample</i>	<i>Silicon Analysis, g/l</i>	<i>Sample</i>	<i>Silicon Analysis, g/l</i>
1278	0.1794	1284	0.0898
1279	0.1794	1285	0.0862
1280	0.1794	1286	0.0862
<u>Avg</u>	<u>0.1794</u>	1287	0.0840
		1288	0.0848
		<u>1289</u>	<u>0.0862</u>
		<u>Avg</u>	<u>0.0862</u>

Irradiated aluminum targets have been shown to have about seven times as much silica present; therefore, the fraction of silica present as solids in irradiated dissolver solution will be much greater. The silica solids can be dissolved with strong hot caustic washes. Samples were taken and analyzed of the filtrate from the 10% sodium hydroxide wash of the dissolver heel of solids and liquid remaining after filtration of the dissolver solution. The wash was heated to 85°C for 4 hr and then cooled to 30°C before filtration. The silicon analysis of Test 2 wash filtrate is shown in Table 30.

TABLE 30

10% NaOH Wash Filtrate

<i>Sample</i>	<i>Silicon Analysis, g/l</i>
1345	0.111
1346	0.118
1347	0.118
<u>Avg</u>	<u>0.116</u>

Filters were flushed with 10% sodium hydroxide to remove silica from the filter after filtration of Test 2 dissolver solution and after filtration of the 10% sodium hydroxide wash. The sodium hydroxide flushes were followed by deionized water flushes. The flush filtrates were analyzed for silicon and hydroxide; the flush data are shown in Table 31.

TABLE 31

Analysis of Flush Filtrates

<i>Filter Flushes</i>				<i>Analysis</i>		
<i>Volume,</i> <i>liters</i>	<i>Composition,</i> <i>%</i>	<i>Temp.,</i> <i>°C</i>	<i>Time,</i> <i>min.</i>	<i>Sample</i> <i>Number</i>	<i>Silicon,</i> <i>g/l</i>	<i>[OH],</i> <i>mol/l</i>
12	10% NaOH	30.5	15	1335	0.1189	3.08
12	Deionized Water	30	5	1336	0.0216	0.03
12	10% NaOH	28.5	15	1343	0.1499	3.11
12	Deionized Water	30	5	1344	0.0153	0.04

The heel of solids and liquid remaining in dissolver after decanting through the supernate dip-leg should be washed at least twice with sufficient volume of 10% sodium hydroxide solution to dilute the aluminum and silicon concentrations sufficiently to prevent precipitation when washing with deionized water later to remove the excess sodium hydroxide. The sodium hydroxide washes should be followed with three deionized water washes to remove the sodium hydroxide. The dilution factor of the soluble impurities can be calculated from the following equation:

$$D.F. = \left[\frac{W + H}{H} \right]^n$$

where D.F. = dilution factor

W = volume of wash added, liters

H = volume of solution left in heel, liters

n = number of washes

The above dilution factor of impurities is limited by the occlusion of impurity-bearing solution in the gelatinous type solids present.

Two 10% sodium hydroxide washes and three deionized washes of about 120 liters each were made on solid-liquid heels from Tests 3 and 4 which were normally about 15-liter volume. The following decontamination factors for aluminum and sodium were determined by dividing the quantity of each impurity in the original dissolver solution by the quantity remaining in the nitric acid dissolution of the product fraction:

Impurity	Decontamination Factors	
	Test 3	Test 4
Aluminum	590	660
Sodium	14,680	1,160

The above decontamination factors are probably as good as can be expected for this process.

Nitric Acid Dissolution

The product solids heel is readily dissolved with 32% nitric acid at about 80°C for 1 to 4 hr from either the filter or the dissolver pot. The impurities present in the solids are dissolved along with the product. Nonradioactive erbium oxide and dysprosium oxide solids were added into the dissolver with the aluminum target tube on both Tests 3 and 4 to simulate the actinide-lanthanide solids which would be present in the irradiated targets. Erbium, dysprosium, and the impurities present were dissolved both from the filter and from the dissolver pot after filtration of the solid-liquid heel remaining in the dissolver after washing in both Tests 3 and 4 (Tables 32 and 33).

TABLE 32

Test 3 Nitric Acid Dissolution of Solids

Description	H+,N	Quantity, g								
		Er	Dy	Al	Na	Fe	Mg	Cu	Cr	Ni
First HNO ₃ wash of filter	4.3	27.1	41.3	5.9	0.2	4.3	22.2	0.01	0.1	0.2
Deionized water rinse	0.1	1.4	2.7	NA ^a	NA	NA	NA	NA	NA	NA
Second HNO ₃ wash of filter	5.6	0.2	0.4	0.04	0.02	0.3	0.07	0.003	0.1	0.1
Deionized water rinse	0.1	<0.07	<0.01	NA	NA	NA	NA	NA	NA	NA
First HNO ₃ wash of dissolver	6.0	78.4	54.5	2.8	0.6	0.7	0.4	0.005	0.02	0.05
Deionized water rinse of dissolver	0.03	0.8	0.5	NA	NA	NA	NA	NA	NA	NA
Second HNO ₃ wash of dissolver	6.3	0.1	0.07	0.07	0.3	NA	NA	NA	NA	NA
Total	—	108.1	99.5	8.8	1.1	5.3	22.7	0.02	0.2	0.4

a. NA means not analyzed.

TABLE 33

Test 4 Nitric Dissolution of Solids

Description	H+,N	Quantity, g								
		Er	Dy	Al	Na	Fe	Mg	Cu	Cr	Ni
HNO ₃ wash of filter	4.3	2.9	6.5	4.9	0.3	7.1	9.8	0.5	0.4	0.3
Deionized water rinse of filter	0.5	0.03	0.09	NA	NA	NA	NA	NA	NA	NA
First HNO ₃ wash of dissolver	5.8	67.9	55.3	2.2	1.8	3.6	2.4	0.03	0.05	<0.02
Second HNO ₃ wash of dissolver	7.8	2.9	2.2	0.8	11.4	1.6	0.1	<0.02	0.2	<0.13
Total	-	73.7	64.1	7.9	13.5	12.3	12.3	0.5	0.7	0.4

The quantities of aluminum and sodium present will be much greater in the nitric acid dissolution of solids from plugged filters because washing of the filter and solids is not possible. The quantities of erbium, dysprosium, and impurities found in filter flushes from Tests 3 and 4 are shown in Tables 34 and 35.

TABLE 34

Test 3 Filter Flushes

Flush Made After Filtration of	Quantity, g								
	Er	Dy	Al	Na	Fe	Mg	Cu	Cr	Ni
Dissolver solution First part	<0.02	0.1	90.0	60.0	1.5	0.2	0.005	0.4	0.3
Above flush	<0.03	0.2	NA	0.1	0.3	0.001	0.002	0.003	0.02
Dissolver solution Last part	0.02	0.01	0.2	36.6	3.2	0.03	0.006	0.7	0.6
Above flush	<0.01	<0.01	0.1	0.1	0.03	0.001	0.001	0.008	0.01
First 10% NaOH wash	0.1	0.5	0.3	10.4	0.9	0.3	0.004	0.3	0.2
Second 10% NaOH wash	2.4	5.0	3.5	15.4	1.7	0.2	0.005	0.4	0.4
Above flush	<0.01	<0.01	0.03	0.04	0.01	0.03	<0.001	0.004	<0.01
First deionized water wash	0.2	0.8	0.3	1.1	0.6	0.01	0.003	0.2	0.2
Above flush	<0.01	<0.01	<0.01	<0.01	0.01	0.001	<0.001	0.004	<0.01
Second deionized water wash	0.04	0.06	0.1	0.5	0.3	0.2	<0.001	0.1	0.2
Above flush	<0.01	<0.01	<0.01	0.5	0.02	0.02	<0.001	0.003	<0.01
Total	2.9	6.7	94.6	125	8.6	1.0	0.05	2.1	2.0

TABLE 35

Test 4 Filter Flush

Flush Made After Filtration of	Quantity, g								
	Er	Dy	Al	Na	Fe	Mg	Cu	Cr	Ni
Dissolver solution	<0.02	0.02	6.2	1.2	0.005	0.1	0.008	0.3	0.2

Material Balance

The material balance is shown in Table 36 for the erbium and dysprosium added to Tests 3 and 4 to simulate the actinide-lanthanide product.

TABLE 36

Material Balance for Erbium and Dysprosium
in Tests 3 and 4

Description	Test 3		Test 4	
	Erbium	Dysprosium	Erbium	Dysprosium
HNO ₃ dissolution, g	108.1	99.5	73.7	64.1
Filter flushes, g	2.9	6.7	0.02	0.02
Filtrates, g	ND ^a	ND	ND	ND
Samples, g	17.8	17.8	10.7	10.8
Total output, g	128.8	124.0	84.4	74.9
Total input, g	126.0	126.0	126.0	126.0
Material balance, %	102.2	98.4	67.0	59.4

a. ND means not detected by analysis.

The material balance was very good for Test 3 and not as good for Test 4. The poor material balance for Test 4 cannot be explained.

REFERENCES

1. W. J. Jenkins. "Technical Data Summary - Multipurpose Dissolver for MPPF." DPST-72-328 (1972).
2. "Californium Separations Facility - Solution Transfer, Project S-1085." DPST-70-119-20, p 16 (October 29, 1970).