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SEPARATION OF FISSION PRODUCTS FROM ALUMINUM WASTE SOLUTIONS BY ION EXCHANGE

R. E. Blanco
I. R. Higgins
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CHEMICAL TECHNOLOGY DIVISION

CHEMICAL DEVELOPMENT SECTION

SEPARATION OF FISSION PRODUCTS FROM ALUMINUM WASTE SOLUTIONS BY ION EXCHANGE

R. E. Blanco
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0.0 ABSTRACT

A process utilizing a scavenging precipitation and ion exchange has been developed for the separation of fission products from aluminum waste solutions. The behavior of individual fission products in the aluminum-resin system was studied.

1.0 INTRODUCTION

The study presented in this report had a twofold objective: (1) an investigation of the practicability of the separation of aluminum from fission products by ion exchange, and (2) an investigation of the behavior of individual fission products in this system.

The disposal of radioactive wastes and the recovery of valuable radioisotopes are problems that will be encountered in conjunction with the U^{235} solvent-extraction process. The 25 Process waste* will be an aqueous aluminum nitrate solution containing about 0.5 curie of fission product activity per milliliter. The waste-disposal plan envisioned at present involves evaporation of the waste and storage in a tank farm. If the bulk of the inactive aluminum salts could be separated from the fission products, the decontaminated aluminum salt could be reused as a salting agent or disposed of easily, and the radioactive waste could be used as a source of valuable isotopes or evaporated to a much smaller volume and stored correspondingly more cheaply.

Ion exchange has several advantages as a unit operation for processing this system: ease of remote control, high yield, and purity of products. It provides an excellent starting point for isotope recovery since many of the isotope production processes utilize ion exchange. The nuclear sulfonic resins have been shown(1) to have sufficient radiation stability for use in this type of operation. The main disadvantage of ion exchange as applied to aluminum waste treatment is the large volume of resin needed, i.e., ~1 liter of Dowex-50 resin per 20.6 g of aluminum per cycle.

The experiments reported here were on a laboratory scale. Thus, before definite conclusions could be reached as to the feasibility of the process, pilot-plant studies would have to be made. The aluminum waste solutions used in the investigation were the raffinates** from the 25 Process solvent-extraction development program.

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*In the 25 Process, aluminum-U^{235} assemblies from the Materials Testing Reactor are dissolved in nitric acid. The uranium is recovered from the aluminum and fission products by solvent extraction.

**The term "raffinate" is used to mean the waste from the solvent-extraction column.

2.0 SUMMARY

A process that utilizes both scavenging and ion-exchange procedures has been developed for the separation of fission products from aluminum wastes. Beta and gamma decontamination factors of 10^3 with an aluminum yield of 97 to 99 per cent are obtained. At lower aluminum yields, factors of greater than 10^4 have been obtained. The volume of radioactive waste to be stored is reduced from its original volume by a factor of 33 to 100.

A plutonium decontamination factor of 5000 may be obtained by the use of an additional resin column preceding the ion-exchange column used for beta and gamma decontamination.

The behavior of the fission products and aluminum was studied in ion-exchange systems utilizing oxalic, nitric, and sulfuric acids and sodium hydroxide.

Alternate processes, in which the aluminum was precipitated as the hydrous oxide or sulfate, or in which the aluminum was passed through a resin bed saturated with aluminum, were investigated but did not prove satisfactory.

3.0 DESCRIPTION OF PROCESS

The recommended waste-decontamination process (see Fig. 3.1) consists of the following steps:

1. The waste is diluted to an aluminum concentration of approximately 0.5 M.

2. The radiocolloids are removed by a scavenging precipitation with hydrous ferric and manganic oxides. The filter cake, which contains 97 per cent of the total ruthenium, 99.5 per cent of the zirconium, and 90 per cent of the niobium, is dissolved in nitric acid–hydrogen peroxide and is either stored or used for recovery of fission products.

2a. If plutonium decontamination is required, the plutonium is reduced to Pu(III) and the solution is passed through a Dowex-50 resin precolumn to adsorb and eliminate the plutonium before step 3 is carried out.

3. The filtrate is diluted to 0.1 M aluminum, made 0.016 M in oxalic acid to complex the fission products, and passed through a Dowex-50 resin column, where the aluminum and a portion of the fission products are adsorbed. The column is washed with 0.016 M oxalic acid.

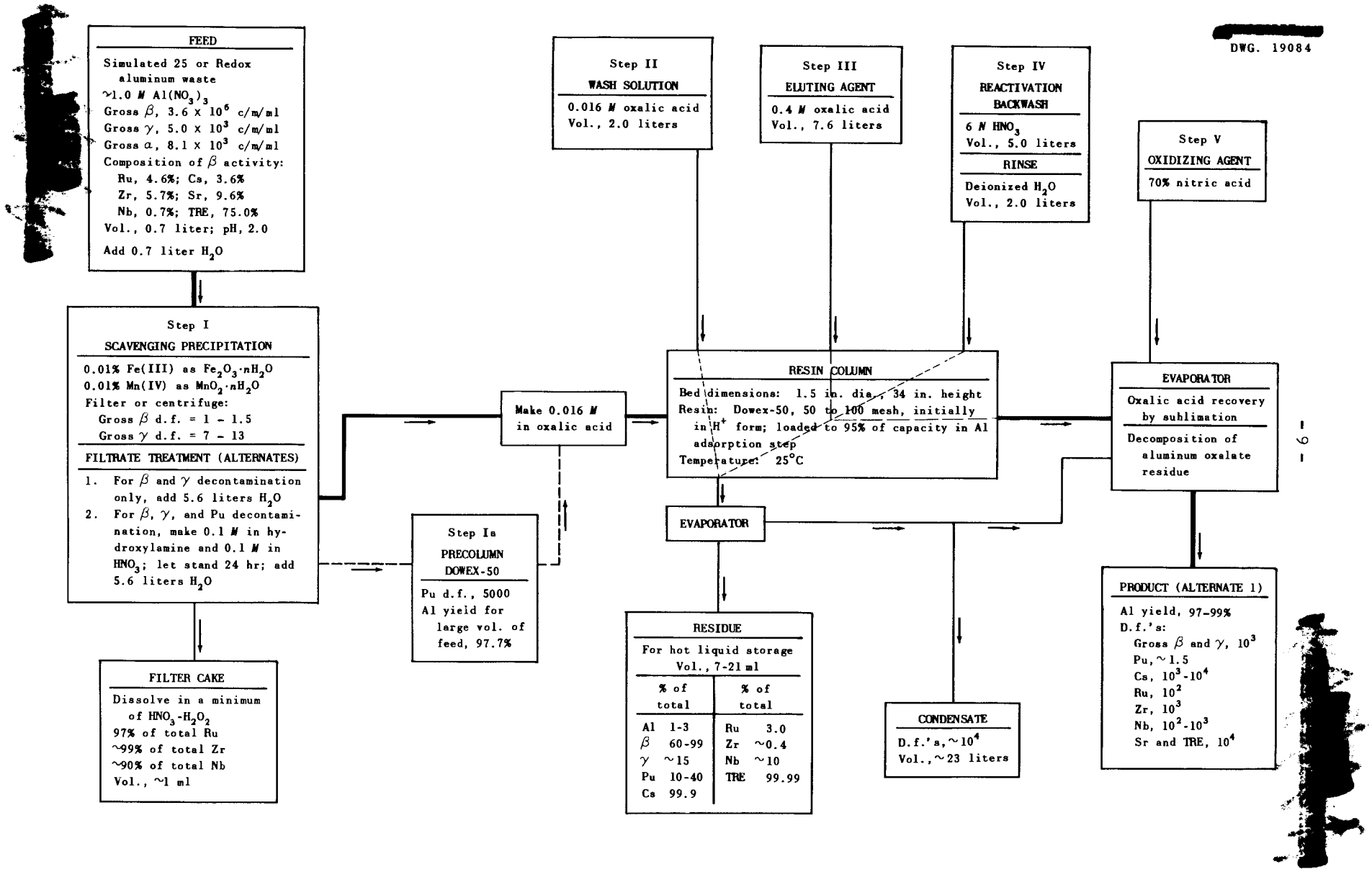

Fig. 3.1. Flowsheet for Aluminum Waste Decontamination.

4. The aluminum is eluted with 0.4 M oxalic acid, and the eluate is evaporated to dryness. Excess oxalic acid is recovered from this residue by sublimation, and the aluminum oxalate is decomposed with nitric acid.

5. The fission products are eluted from the resin column with 6 M HNO_3. This eluate is evaporated and either stored or used for recovery of the fission products.

6. The resin is rinsed with water, and the column is ready for reuse.

The aluminum yield in the process is 97 to 99 per cent. Decontamination factors are of the order of 10^3 to 10^4.

Summaries of various experimental runs are given in Appendix 1 (Tables A1.1 through A1.5).

4.0 SCAVENGING PRECIPITATION

The purpose of the scavenging precipitation is to remove the radiocolloidal activity, which caused erratic results in initial attempts to separate the fission products from aluminum by ion exchange. The following procedure, which gives a gross beta decontamination factor of 1 to 2 and a gross gamma decontamination factor of 7 to 13, is recommended:

1. The 25 Process waste is diluted to 0.5 M aluminum, giving a solution of pH approximately 2, and heated to boiling. Ferric nitrate, as crystals or in solution, is added, and the mixture is boiled for 1 min. Red colloidal hydrous ferric oxide is formed.

2. Manganous nitrate is added to the waste solution, and hydrous manganese dioxide is precipitated with the ferric oxide by adding potassium permanganate slowly to the boiling solution.

3. The precipitate is digested for half an hour and is then removed by filtering, centrifuging, or settling.

Both the iron and manganese should be limited to 0.01 per cent, by weight, of the waste solution. The investigations that led to the recommendation of this procedure were made on aqueous waste solutions that were 0.1 to 1.0 M in aluminum nitrate and that varied in pH from 2.0 to 3.0.

Previous work had shown that up to 96 per cent of the ruthenium and zirconium and nearly quantitative amounts of niobium can be carried on a precipitate of hydrous manganese dioxide.[2,3,4] The coprecipitation of hydrous ferric oxide with the manganese dioxide was recommended after it was shown to raise the ruthenium decontamination factor to 33-100.

It was known from previous work[3] that the efficiency of carrying varies inversely with variations in the bulk ionic strength. Dilution of the waste to approximately 0.5 M aluminum is therefore recommended, not only because it increases the decontamination, but also because it was shown that the lower ionic strength decreased the tendency of the manganese dioxide to become colloidal.

In a series of scouting runs made on a resin column, 16.5 cm height by 1.4 cm diameter, a scavenging step preceding the adsorption step was shown to increase the overall decontamination of ruthenium, zirconium, and niobium as much as 100-fold. The increase in gross beta and gamma decontamination factors was limited to a factor of 10, however, because of the poor decontamination of cesium under these column conditions (see Table A1.2).

4.1 Effect of pH

The pH used for the scavenging precipitation in all experimental runs was approximately 2, which is the pH of the 25 Process waste as it comes from the columns.

Efficient precipitation of hydrous ferric oxide was obtained at pH 11, with a gross gamma decontamination factor of approximately 4. The aluminum remained in solution as the aluminate ion. The large amount of sodium that would have to be added to the waste to adjust the pH in this case, however, would increase the storage problem, and scavenging precipitations at high pH values are therefore not considered practical.

4.2 Effect of Digestion Time

Studies on the effect of total digestion time on decontamination by scavenging showed little variation of gross gamma decontamination with digestion time of the ferric oxide. Approximately 1 min was therefore considered satisfactory (see Table 4.1).

Boiling coformed manganese dioxide in the aluminum waste (0.10 M aluminum) for 24 hr caused the gross gamma decontamination factor to drop from 9 after 5 min to 6 after 24 hr. To ensure complete reaction of the manganese, 0.5 hr was taken as the optimum digestion time.

4.3 Effect of Form of Carrier

Freshly prepared and digested hydrous ferric oxide was added as a slurry to the boiling waste, digested for 1 min, and filtered. This step was repeated and was then followed by coprecipitation of manganese dioxide, digestion, and filtration, as described in the recommended procedure. The decontamination factors observed were no higher than those obtained with ferric oxide prepared in the solution.

Commercial manganese dioxide was shown to be effective as a scavenger, although it probably would not remove peptized ferric oxide as efficiently as would coformed manganese dioxide (see Tables 4.2 and 4.3). The individual fission product decontamination factors obtained by scavenging are shown in Table 4.4.

4.4 Effect of Amount of Carrier

Varying the amount of iron carrier from 0.01 to 0.1 per cent by weight of the waste solution and the manganese from 0.01 to 0.05 per cent made no difference in the gross decontamination factors (see Tables 4.1 through 4.4 and A1.2 through A1.4). In order to keep added solids at a minimum, 0.01 per cent of each element is recommended for the scavenging procedure.

Table 4.1

Effect of Digestion Time on Scavenging Decontamination Factor

Waste solution used: 25 Process aluminum waste diluted to 0.1 M aluminum, pH 2.5

Scavenger	Total Time in Boiling Solution (min)	Gross Decontamination Factors	
		β	γ
0.01% Fe(III) as preformed hydrous ferric oxide, added as a slurry	5 (at room temp.)	1	1.6
	1	1	2.1
	15	1	2.0
	30	1	2.3
0.01% Fe(III) as coformed hydrous ferric oxide, precipitated at pH $\sim$11 by NaOH at 100°C	5	5	3.5
	90	5.3	3.2
	24 x 60	6.2	4.3
0.01% Mn(II) as coformed hydrous manganese dioxide precipitated by addition of $KMnO_4$ to $Mn(NO_3)_2$ at 100°C	5	1.5	9.2
	30	1.5	6.7
	60	1.5	7.8
	24 x 60	1.5	5.8

Table 4.2

Scavenging of 25 Process Aluminum Waste by Preformed Commercial Manganese Dioxide

Conditions: solution boiled for 1 hr after the addition of MnO_2

MnO_2 (%)	Al in 25 Process Waste (M)	Gross Decontamination Factors	
		β	γ
0.1	0.1	1.2	3.3
2.0	0.1	1.0	7.0
2.0	1.0	3.6	6.0

Table 4.3

Scavenging of 25 Process Aluminum Waste with Preformed Hydrous Ferric Oxide and Coformed Hydrous Manganese Dioxide

Waste solution used: 25 Process aluminum waste, diluted to 0.1 M aluminum

Run No.	Scavenger	Step No.*	Gross Decontamination Factors			
			Stepwise		Overall	
1	0.1% Fe(III)	1	1	6.3		
	+ 0.05% Mn(II)	2	1	1		
		3	2.0	2.1	2.0	13.2
2	Same	1, 2, 3			1.1	8.0
3	Same	1, 2, 3			1.0	8.0
4	0.02% Fe(III)	1	1	3		
	+ 0.01% Mn(II)	2, 3	1.3	2.6	1.3	7.8
5	Same	1	1	3.4		
		2, 3	1.3	3.4	1.3	11.5
6	Same	1, 2, 3			1.6	7.6
7	Same	1, 2, 3			1.6	10.8

*See Sec. 4.3.

Table 4.4

Individual Fission Product Decontamination Factors Obtained by Scavenging with Hydrous Manganese Dioxide and Hydrous Ferric Oxide

Waste solution used: 25 Process aluminum waste diluted to 0.1 M aluminum

Scavenger	Decontamination Factors					
	Ru	Zr	Nb	Cs	Sr	Rare Earths
0.05% Fe(III) as coformed $Fe_2O_3 \cdot nH_2O$	30 - 76	8.5	1 - 3	2	1	1
0.02% Fe(III) as coformed $Fe_2O_3 \cdot nH_2O$ and 0.01% Mn(II) as coformed $MnO_2 \cdot nH_2O$	30 - 300	20 - 190	7 - 10	1	1	2
2% preformed MnO_2	6.5	47	11	1.9	2	4

5.0 ION-EXCHANGE OPERATING CONDITIONS

5.1 Type of Resin

Dowex-50 resin was chosen for waste-treatment studies because of its high exchange capacity and because it is chemically resistant to the 6 M HNO_3 used to remove adsorbed activities and to regenerate the column. The resins IR-100 and Dowex-30 were investigated briefly, but their adsorption capacities were shown to be only one-fifth to one-third of the capacity of Dowex-50, and they were readily attacked by strong nitric acid.

5.2 Resin Particle Size and Flow Rates

Both 60- to 80- and 100- to 200-mesh resins were used in these studies. No difference due to the resin particle size was noted in the results. General experience, however, has indicated that 50- to 100-mesh is a satisfactory compromise between hydraulic and diffusion efficiencies (see below). All flow rates were at 1 $ml/m/cm^2$ or less.

The efficiency of the ion-exchange process is known to be controlled by a combination of the rates at which equilibrium is reached in the diffusion and mass action systems.[5,6,7] Consequently, the resin particle size is of considerable importance in ion-exchange processes, the use of smaller resin particles resulting in more rapid equilibrium during adsorption and in sharper band edges[8] during elution. Thus, faster

flow rates can theoretically be tolerated with smaller particle sizes. Practically, however, extremely small resin particles produce a high resistance to the flow of liquid in the column (Table 5.1), and a compromise must be made between resin size and the hydrostatic head needed to maintain an efficient but economic flow rate.

Table 5.1

Pressure Required to Maintain a Given Flow Rate in Resin Columns of Various-Sized Particles

Resin Particle Size (mesh)	Height of Resin Column (cm)	Liquid Flow Rate ($ml/m/cm^2$)	Pressure Head Required to Maintain Flow Rate (ft H_2O)
60 - 80	106	1	1
100 - 200	69	0.5	12
200 - 500	69	0.5	28

5.3 Column Dimensions

Decontamination of the aluminum was increased by increasing the height of the resin column, but variations in the column diameter did not affect the decontamination noticeably.

The heights and diameters of the columns used in the study were: 16.5 by 1.4 cm, 69 by 0.6 cm, 106 by 0.6 cm, and 106 by 3.8 cm. The increase in decontamination that was observed with the longer column was primarily a reflection of the movement of the cesium band, which was, necessarily, close to the bottom of the column in the short column and was therefore more likely to be eluted with the aluminum. Varying the column diameter from 0.6 to 3.8 cm did not noticeably affect the decontamination (see Tables A1.1 through A1.5).

The effect of column height can be seen by comparing run 3 (Table A1.2) with run D-50-20 (Table A1.3). In this case the gross beta and gamma decontamination factors rose from 56 and 42 to 3.3×10^3 and 1.5×10^3, respectively, as the column height increased from 16.5 to 106 cm. The other experimental conditions were approximately the same in these two runs.

6.0 ADSORPTION STEP

6.1 Complexing Agents in Feed

The use of a complexing agent in the feed to prevent adsorption of the fission products by the resin achieves a partial decontamination of the aluminum. The fission products pass into the feed effluent along with other anions and are either recovered or stored.

Oxalic Acid. Oxalic acid complexes zirconium and niobium, but its concentration must be kept low to prevent complexing of the aluminum. An oxalic acid concentration of 0.016 M in the feed has been found satisfactory (see Tables A1.2 through A1.4).

Fluorides. The use of 0.1 per cent HF in the feed to complex zirconium and niobium (and 0.5 M HF as the elutriant) gave beta and gamma decontamination factors of 20 and 2, respectively (see Table A1.1); 0.5 per cent of the gamma activity was eliminated. In one run in which 0.016 M oxalic acid was used as the complexing agent (and 0.5 M NH_4F as the elutriant; see Table A1.1), cesium and gross gamma decontamination factors of 10 and 20, respectively, were obtained; 61 per cent of the gamma activity was present in the feed effluent. Although the two runs are not strictly comparable, since there were variables other than the complexing agent, the primary cause for the lower gross gamma decontamination obtained in the first case is considered to be the complexing agent used. The cesium decontamination factor was expected to be much lower in the second case than in the first because highly ionized ammonium fluoride was used as the elutriant instead of the weakly ionized hydrofluoric acid. Unfortunately, the hydrofluoric acid eluate was accidentally destroyed before a radiochemical analysis for cesium could be completed. Since the hydrogen fluoride offered no particular advantages and had the disadvantages of corrosiveness and of increasing the solids content of the waste, study of fluorides as a complexing agent was not continued.

6.2 Effect of Aluminum Concentration

The 0.5 M aluminum waste solution was diluted to 0.1 M aluminum before being fed to the column. The mass ion theory of ion exchange predicts that the higher the bulk ionic strength of the feed solution, the farther the fission products will travel down the column before being adsorbed. The use of a dilute feed solution keeps the fission products relatively high on the resin column and permits their maximum separation from aluminum in the elution.

6.3 Effect of Percentage of Resin Saturated with Aluminum

With increasing saturation of the resin with aluminum, cesium decontamination is decreased and ruthenium decontamination is increased. Adsorption conditions were chosen to favor cesium decontamination since ruthenium decontamination is higher in the scavenging step.

The highest cesium decontamination factors were obtained when the resin was less than 95 per cent saturated with the aluminum (see Tables A1.2 through A1.4). The factors were at least 10^3 lower when the aluminum in the feed was equal to or greater than the capacity of the resin. This effect is attributed to the fact that the cesium is adsorbed lower on the column (i.e., adsorbed less strongly), than the aluminum. Large amounts of aluminum therefore force the cesium so far down the column that it is readily eluted with the aluminum.

Since the exchange potential of ruthenium is lower than that of either cesium or aluminum, saturation of the resin with aluminum forces the ruthenium off the column during the adsorption step. However, if the resin is not saturated with aluminum, and some ruthenium is adsorbed, it is readily eluted along with the aluminum in the elution step.

7.0 SEPARATION OF PLUTONIUM FROM ALUMINUM

If the decontaminated aluminum nitrate is to be stored or reused in the Redox process, removal of the small amounts of plutonium expected to be present will not be necessary. However, if removal of the plutonium is desired, a decontamination factor of 5000 may be obtained by the following process: The waste, which in the case of the 25 Process or Redox raffinate is expected to be approximately 1.0 M in aluminum and to have a pH of 1.8 to 2, is diluted to 0.5 M aluminum and made 0.1 M in HNO_3 and 0.1 M in hydroxylamine and allowed to stand for 24 hr. This procedure reduces Pu(VI) and Pu(IV) to Pu(III), which is more strongly held on a resin column than is aluminum. Indications are that a cheaper reducing agent, such as formaldehyde, and a shorter reduction time are possible. The feed is then diluted to 0.1 M aluminum and run into a Dowex-50 resin column, where the plutonium is adsorbed. The aluminum, after saturating the resin, passes (breaks) through the column.

Runs were made in which nine times the amount of aluminum held on the resin passed through with no indication of plutonium breakthrough. Experiments using macro amounts of plutonium must be made to determine the capacity of a resin column for plutonium under those conditions. An estimate of the maximum amount of trace plutonium that could be held, however, can be made by using the distribution coefficient, K_d:

$$K_d = \frac{\text{mass of Pu per ml of resin}}{\text{mass of Pu per ml of solution}} = 679.$$

This value was determined by equilibrating aluminum-form resin and 0.1 M $Al(NO_3)_3$ solution containing plutonium tracer; it indicates that 679 liters of 0.1 M aluminum solution containing plutonium in tracer concentration can be passed through a column containing 1 liter of resin before any plutonium appears in the effluent stream. This would correspond to an

[redacted]

Table 7.1

Plutonium Decontamination by Breakthrough Method

Resin: Dowex-50, 60 to 80 mesh, initially in acid form
Column dimensions: 106 cm height, 6 mm diameter
Flow rates: 0.5 ml/m/cm^2
Procedure: feed was passed through column and effluent analyzed for Pu
Al yield (%): $\frac{\text{Total Al in effluent} \times 100}{\text{Total Al in feed}}$

Run No.	Feed	Reduction Conditions		Pu α in Diluted Feed (c/m/ml)	Al Yield (%)	Overall Pu Decontamination Factor
		Acidity	NH_2OH Conc. (M)			
1	25 Process waste previously scavenged with $Fe_2O_3 \cdot nH_2O$ and $MnO_2 \cdot nH_2O$, diluted 10 times to 0.1 M Al	pH ~2.5	0	946*	91.5	3.5
2	Unscavenged 25 Process waste diluted to 0.1 M Al, then reduced with hydroxylamine over a 24-hr period	pH ~2.5	0.01	1146**	86	53
3	Same	pH ~2.5	0.10	1146	76	48
4	25 Process waste made 0.05 M in $K_2Cr_2O_7$ to simulate Redox waste; reduction made before dilution to 0.1 M Al	1.0 M HNO_3	1.00	634	92	158
5	Pu tracer reduced in 10 ml volume before dilution to 1 liter with 0.1 M Al	1.0 M HNO_3	1.00	97,000	75.5	1000
6	Same as Run 4 except Pu tracer used and no $K_2Cr_2O_7$	0.1 M HNO_3	0.10	99,000	88.5	5824
7	Same as Run 6 except no acid used	pH 2.5	0.10	97,000	~88.5	~4292

*Pu(III) = 4425 c/m/ml, Pu(VI) = 2636 c/m/ml.
**Pu(III) = 1038 c/m/ml, Pu(VI) = 126 c/m/ml.

[redacted]

aluminum yield of 98.9 per cent, free from plutonium. The other 1.1 per cent aluminum would remain on the resin with the plutonium.

The acidity at which the plutonium was reduced was found to be critical when actual raffinates from 25 Process runs were used. Plutonium decontamination factors varying from 15 to 20 were the highest that could be obtained when the reduction was performed at pH's of 2.0 to 2.5. However, when the solution was made 1.0 M in HNO_3 during the reduction, the plutonium decontamination factor rose to 150. This decontamination is considered excellent for a solution originally containing only 1000 plutonium alpha c/m/ml, because of the low counting accuracy at background levels. When the original level of the plutonium alpha activity was raised to 10^5 c/m/ml by the addition of plutonium tracer, the plutonium decontamination factor rose to 5800. Under these conditions, the acidity of the solution during reduction did not materially affect the plutonium decontamination (see Table 7.1). It should be noted that the plutonium contained in the actual raffinates is much more likely to be polymerized or colloidal than is tracer plutonium. It is therefore recommended that the raffinates be acidified before reduction.

This plutonium decontamination process could easily be combined with the oxalic acid elution process described in Sec. 3.0. The filtrate from the scavenging precipitation would be reduced with hydroxylamine, diluted, and passed through the pretreatment column to remove the plutonium. The effluent would be made 0.016 M in oxalic acid and passed into the aluminum adsorption column for beta-gamma decontamination. The pretreatment column would be reactivated with 6 M HNO_3 when the alpha count in the effluent started to rise, indicating plutonium breakthrough.

8.0 ELUTION STEP

Oxalic acid, at a concentration of 0.4 M and reagent pH (0.78) is recommended for aluminum elution (see Figs. 8.1 through 8.3). The advantages of oxalic acid are:

1. A relatively small volume of elutriant is required because an aluminum oxalate complex is formed.

2. The decontamination obtained with oxalic acid is nearly 100 times higher than that obtained with other elutriants. This is the result of the low ionization of both the oxalic acid and the aluminum oxalate, which makes the cation concentration low during elution so that only a small amount of activity is displaced from the column with the aluminum.

3. No solids are added to the waste solution since the oxalates can be decomposed to carbon dioxide by boiling with nitric acid.

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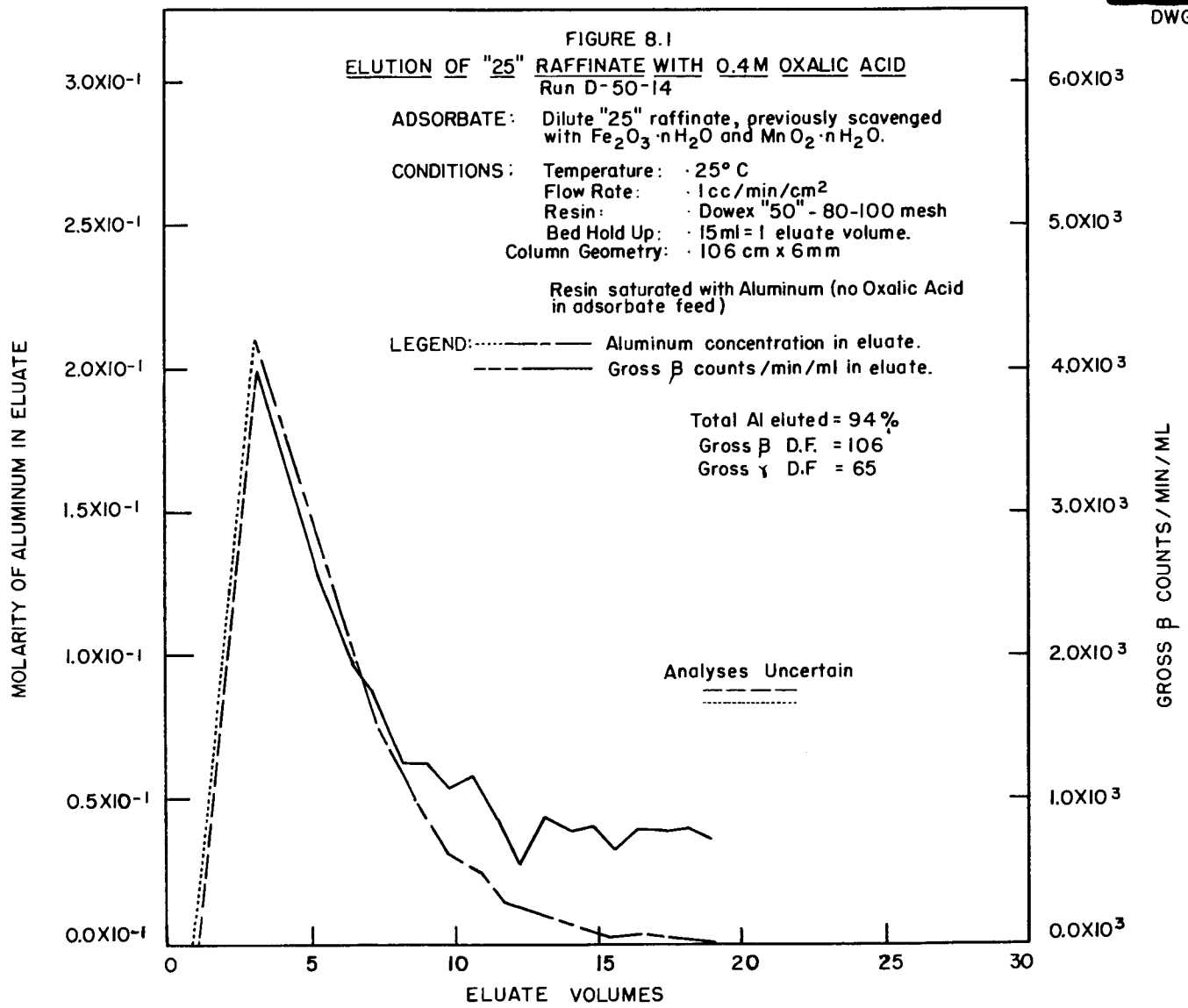

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FIGURE 8.2

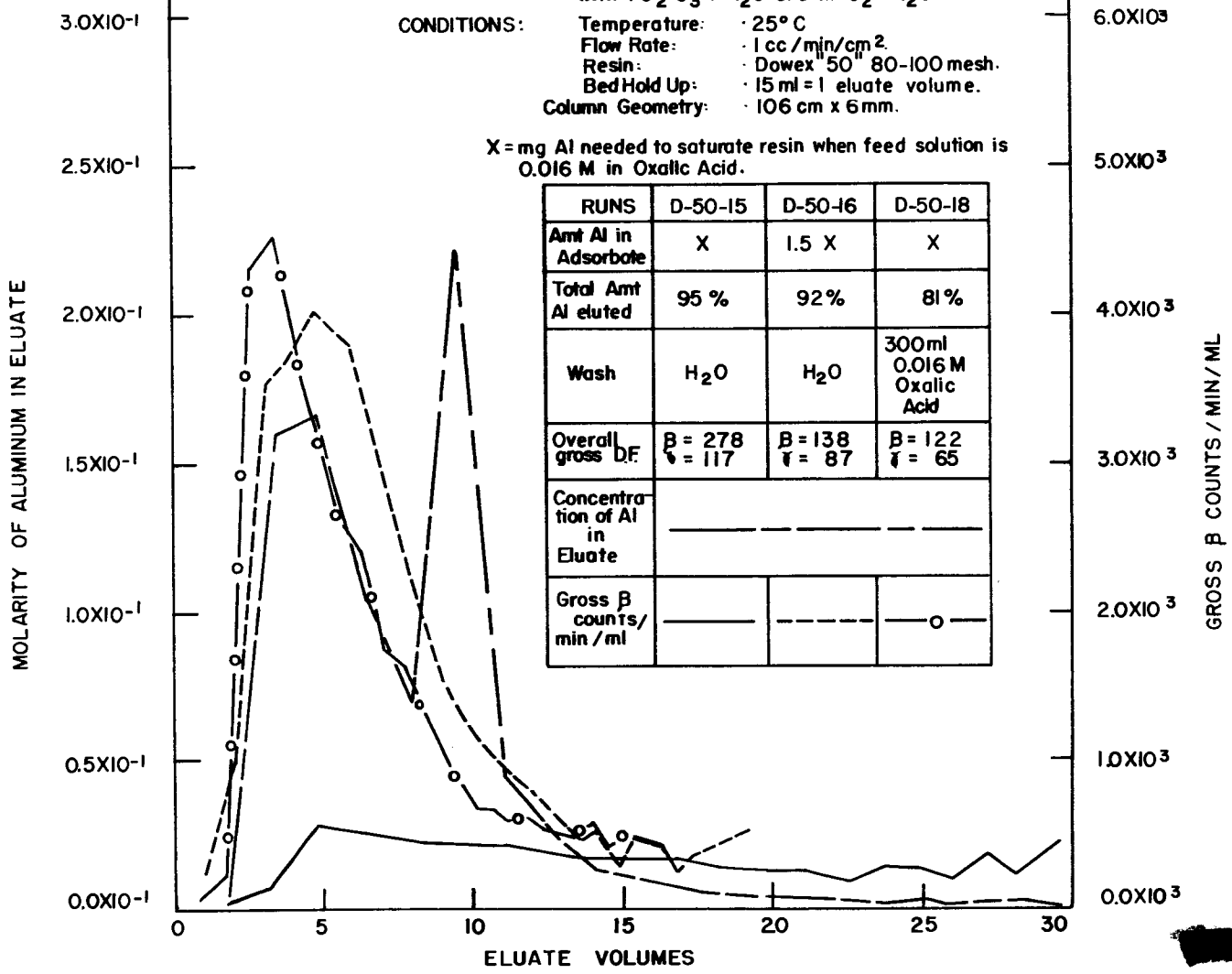
ELUTION OF "25" RAFFINATE WITH 0.4M OXALIC ACID

ADSORBATE: Dilute "25" raffinate, previously scavenged with $Fe_2O_3 \cdot nH_2O$ and $MnO_2 \cdot nH_2O$.

CONDITIONS:
- Temperature: · 25° C
- Flow Rate: · 1 cc/min/cm^2.
- Resin: · Dowex "50" 80-100 mesh.
- Bed Hold Up: · 15 ml = 1 eluate volume.
- Column Geometry: · 106 cm x 6 mm.

X = mg Al needed to saturate resin when feed solution is 0.016 M in Oxalic Acid.

RUNS	D-50-15	D-50-16	D-50-18
Amt Al in Adsorbate	X	1.5 X	X
Total Amt Al eluted	95 %	92 %	81 %
Wash	H_2O	H_2O	300 ml 0.016 M Oxalic Acid
Overall gross D.F.	β = 278 γ = 117	β = 138 γ = 87	β = 122 γ = 65
Concentration of Al in Eluate	———	———	———
Gross β counts/min/ml	———	- - - -	—o—



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Dwg No 6671

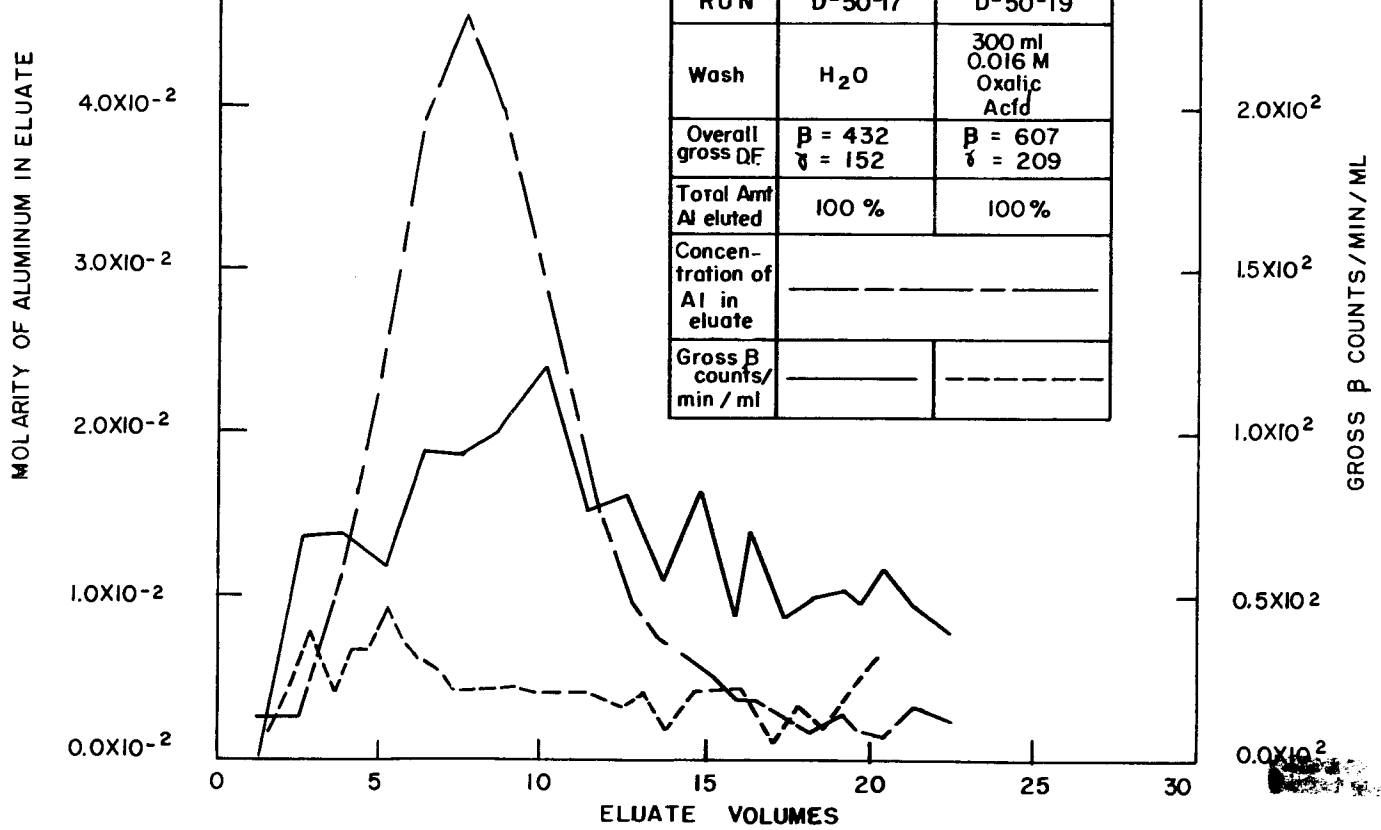
FIGURE 8.3

ELUTION OF "25" RAFFINATE WITH 0.4 M OXALIC ACID

ADSORBATE: Dilute "25" raffinate, previously scavenged with $Fe_2O_3 \cdot nH_2O$ and $MnO_2 \cdot nH_2O$, made 0.016 M in Oxalic Acid.

CONDITIONS:
- Temperature: 25° C
- Flow Rate: 1 cc/min/cm²
- Resin: Dowex "50"-80-100
- Bed Hold Up: 15ml = 1 eluate volume
- Column Geometry: 106 cm x 6mm
- Resin 20% saturated with aluminum

RUN	D-50-17	D-50-19
Wash	H_2O	300 ml 0.016 M Oxalic Acid
Overall gross DF	β = 432 γ = 152	β = 607 γ = 209
Total Amt Al eluted	100 %	100 %
Concentration of Al in eluate	———	- - - - -
Gross β counts/min/ml	———	- - - - -



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The chief disadvantage of oxalic acid is that it complexes plutonium, so that the bulk of the plutonium is eluted with the aluminum, giving low plutonium decontamination factors.

8.1 Effect of Oxalic Acid Concentration

Overall gross beta and gamma decontamination factors as high as 10^3 were obtained in runs in which 0.4 M oxalic acid at its natural pH of 0.78 was used as the elutriant (see Tables A1.3 and A1.4). The aluminum yield varied from 97 to 99 per cent. Increasing the oxalic acid concentration of the elutriant to 0.71 M increased the speed of the elution but did not increase the decontamination; also, the amount of oxalic acid required per mole of aluminum was actually higher in this case. Thus, it was concluded that there was no advantage in increasing the concentration.

8.2 Effect of pH

Increasing the pH of the oxalic acid elutriant from its natural value of 0.78 to 2.5 (with ammonium hydroxide) increased the gamma decontamination factor to 10^4 for 95 per cent aluminum elution (see Table A1.3), but the beta and gamma decontamination factors dropped to 1.8 and 12.9, respectively, for 99 per cent aluminum elution. This drop in decontamination was due to the sudden breakthrough of cesium (see Table A1.3).

8.3 Effect of Temperature

The exact effect of temperature on the aluminum distribution coefficient in the oxalic acid—aluminum—resin system is not known, but the rate at which equilibrium is attained would be expected to increase with an increase in temperature, from analogy with the rare earth—citric acid system described by Ketelle and Boyd.[8] In one run with 0.4 M oxalic acid elutriant at 90°C, the aluminum yield was as high as that at 25°C, but gross beta and gamma decontamination factors were only 15 and 2, as compared with corresponding values of 25 and 7 obtained at 25°C. The effect of temperature on oxalic acid elution has not been fully investigated.

8.4 Other Elutriants Studied

Sodium Hydroxide. Elution of the aluminum with sodium hydroxide is rapid and can be carried out with a small volume of elutriant, because of the formation of the complex AlO_2^- ion. Elution with sodium hydroxide is not recommended for process use, however, because concentrations of sodium hydroxide high enough to hold the complex anion in solution increase the cation concentration sufficiently to elute a portion of the cesium and other fission products with the aluminum. Thus, the aluminum decontamination is lowered and the bulk of the materials to be stored is increased owing to the large amount of sodium added to the system.

Studies were made with 0.1, 0.3, and 1.0 M NaOH (see Figs. 8.4 through 8.6 and Table 8.1). Gross beta and gamma decontamination factors of 8 and 3, respectively, were obtained with 85 per cent aluminum elution. With 0.1 M NaOH the pH of the column (7 to 8) was not high enough during the initial surge of aluminum elution to hold the AlO_2^- in solution, and a mixture of $NaAlO_2$ and $Al_2O_3 \cdot nH_2O$ precipitated in the column. This never completely redissolved.

Table 8.1

Decontamination Factors Obtained by Elution of Aluminum with Sodium Hydroxide

Feed to process: 25 Process aluminum waste, diluted to 0.1 M aluminum
Resin: Dowex-50, 60 to 80 mesh
Column specifications: 16.5 cm height, 1.4 cm diameter;
1 column volume = 15 ml
Temperature: 25 ± 1°C
Flow rates: 1 ml/m/cm²

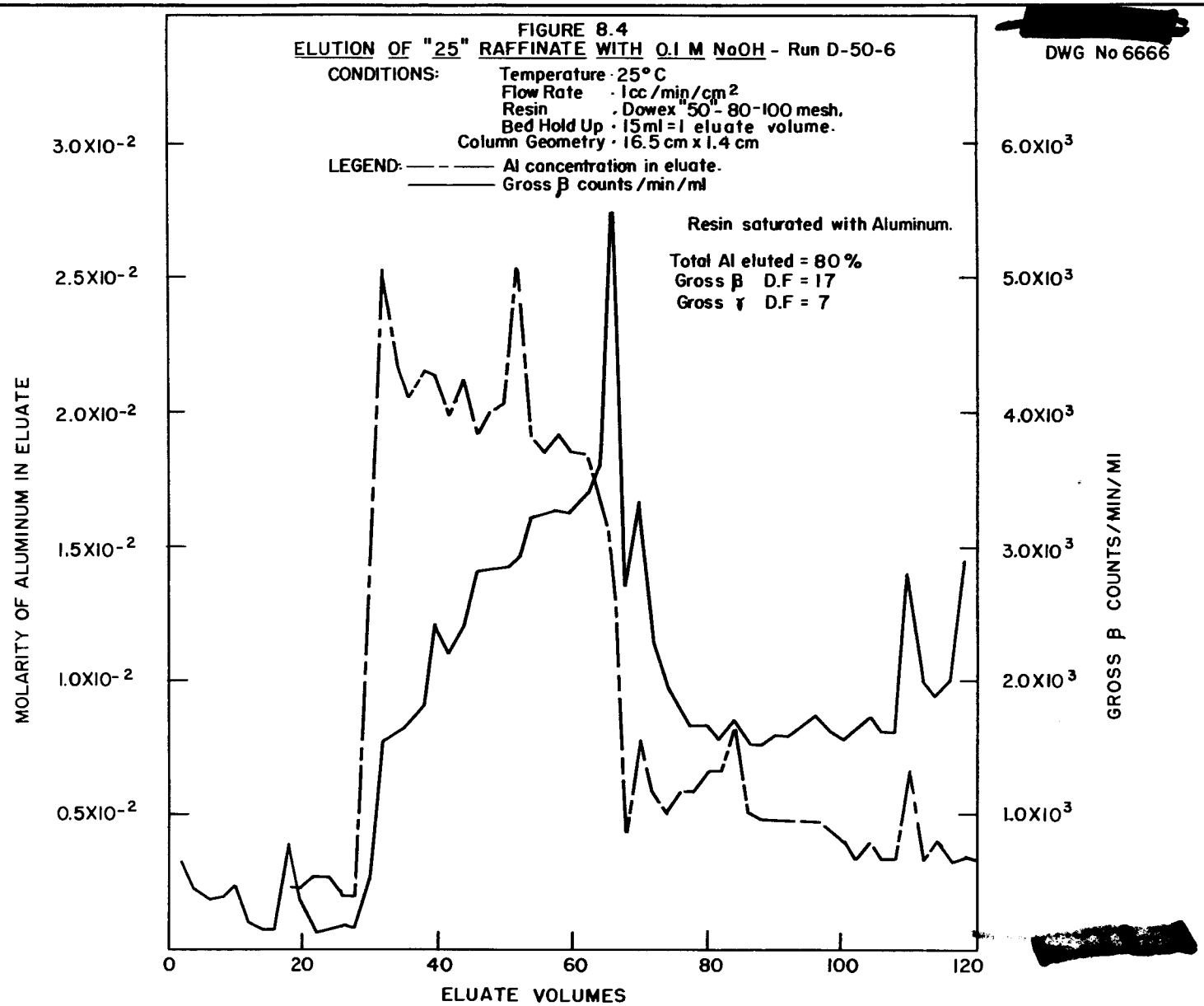
Run No.	NaOH Conc. of Elutriant (M)	Al Eluted (%)	Decontamination Factors							
			Gross		Individual Fission Products					
			β	γ	Ru	Zr	Nb	Cs	Sr	TRE
1	0.1	65*	17	7	5.5	72	35	3	780	350
2	0.3	40	99	196	30	128	37	79	5,700	80,000
		94	15	8	5	---	6	1.7	3,000	5,000
3	0.5	85	8.4	2.6	2.7	8.3	4.6	1.3	190	3,000

*Aluminum precipitated in column.

About 60 per cent of the beta activity in the sodium hydroxide eluate was cesium.

Fluorides. Fluoride forms a strong complex with aluminum, and aluminum elution with fluorides is rapid and complete. However, the cation concentration, because of ionization of the ammonium fluoride, is so high that low decontamination from cesium is obtained. Since hydrofluoric acid is considered too corrosive for use, and the presence of fluorides would increase the bulk of the waste to be stored, fluorides are not recommended.

Nitric Acid. If satisfactory elution could be obtained with nitric acid, there would be the advantage that no undecomposable ion was being added to the waste. However, elution with either 0.3 or 0.5 M HNO_3 was slow and incomplete, less than 50 per cent of the aluminum being eluted in most runs (see Figs. 8.7 through 8.9 and Table 8.2). Gross beta and gamma



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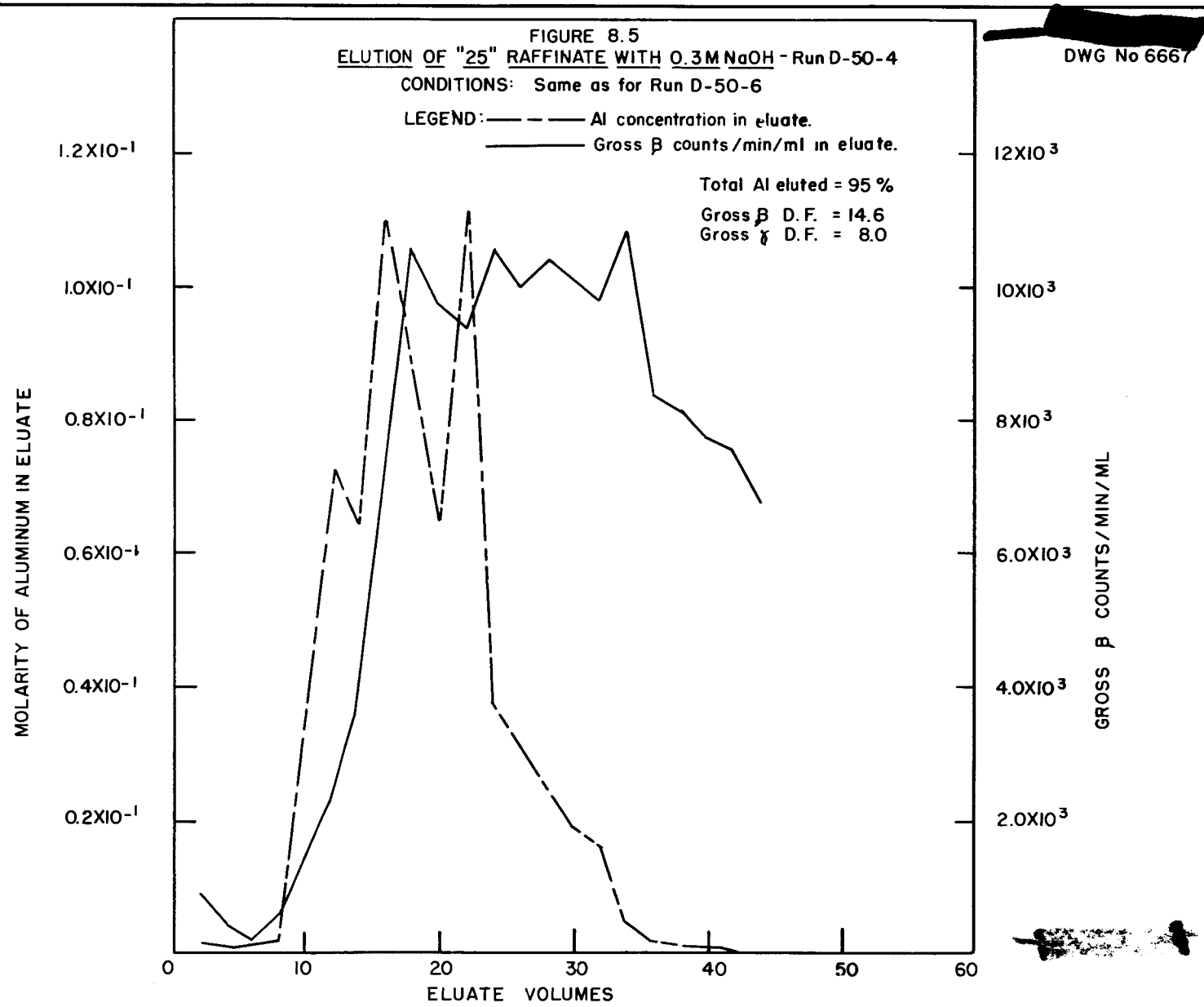

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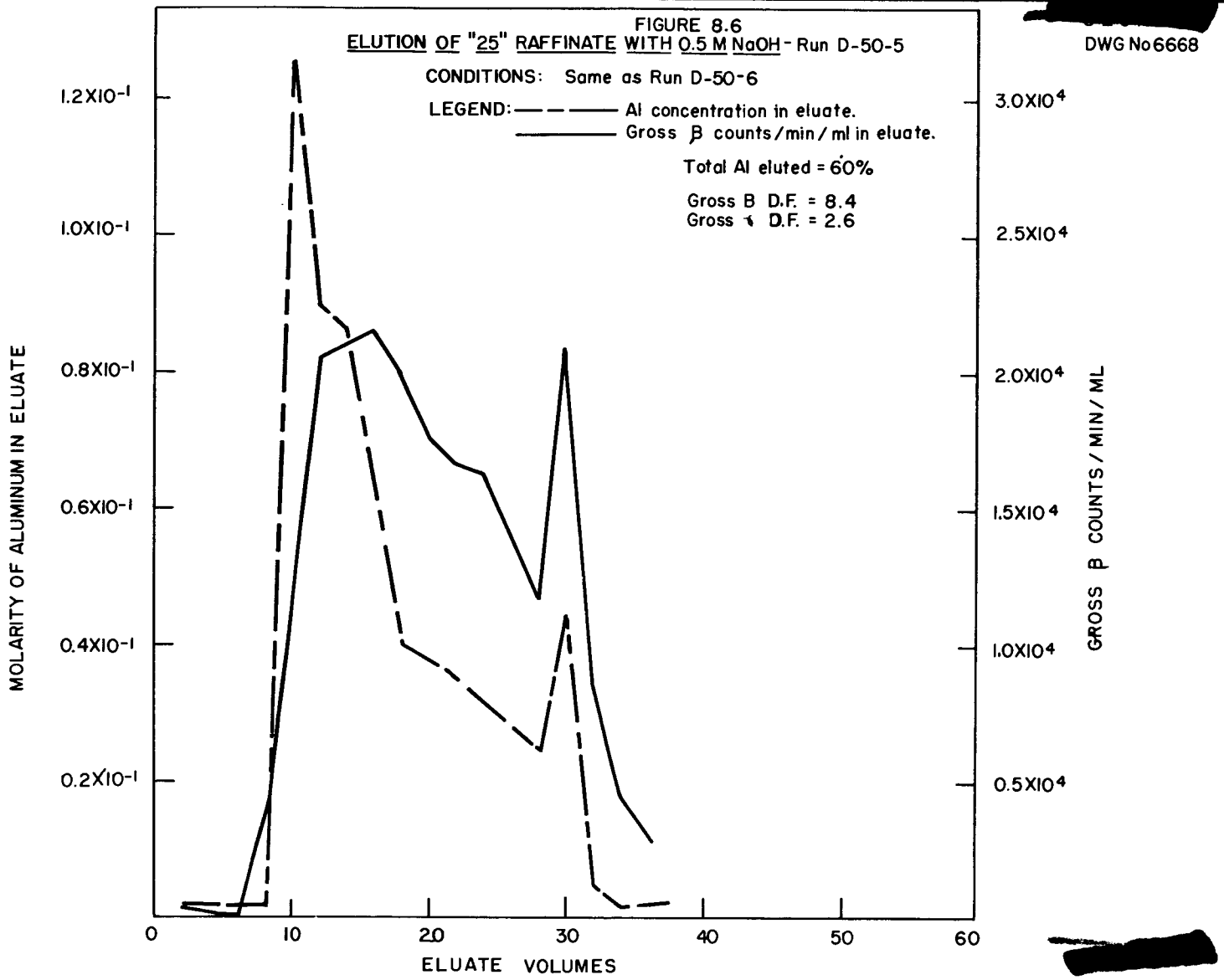

FIGURE 8.6
ELUTION OF "25" RAFFINATE WITH 0.5 M NaOH - Run D-50-5

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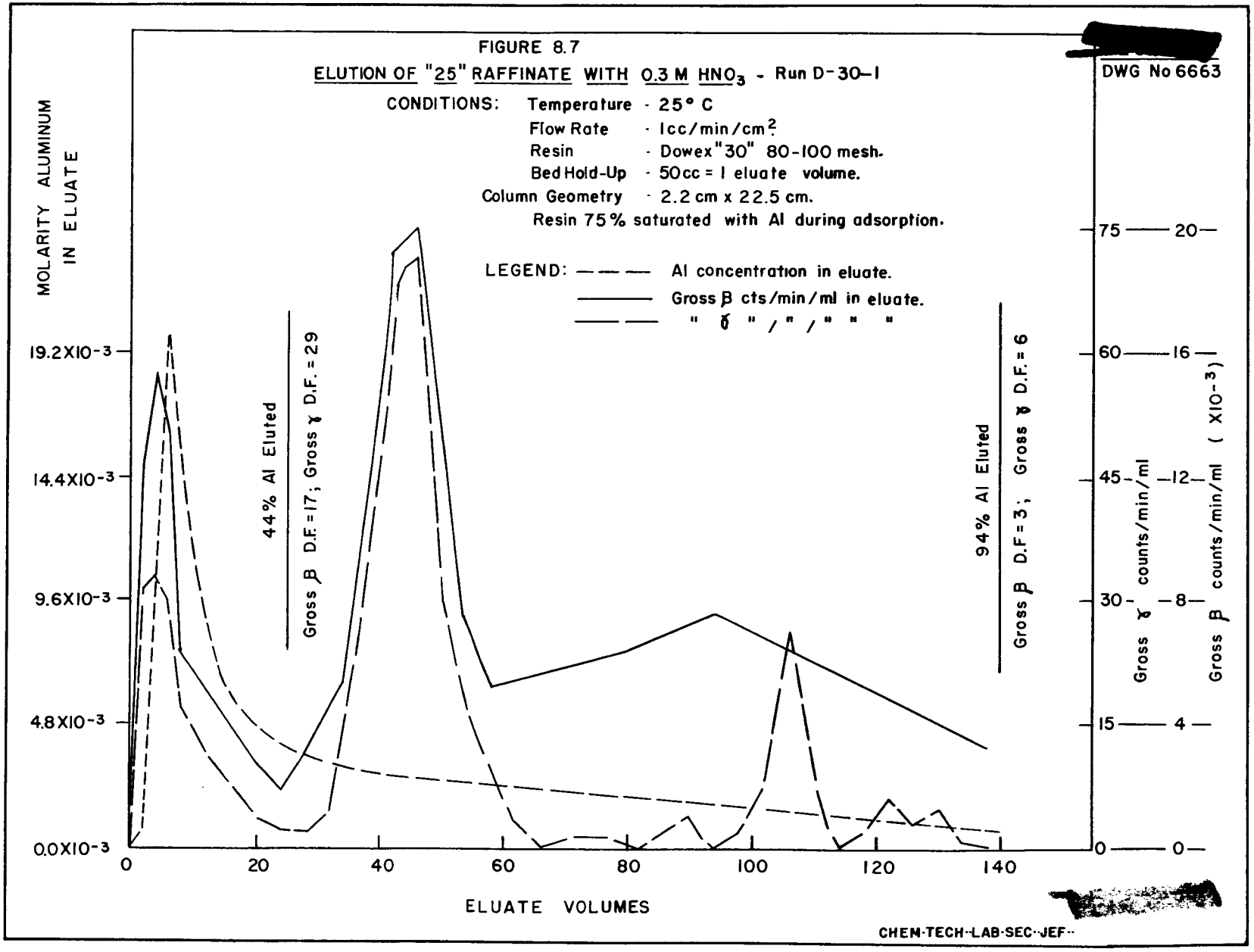
FIGURE 8.7
ELUTION OF "25" RAFFINATE WITH 0.3 M HNO_3 - Run D-30-1
CONDITIONS: Temperature - 25° C
Flow Rate - 1cc/min/cm²
Resin - Dowex "30" 80-100 mesh.
Bed Hold-Up - 50cc = 1 eluate volume.
Column Geometry - 2.2 cm x 22.5 cm.
Resin 75% saturated with Al during adsorption.
LEGEND: Al concentration in eluate.
Gross β cts/min/ml in eluate.
" γ " / " / " " "
DWG No 6663
MOLARITY ALUMINUM IN ELUATE
19.2X10⁻³
14.4X10⁻³
9.6X10⁻³
4.8X10⁻³
0.0X10⁻³
44% Al Eluted
Gross β D.F. = 17; Gross γ D.F. = 29
94% Al Eluted
Gross β D.F = 3; Gross γ D.F. = 6
0
20
40
60
80
100
120
140
ELUATE VOLUMES
Gross γ counts/min/ml
Gross β counts/min/ml (X10⁻³)
75
60
45
30
15
0
20
16
12
8
4
0
CHEM-TECH-LAB-SEC-JEF

[redacted]

DWG No 6664

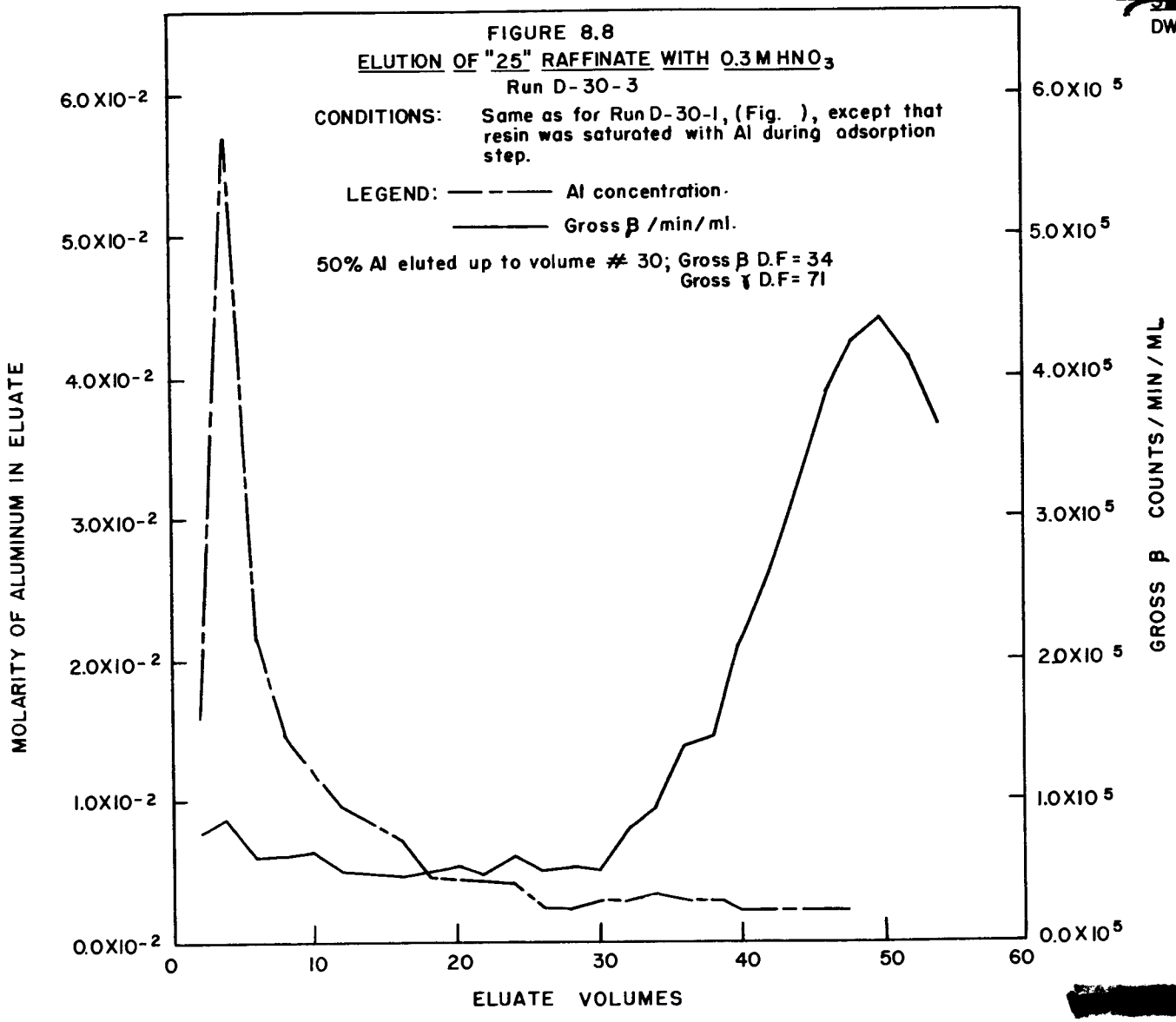

FIGURE 8.8

ELUTION OF "25" RAFFINATE WITH 0.3 M HNO_3

Run D-30-3

CONDITIONS: Same as for Run D-30-1, (Fig.), except that resin was saturated with Al during adsorption step.

LEGEND: — - - — Al concentration.

——— Gross β /min/ml.

50% Al eluted up to volume # 30; Gross β D.F = 34
Gross γ D.F = 71

[redacted]

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DWG No 6665

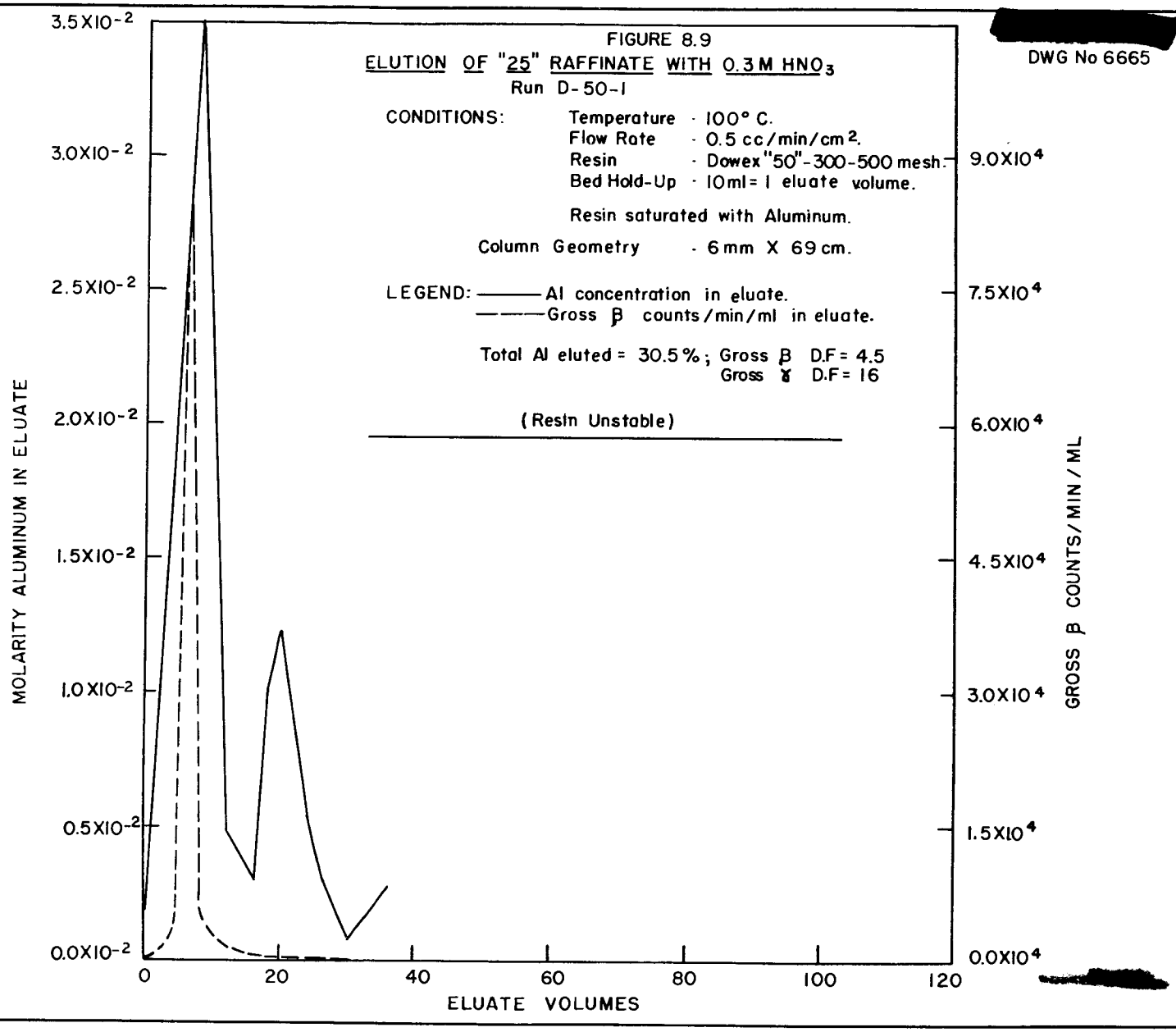

FIGURE 8.9

ELUTION OF "25" RAFFINATE WITH 0.3 M HNO_3

Run D-50-1

CONDITIONS:
- Temperature - 100° C.
- Flow Rate - 0.5 cc/min/cm².
- Resin - Dowex "50"-300-500 mesh.
- Bed Hold-Up - 10 ml = 1 eluate volume.

Resin saturated with Aluminum.

Column Geometry - 6 mm X 69 cm.

LEGEND: ——— Al concentration in eluate.
– – – Gross β counts/min/ml in eluate.

Total Al eluted = 30.5%; Gross β D.F = 4.5
Gross γ D.F = 16

(Resin Unstable)

[redacted]

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DWG No 5656

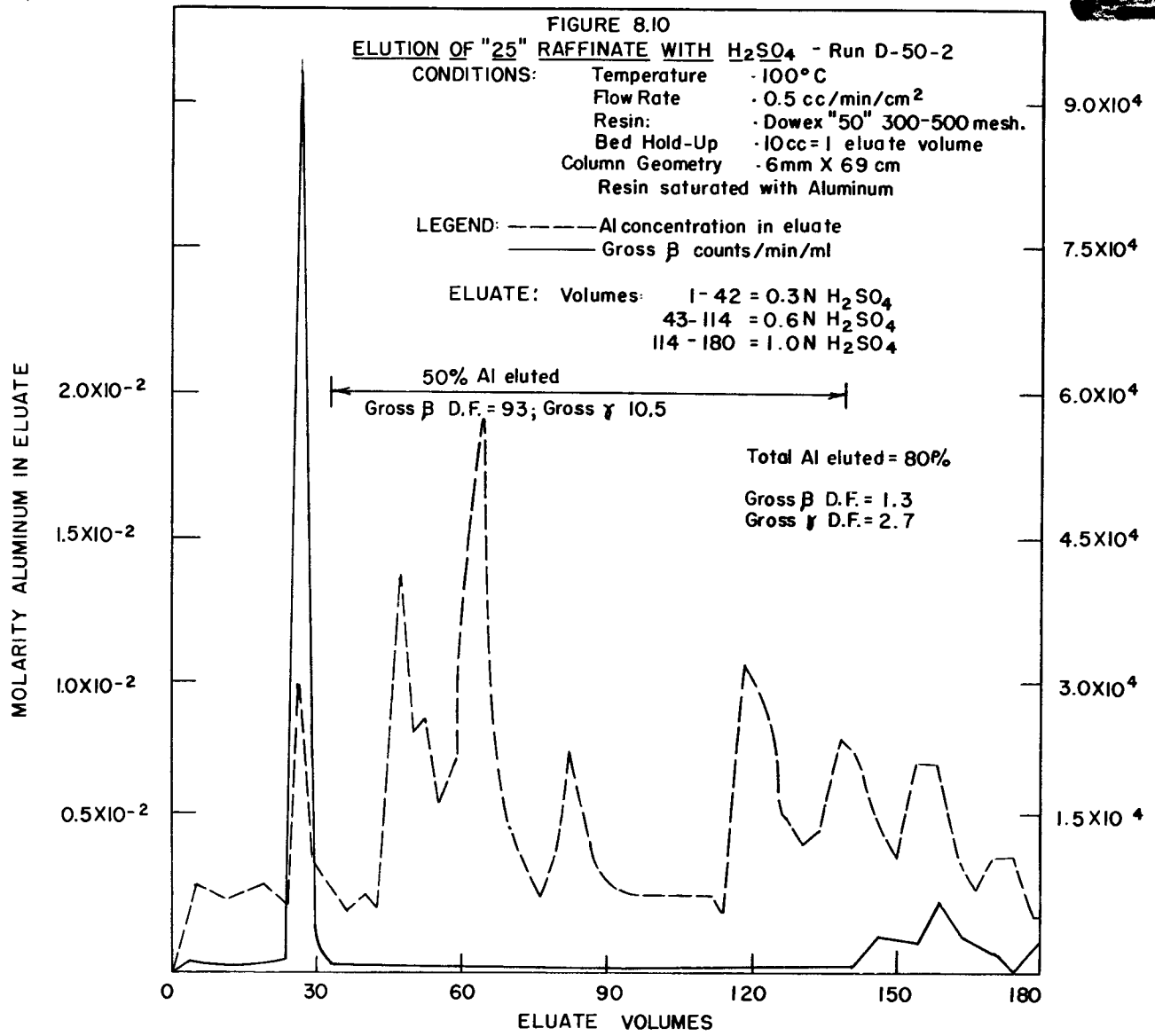

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Table 8.2

Elution of Aluminum with Nitric or Sulfuric Acid

Feed: 25 Process waste, diluted
Resin saturation with aluminum: Run 1, 75%; Runs 2 through 9, 100%

Run No.	Column Ht.	Column Dia.	Dowex Resin Type	Dowex Resin Mesh	Temp. (°C)	Flow Rate (ml/m/cm^2)	Feed Al Conc. (M)	Elutriant	Al Yield (%)	Decontamination Factors: Gross β	Gross γ	Individual Fission Products: Ru	Zr	Nb	Sr	Cs	TRE
1	22.5 cm	2.2 cm	30	80-100	25	1.0	0.1	0.3 N HNO_3	44	17	29	2	6.5	30	100	10,000	1000
2	22.5 cm	2.2 cm	30	80-100	25	1.0	0.1	Same	94	3	6	1.7	1.6	27	1.1	---	1.3
4	22.5 cm	2.2 cm	30	80-100	25	1.0	0.1	Same	56	34	71	---	---	---	---	---	---
3	69 cm	0.6 cm	30	80-100	100	2.0	0.1	Same	Resin unstable								
5	69 cm	0.6 cm	50	300-500	100	0.5	0.1	Same	Resin unstable								
7	8 ft	1 in.	50	60-80	25	3.2	0.2	Same	43	19	6	7	60	15	256	3	1380
8	4 ft	1.5 in.	50	250-300	25	0.2	0.25	0.5 N HNO_3	41	10	6	5	150	9	---	3	---
9	4 ft	1.5 in.	50	250-300	90	0.4	0.25	Same	37	12	7	---	---	---	---	---	---
6	69 cm	0.6 cm	50	300-500	100	0.5	0.1	H_2SO_4*	50	93	10.5	165	40	5.6	460	950	8000
									80	2.7	1.3	---	---	---	---	---	---

*See Fig. 8.10 for elutriant acid concentrations.

decontamination factors varied from 3 to 19 and 6 to 71, respectively. Increasing the temperature in the elution step to 90 or 100°C increased the concentration of aluminum in the eluate, but the elution was still incomplete. In addition, the resin was attacked by the acid at the higher temperatures, resulting in colored effluents.

It is interesting to note that the elution peaks for aluminum and ruthenium coincided during elution with 0.3 M HNO_3 although aluminum was expected to be much more strongly adsorbed than ruthenium. It is known that the distribution coefficient for low concentrations of aluminum in 0.3 M HNO_3 is approximately 1000, indicating that the aluminum is very strongly adsorbed. It was therefore concluded that the distribution coefficient of the aluminum was greatly decreased in the waste system in question when the aluminum concentration was high. Under these conditions the aluminum is eluted with the weakly held ruthenium.

Sulfuric Acid. Sulfate does not form a strong complex with aluminum; elution with sulfuric acid is very slow and yields are low. Gross beta and gamma decontamination factors of 93 and 100, respectively, were obtained with sulfuric acid (see Table 8.2). The addition of sulfate ion is objectionable because it increases the bulk for waste storage. However, the $Al_2(SO_4)_3$ produced can be decontaminated further (see Sec. 9.3).

Sulfuric acid (0.3 N) was found to elute aluminum much more slowly than 0.3 N HNO_3. Only ~10 per cent of the aluminum was eluted by the first 30 column volumes of 0.3 N H_2SO_4. Increasing the H_2SO_4 concentration to 0.6 and 1.0 N did not bring the elution rate to an acceptable level. The high activity peak (see Fig. 8.10) at 27 column volumes of elutriant was 63 per cent cesium and 35 per cent zirconium. The overall beta and gamma decontamination factors of aluminum elution with sulfuric acid were 2.7 and 1.3, respectively, indicating that elution with sulfuric acid is impractical.

9.0 INVESTIGATION OF ALTERNATE METHODS OF ALUMINUM WASTE DECONTAMINATION

9.1 Breakthrough Method

The breakthrough method was investigated for aluminum decontamination. It consisted in passing diluted 25 Process aluminum waste through a resin column until the resin was saturated and the excess aluminum broke through the column. An ion having a greater affinity for the resin than aluminum would be expected to remain on the column and thus be separated from the aluminum in the breakthrough solution. Results indicated that all the anions, part of the radiocolloids of ruthenium, zirconium, and niobium, and any activity more weakly held than aluminum passed through the column

with the aluminum. As more feed passed through, more activity was displaced from the resin by the developing action of the aluminum. Cesium was displaced first, followed by strontium, barium, and finally the rare earths. About 80 per cent of the total beta and 50 per cent of the total gamma activities were separated from the aluminum by this process. A follow-up scavenging of the aluminum breakthrough solution with hydrous ferric oxide and manganese dioxide removed 80 to 90 per cent of the remaining gamma activity, giving an overall gross decontamination factor of about 10 for both beta and gamma (see Table 9.1).

The experiments were carried out in semi-pilot-plant scale columns ranging from 3 ft height by 1 in. diameter to 8 ft height by 3 in. diameter. Dowex-50 resin was used. The feed solution was diluted to 0.1 M aluminum so that the mass effect of the aluminum ion would cause less movement of the activity bands toward the bottom of the column and actual breakthrough of the more weakly held activities.

Although the method was shown to be unsuitable for 25 Process aluminum waste decontamination owing to the low decontamination factors obtained, the experiments were useful in obtaining information regarding the relative exchange potentials of the fission products in the presence of a high concentration of aluminum. For example, it had been assumed that the aluminum would be held strongly by the resin because of its small ionic radius and high charge. However, it was found that aluminum displaced cesium very slowly from the resin, although the cesium is only singly charged and has a large ionic radius. To explain this phenomenon, it was assumed that the aluminum is highly hydrated and thus its effective ionic radius is increased and its exchange potential is lowered.

9.2 Precipitation

Precipitation of the aluminum as the hydroxide achieves practically no decontamination or volume reduction. In precipitation experiments, decontamination factors of the order of 1 were obtained, and the bulk of the hydroxide was found to be almost as large as that of a saturated solution of aluminum nitrate. The precipitate was very difficult to filter or centrifuge. It is possible to precipitate thorium hydroxide in a compact granular form by raising the pH of the solution by the slow decomposition of urea in the presence of certain anions such as formate.[9] Precipitation of aluminum from the waste solution by this method was successful in reducing the volume of the precipitate and in simplifying any necessary mechanical processing, but no decontamination was achieved. Because of the inactive chemicals added to the waste, the process is not considered attractive.

9.3 Recrystallization

An efficient process for the separation of aluminum sulfate from iron sulfate by recrystallization of the aluminum salt from an aqueous

Table 9.1

Beta and Gamma Decontamination by Breakthrough Method

Feed: diluted 25 Process aluminum waste
Resin: Dowex-50, 60 to 80 mesh, initially in acid form
Temperature: 25°C
Aluminum yield (%): 100 x g of Al in effluent ÷ g of Al in feed

Run No.	Column Ht. (ft)	Column Dia. (in.)	Al in Feed (M)	Feed and Elutriant Flow Rate (ml/m/cm²)	Al Yield (%)	Decontamination Factors: Gross, Resin β	Gross, Resin γ	Gross, Scavenging β	Gross, Scavenging γ	Individual Fission Products, Resin: Ru	Zr	Nb	Cs	Sr	Ba	TRE	Scavenging*: Zr	Nb	Cs
200	8	3	0.1	7.5	74	3.3	3.6												
					82	2.6	5.3												
					88	2.7	4.5												
					90	1.7	3.0												
					92	1.6	2.7			2.5	10	5	6	500	>50	330			
					93	1.6	3.4												
					94	1.4	3.0												
					95	1.1	2.0												
202	3	1	0.1	2.4	75	24.0	4.5												
					79	6.3	1.7			2.5	1.3	1.6	1.7	14	>62	225			
					85	5.7	1.4												
					91	4.0	1.8	1	4.5	2.5	1.4	1.5	1.03	1.4	>26	30	93	3	1.2
203	3	1	0.2	2.4	70	9.3	2.0												
					90	3.5	1.4												
					92	3.8	2.2												
					95	4.1	2.2												
					96	3.8	1.9	1	9.0										
204	8	1	0.1	2.4	44	13.0	2.0												
					72	6.0	1.6												
					85	10.0	3.0			3.2	1.7	2.0	1.3	1.7		28			

*Run 202 eluate was scavenged at pH 12 in presence of sodium hydroxide with 0.1% Mn as MnO_2 and 0.01% Fe as $Fe(OH)_3$. Run 203 eluate scavenging was the same except that the manganese precipitation was omitted.

ethyl alcohol solution has been reported.[10] The method was studied on 25 Process aluminum waste to which sulfuric acid had been added in excess of the stoichiometric amount of aluminum present. Gross gamma decontamination factors ranging up to 20 were obtained. Recrystallization from a 0.5 to 1.0 M aluminum solution gave the highest decontamination factors. The individual decontamination factors for ruthenium, zirconium, and niobium ranged from 200 to 500, while those for cesium, strontium, and rare earths were in the range 5 to 40. When the recrystallization was carried out on a sulfuric acid eluate from the ion-exchange process, from which strontium, cesium, and rare earths had been removed by the resin, an additional gross gamma decontamination factor of 7 to 18 was obtained.

An important objection to this process is the large amount of sulfuric acid that must be added to the waste solution. This would interfere with the recycling of the aluminum and would materially increase the bulk of waste to be stored.

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A. H. Kibbey

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REB:IRH:AHK:ig

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Appendix 1. SUMMARY OF DATA OBTAINED IN VARIOUS RUNS

Information obtained in various runs is summarized in the tables in this appendix. The cross-linkage of the Dowex-50 resin used in the experiments reported throughout this work was reported by the Dow Chemical Company to be approximately 10 per cent. No value of the cross-linkage of Dowex-30 is known.

Table A1.1

Decontamination of 25 Process Aluminum Waste, Runs 210-214

Feed: 25 Process waste, diluted to 0.25 M aluminum
Scavenger: None
Resin: Dowex-50, initially in acid form, finally 100% saturated with aluminum
Column specifications: 88 cm (34 in.) height, 3.8 cm (1.5 in.) diameter
Aluminum yield: 96 to 99% in all cases
In each run, before the elution was started, the column was washed with 1500 ml of complexing agent in the same concentration as in the feed

Run No.	Resin Size (mesh)	Temp. (°C)	Feed and Elutriant Flow Rate (ml/m/cm^2)	Complexing Agent in Feed	Elutriant	Decontamination Factors							
						Gross			Individual Fission Products				
						Puα	β	γ	Zr	Ru	Cs	Sr	TRE
210	250-300	25	0.2	0.016 M oxalic acid	0.4 M oxalic acid	47	25	7	18	3	620	---	1100
211	250-300	90	0.3	Same	Same	---	15	2	---	---	---	---	---
212	60-80	25	0.5	Same	Same	150	190	10	40	8	925	21,000	2900
213	60-80	25	0.5	0.05 M HF	0.5 M HF	---	20	2	---	---	---	---	---
214	60-80	25	0.5	Same as Run 210	0.5 M NH_4F	---	130	20	220	20	10	7,000	3500

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Table A1.2

Decontamination of 25 Process Aluminum Waste, Runs 1 through 6

Feed: 25 Process waste diluted before scavenging to 0.1 M Al, pH 2.5
Resin: Dowex-50, 60 to 80 mesh, initially in acid form
Column dimensions: 16.5 cm height, 1.4 cm diameter; aluminum capacity of column in presence of 0.016 M oxalic acid, 620 mg
Temperature: 25 ± 1°C
Elutriant: 0.4 M oxalic acid
Flow rates: 1 ml/m/cm^2
Aluminum yield: 97 to 99%

Run No.	Al in Feed (mg)	Scavenger*	Complexing Agent in Feed	Wash Solution	Decontamination Factors									
					Gross				Individual Fission Products, Overall					
					Scavenging		Overall							
					β	γ	β	γ	Zr	Ru	Cs	Sr	Nb	TRE
1	1.2x620	None	0.032 M oxalic acid	60 ml H_2O	---	---	20	10.5	58	3.8	6.8	176	3.9	1,260
2	1.3x620	0.01% Fe +0.01% Mn	Same	Same	1.3	11.5	32	39	1150	250	2.0	50	90	1,700
3	0.75x620	0.05% Fe +0.01% Mn	0.016 M oxalic acid	300 ml 0.016 M oxalic acid	2.0	13.0	56	42	126	220	4.0	1,000	460	250
4	1.2x620	Same	Same	Same	1.0	8.0	27	43	790	550	5.3	---	106	---
5	1.5x620	Same as Run 2	Same	Same	1.3	7.8	49	73	950	538	6.3	---	140	---

*Iron and manganese added as hydrous oxides.

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Table A1.3

Decontamination of 25 Process Aluminum Waste, Runs D-50-14 through D-50-22

Feed: 25 Process waste diluted before scavenging to 0.5 M aluminum, pH 2.5
Resin: Dowex-50, 60 to 80 mesh, initially in acid form
Column specifications: 106 cm height, 6 mm diameter; aluminum capacity, 578 mg in presence of 0.016 M oxalic acid, 610 mg in absence of oxalic acid
Temperature: 25 ± 1°C
Scavenger: 0.01% Fe as $Fe_2O_3 \cdot nH_2O$, 0.01% Mn as $MnO_2 \cdot nH_2O$; β d.f. = 1.6, γ d.f. = 7.6
Aluminum yields: approximately 97 to 99%

D-50 Run No.	Al in Feed (mg)	Complexing Agent in Feed	Wash Solution	Al Lost in Wash (%)	Conc. of Oxalic Acid in Elutriant	Decontamination Factors				
						Gross				
						Resin Column		Overall		
						β	γ	Pu α	β	γ
14	1x610	None	300 ml 0.016 M oxalic acid	1.2	0.4 M pH 0.78	66	8.6	1.2	106	65
15	1x578	0.016 M oxalic acid	60 ml H_2O	1.0	Same	174	15.5	1.0	278	118
16	1.5x578	Same	Same	2.3	Same	86	11.4	2.2	138	87
17	0.2x578	Same	Same	1.6	Same	270	20	1.6	432	152
18	1x578	Same	Same as Run 14	15	Same	76	8	2.1	122	61
19	0.2x578	Same	Same	1.5	Same	850	46	1.4	1.4×10^3	350
20	0.9x578	Same	45 ml 0.016 M oxalic acid	0.1	Same	2.0×10^3	192	1.4	3.3×10^3	1.5×10^3
21	0.9x578	Same	Same	0.5	0.4 M pH 2.5	For 96% Al Elution: 1.6×10^3	2.0×10^3	---	2.6×10^3	$1.[illegible] \times 10^4$
						For 99% Al Elution: 1.2	1.7	---	1.9	[illegible]
22	0.9x578	Same	Same	0.5	0.72 M pH 0.78	1.4×10^3	155.3	1.3	2.2×10^3	1.2×10^4

For D.F.'s for Zr, Ru, Cs, Sr, Nb, and TRE, see next page

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Table A1.3 (Continued)

D-50 Run No.	Decontamination Factors Individual Fission Products, Overall					
	Zr	Ru	Cs	Sr	Nb	TRE
14	107	143	6.6	1.4×10^4	103	2.0×10^4
15	426	143	16.8	5.2×10^3	98	8.4×10^3
16	134	247	7.6	3.9×10^3	50	4.4×10^4
17	---	130	150	---	10.7	---
18	258	351	6.7	4.7×10^3	212	2.8×10^3
19	187	$\sim 6.8 \times 10^3$	$\sim 5.6 \times 10^3$	$\sim 1.4 \times 10^4$	61	$\sim 1.1 \times 10^5$
20	1.8×10^3	377	2.5×10^4	$\sim 6.3 \times 10^4$	2.4×10^3	3.6×10^4
21	For 96% Al Elution:					
	---	---	---	---	---	---
	For 99% Al Elution:					
	86	143	1.3	2.7×10^3	266	2.8
22	1.5×10^3	293	1.2×10^3	6.3×10^3	117	5.0×10^4

[redacted]

Table A1.4

Decontamination of 25 Process Aluminum Waste, Runs 216 through 221

Feed: diluted 25 Process waste
Resin: Dowex-50, initially in acid form, 60 to 100 mesh
Column dimensions: 88 cm (34 in.) height, 3.8 cm (1.5 in.) diameter
Flow rates: 1 ml/m/cm^2
Temperature: 25 ± 1°C
Scavenger: 0.01% Fe as $Fe(OH)_3$; 0.01% Mn as MnO_2, pH 2
Elutriant: ~ 0.4 M oxalic acid
Aluminum yield: 97 to 99%
In each run, before the elution was started, the column was washed with 1500 ml of complexing agent in the same concentration as in the feed
No plutonium decontamination was obtained in the scavenging step

Run No.	Resin Size (mesh)	Resin Satn. with Al (%)	Feed Al Conc. (M)	Complexing Agent in Feed	Decontamination Factors											
					Gross					Individual Fission Products						
					Scavenging		Overall			Scavenging		Overall				
					β	γ	Pu α	β	γ	Zr	Ru	Zr	Ru	Cs	Sr	TRE
216	60-80	100	0.25	0.016 M oxalic acid	2	10	---	600	>200	63	140	760	86	1000	3300	420
217	60-80	100	0.5	Same	2	5	5	600	>300	190	26	2200	290	90	500	1100
219	100-200	92	0.5	0.008 M oxalic acid	1	7.5	---	3000	600	8	33	---	---	---	---	---
220	100-200	100	0.5	Same	1	7.5	1.2	880	470	---	---	---	---	---	---	---
221	100-200	94	0.25	Same + 0.05% thio-urea	2.5	7	380*	1200	>600	---	---	6900	620	3600	1160	1450

*Probably error in alpha determination.

[redacted]

Table A1.5

Distribution of Fission Products in Adsorption Step Effluent and Oxalic Acid Wash

Feed: diluted 25 Process aluminum waste, previously scavenged with $Fe(OH)_3$ and MnO_2
Resin: Dowex-50, 60 to 80 mesh
Column dimensions: 106 cm height, 6 mm diameter
Aluminum capacity: in presence of 0.016 M oxalic acid, 578 mg; in absence of oxalic acid, 610 mg
Flow rates: 1 $ml/m/cm^2$
Column wash: 300 ml of 0.016 M oxalic acid

Note: In Runs D-50-16, 17, and 18, 45 ml of water was combined with the adsorption effluent

D-50 Run No.	Al in Feed (mg)	Complexing Agent in Feed	Contaminant (% of total in feed)													
			Adsorption Effluent							Column Wash						
			Ru	Zr	Nb	Cs	Sr	TRE	Pu	Ru	Zr	Nb	Cs	Sr	TRE	Pu
14	1 x 610	None	44	3.3	1.6	0.0089	---	---	---	35	7	4.1	4.2	---	---	---
15	1 x 578	0.016 M oxalic acid	(38)*	10	2.6	0.04	---	---	---	(91)*	4.9	6.8	6.0	0.02	0.02	---
16	1.5 x 578	Same	(67)*	8	2.9	27	---	---	---	(51)*	15.6	13.5	13.1	<0.03	<0.005	44.5
17	0.2 x 578	Same	---	19	3.4	---	---	---	---	---	---	---	---	---	---	---
18	1 x 578	Same	---	---	5.8	---	0.02	<0.03	---	18.5	3.6	1.3	2.3	---	0.002	0.9
19	0.2 x 578	Same	57	---	---	<0.03	---	---	7.8	27	---	---	0.07	---	---	2.4

*Parentheses indicate questionable accuracy.

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