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PILOT PLANT DEVELOPMENT STUDIES
OF A CONTINUOUS PROCESS FOR RECOVERING URANIUM FROM
NICHROME FUELS

H. V. Chamberlain

**PHILLIPS
PETROLEUM
COMPANY**

ATOMIC ENERGY DIVISION

**NATIONAL REACTOR TESTING STATION
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PILOT PLANT DEVELOPMENT STUDIES
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A B S T R A C T

Pilot plant studies were conducted to develop a continuous process for recovering uranium from nichrome fuels (HTRE). The process consisted of dissolution of fuel in mixed HCl-HNO_3 solution, removal of the chloride ion by stripping with HNO_3 in a packed column, and then recovery of the uranium by TBP solvent extraction. Recovery of uranium from nichrome fuels at satisfactory rates and efficiencies can be obtained by this process. Reactor fuels containing large amounts of silica may present a solids problem during chloride removal. Titanium is a suitable material of construction for the dissolver, stripping column, and their associated equipment (feed, off-gas and product vessels). Stainless steel equipment is suitable for the solvent extraction system.

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I. SUMMARY

A continuous process was developed and demonstrated for recovering uranium from nichrome fuels. The process consists of dissolution in dilute solutions of mixed HCl-HNO_3 , removal of chloride ion from dissolver effluent by stripping with nitric acid in a packed column, and uranium purification by TBP extraction.

Variables studied in the dissolution step were acid feed rate and fuel element height. Acid feed was 1.0M hydrochloric acid and 6.0M nitric acid, and fuel element heights were 3.5 and 5.5 feet. The effects of increasing the ratio of the acid feed rate to fuel surface area (FSR) from 0.004 to 0.024 cm/min were found to be as follows:

1. Dissolution rate increased from 0.66 to 1.8 mg/(cm²)(min).
2. Metal concentration of the dissolver effluent decreased from 185 to approximately 50 grams per liter.
3. Off-gas rate increased from 0.07 to 0.3 scfm.
4. Acid consumption increased from 2.2 to 3.2 moles of acid per mole of fuel dissolved.

Dissolver off-gas consisted of nitrogen oxides and nitrosyl chloride; essentially all the chloride in the dry off-gas, before and after the downdraft condenser, was present as nitrosyl chloride. The chloride absorption efficiency of the downdraft condenser increased from 15.4 to 84.9 per cent as the final off-gas to condensate ratio (ft³/l) decreased from 13 to 1. Residues in the dissolver effluent consisted of the oxides of chromium, nickel, niobium, and silicon at total concentrations of up to 2.8 grams per liter. Negligible quantities of uranium were found in the residues. Average particle diameter of the residue was 35 microns. Chemical stability studies on dissolver effluents indicated that approximately 20 per cent of the niobium remains in solution after the dissolution step and that it will precipitate as a gelatinous mass on reheating the effluents. Some silicon also remains in solution.

Removal of the chloride ion is performed by adjusting the dissolver effluent to be approximately 7.5M in total acid content and then contacting the resulting solution with concentrated nitric acid vapors in a packed stripping column. Increasing the molar vapor/liquid ratio (V/L) in the column from 0.27 to 0.67 produced the following results:

1. Chloride concentration in the uranium-bearing stream decreased from 16,000 to 2,000 parts per million (ppm).
2. The chloride removal performance (conc. of chloride in feed/conc. of chloride in kettle) increased from 5 to 12,500.
3. Height of a transfer unit decreased from 3.2 to 1.0 feet.

A molar V/L of less than 0.4 is required to produce an overhead product stream that can be recycled as a dissolvent. Increasing the packing height from 4.0 to 9.5 feet increased the chloride removal performance from 20 to 350 at a V/L of 0.4. Stripper off-gas contained nitrogen oxides and nitrosyl chloride. Again, essentially all chloride in the off-gas was present as nitrosyl chloride. The downdraft condenser used had 13.5 ft² of heat transfer surface and was very efficient in removing chloride from the off-gas. Increasing the stripper overhead product rate from 25 to 110 cc/min increased the off-gas rate (non-condensables) from 0.05 to 0.12 scfm. Flooding capacity data of the four-inch diameter column agreed satisfactorily with experimental data obtained with other systems.

Limited solvent extraction data obtained indicate that very few operational problems should be encountered when processing solutions containing dissolved nichrome. Uranium waste losses were somewhat high, 0.02 per cent when using five volume per cent TBP in Amsco 125-90W. These high waste losses were due to an insufficient number of stages in the column. Presence of aluminum in the feed solution (7 to 37 grams per liter) increased uranium waste losses from 0.06 to 0.15 per cent.

Titanium is a suitable material of construction for the dissolver and stripping column systems, and stainless steel is suitable for the solvent extraction system.

II. INTRODUCTION

Early in 1954, Phillips Petroleum Company was assigned the task of developing a wet chemical method for the recovery of uranium from nichrome fuel. This nichrome fuel was then being used by General Electric in their ANP program. A description of this fuel is given in Appendix A. Scoping tests ⁽¹⁾ by Dunleavy of G. E. indicated that both a mixed HCl-HNO₃ solution and a perchloric acid solution would be suitable solvents for nichrome. The mixed HCl-HNO₃ system was chosen by Phillips for further study because it is safer to handle than is perchloric acid.

A suitable process using mixed HCl-HNO₃ was visualized by Slansky, et al. ⁽²⁾ to consist of the following steps: dissolution in mixed HCl-HNO₃, adjustment of the dissolver effluent with concentrated nitric acid for removal of the chloride ion by a batch boil-off procedure, and then adjustment of the resulting solution with water for uranium purification via solvent extraction. Since existing solvent extraction systems are constructed of stainless steel, removal of the chloride ion is required to minimize corrosion. Work conducted by Walters, et al. ⁽³⁾ indicated that in nitric acid solutions (7.8M or greater) corrosion rates of the 300 series stainless steels were less than 0.007 ipy with chloride concentrations up to 0.1 w/o (1000 ppm); solutions with 2.6M HNO₃ could contain as much as 0.02 to 0.05 w/o chloride. A goal of 25 ppm chloride in the stripping column product was chosen. A large safety factor in concentration was then available to allow for any maloperation of equipment that might occur.

Dissolution studies were initiated at the Idaho Chemical Processing Plant in late 1954. Initial work ⁽²⁾ was on the development of a batch process for the nichrome fuel, but it was soon recognized that a continuous process, although not required from a plant capacity standpoint, seemed to offer several advantages such as process control and criticality control. Also, work at Oak Ridge National Laboratory on the Darex process for stainless steel fuels ⁽⁴⁾ indicated that the processes were very similar for the two fuels and that both could use the same system. Therefore, very limited pilot plant work was done on developing a batch process.

Corrosion work by Klemens ⁽⁵⁾ indicated that titanium would be a suitable material of construction for the mixed HCl-HNO₃ system. Therefore, titanium, as well as Pyrex* glass and polyvinyl chloride, was used in construction of the pilot plant equipment.

This report presents the results of the pilot plant effort to obtain process design information for the recovery of uranium from nichrome fuels.

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PART I

BATCH STUDIES

PART I

BATCH STUDIES

The purpose of this portion of the program was to obtain process design information for the batch process reported by Roberts, et al. (6). This process consisted of batch dissolution of the fuel elements using mixed HCl-HNO_3 , followed by removal of the chloride ion using a batch buildown procedure. This last step was also performed in the dissolver.

I. DISSOLUTION

A. Objectives

The objectives of this portion of the study were to determine the controllability of the batch dissolver during dissolution of the fuel, to determine the efficiency of the downdraft condenser for chloride absorption, and to characterize the type of solids produced.

B. Equipment and Procedures

1. Equipment

The dissolver system consisted of a batch dissolver, downdraft condenser, pump for returning condensate to the dissolver, and rotameters for measuring condensate and off-gas flows. Figure 1 is a schematic equipment flowsheet of the dissolution equipment.

The dissolver was made of a four-foot length of four-inch glass pipe equipped with titanium end plates and lines. Off-gas and condensate lines were of Pyrex glass pipe or PVC tubing. The condenser had 1.37 square feet of heat transfer surface. The off-gas line from the dissolver to the condenser was heated to prevent condensation of water vapor ahead of the condenser.

2. Procedures

A charge of 277 grams of fuel was used for each run. This consisted of 232 grams of nichrome-uranium oxide meat and 45 grams of nichrome cladding material containing one per cent niobium. Two liters of acid (40 volumes of 37 weight per cent HCl , 60 volumes

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Figure 1

Schematic Equipment Flowsheet
of Batch Dissolution System

of 70 weight per cent HNO_3 , 90 volumes of H_2O) were used as dissolvent for each run.

Initiation of the reaction was spontaneous, violent ebullition occurring within five minutes after introduction of dissolvent; however, the reaction quickly subsided to a gentle boiling. After the initial rapid reaction, low-pressure steam (10 to 15 pounds) was used to maintain a gentle reaction, with dissolver temperatures varying between 175 and 185°F. Samples of condensate and off-gas were taken periodically throughout a total run time of approximately four hours.

C. Results

1. Dissolver Operation

The dissolver feed composition was nominally 7.5M acid, 2.5M chloride, and 5.0M nitrate. The actual concentrations varied between 6.79 to 7.66M acid, 2.34 to 2.75M chloride, and 3.76 to 5.07M nitrate. Dissolver product samples had a composition range of 0.1 to 1.36M acid, 1.03 to 2.38M chloride, and 2.50 to 3.57M nitrate. The data show that with the proper dissolution conditions, i.e., a low boiling rate, the losses of chloride and nitrate can be minimized. With the high acid losses, the addition of a large quantity of nitric acid will be required before the dissolver product solution can be sent to the stripper for chloride removal. Table 1 summarizes the feed and product solutions analyses.

2. Dissolver Off-Gas

The maximum off-gas rate past the downdraft condenser was approximately 0.5 standard cubic foot per minute during the initial surge. The fuel charged to the dissolver to begin the run was approximately one square foot in surface area; consequently, off-gas rates also express cubic feet per minute per square foot of fuel. After the initial surge, i.e., during the gentle boiling period, the off-gas rate was a maximum of about 0.2 cubic foot per minute, but fluctuated between 0.2 and zero, averaging probably less than 0.1 cubic foot per minute.

TABLE 1
DISSOLVER FEED AND PRODUCT ANALYSES FOR BATCH
DISSOLUTION OF NICHROME-URANIUM OXIDE MEAT

Feed: 40-60-90 volume mixture of - 37 per cent HCl - 70 per cent HNO_3 - H_2O Two liters per batch 277 grams fuel per batch Alloy - 82 weight per cent Nichrome V					
Run No. and Sample	H^+ (N)	Cl^- (mg/ml)	NO_3^- (N)	Sp. Gr.	Remaining Volume (cc)
4 and 6 DP (1)	(2)	(2)			
4 DP (1)	0.12	37.66	3.11	1.336	1600
6 DP	0.58	65.5	3.57	---	1800
7 DP	7.25	100.3	4.91		
7 DP	0.43	79.0	3.40	---	1750
8 DP	7.56	94.7	4.61		
8 DP	0.19	79.0	2.50	1.328	~1800
9 DP	(2)	(2)	(2)		
9 DP	0.234	46.5	3.36	1.395	1350
10 DP	6.79	85.5	3.76		
10 DP	0.09	82.5	3.36	1.415	1325
12 DP	7.66	93.0	5.069		
12 DP	1.38	84.5	3.56	1.335	1700 (3)
(1) DP-Feed, DP-Product					
(2) Samples not available. Nominal composition assumed.					
(3) 1800 cc feed dissolvent, instead of 2 liters.					

A suitable analytical technique was not available at the time of this work to enable direct analysis for all components of the off-gas. The method used reported the acidic components as either chloride or nitrate; some of the non-acidic components, such as NO which is easily oxidized, could also appear as an acidic component.* This method has serious limitations if certain species of chlorine (i.e., nitrosyl chloride) are present.

Analyses of the off-gas showed that the chloride concentration in the off-gas stream past the condenser varied between 0 and 47 mole per cent (air-free basis), and was dependent on the reaction rate or ebullition rate. In most cases, off-gas samples were not obtained during the initial spontaneous reaction, and gas evolution after this time was dependent on the steam pressure applied to the dissolver coil. Unfortunately, the manner in which the off-gas composition changes as the run proceeds cannot be determined from the data available. With an average off-gas rate after the condenser of 0.1 cubic foot per minute, the loss of chloride was 0.3 mole for a four-hour run, or 12 per cent of the total chloride.

The nitrate concentrations in the final off-gas stream ranged from 15 to 97 mole per cent which would give a loss of 0.6 mole per run. The nitric and nitrous oxide concentrations ranged from 0 to 82 and 1 to 11 mole per cent, respectively. This would give up to 0.5 mole of nitric oxide and 0.1 mole of nitrous oxide loss per run. The total loss of nitric acid (not absorbed in the condenser) was about 1.3 moles per run, or 26 per cent of the total nitrate. Off-gas data for these runs are summarized in Table 2.

TABLE 2
OFF-GAS ANALYSES FOR BATCH DISSOLUTION
OF NICHROME-URANIUM ALLOY

241 grams fuel per batch 2 liters (40-60-90) mixtures of 37 per cent HCl - 70 per cent HNO_3 - H_2O Reaction temperature - $135 \pm 5^\circ \text{F}$ Average off-gas rate - 0.1 to 0.2 ft^3/min standard conditions Alloy - 82 weight per cent Nichrome V					
Run No. and Sample	Time, Hrs from Beginning of Run	Off-Gas Analyses (Air-Free Basis) Mole Per Cent*			
		Cl^-	NO_3^-	NO	H_2O
6-1	1.0	11.2	65.7	0	3.2
-2	3.25	47.0	47.0	0	6.0
7-1	0.33	29.4	59.9	8.8	1.9
-2	2.75	24.6	22.7	51.7	1.0
8-1	1.1	6.6	52.2	39.6	1.7
-2	2.2	0.5	96.7	0.6	2.2
-3	3.5	5.2	80.1	3.6	11.1
9-1	0.7	23.3	33.2	35.8	1.7
-2	0.25	21.8	61.5	0.8	9.9
-3	2.0	19.6	55.0	23.2	2.2
-4	3.25	41.7	50.3	2.0	6.0
-5	4.0	46.8	30.3	7.4	15.4
10-1	0.25	21.0	68.2	0.7	10.1
-2	1.25	6.8	47.3	39.3	6.6
-3	3.5	3.4	85.6	2.2	8.8
12-1	1.4	36.2	39.6	22.6	1.6
-2	3.25	7.4	14.9	76.7	1.0
-3	4.5	0.2	16.0	81.8	2.0

* Off-gas samples were taken at the condenser exit.

*The off-gas sample is contacted with potassium hydroxide and the resulting solution is analyzed by wet chemical methods for chloride and nitrate. The remaining gases are analyzed by the mass spectrometer.

3. Downdraft Condenser Evaluation

The condensate rates from the downdraft condenser averaged 50 to 60 milliliters per minute with acid and nitrate concentrations averaging about 0.2M each. The chloride concentration averaged about 2 grams per liter (0.05M). As was expected, the amount of chloride removed from the dissolver depended on how vigorously the reaction was maintained. Run 12 was made with a very gentle reaction in the dissolver, and the total chloride loss amounted to about 0.2 mole for the four hours of run time. In comparison, during Run 4 there were several instances of vigorous boiling, and the chloride loss was approximately 1.5 moles for an equivalent run time. Table 3 presents these data.

The absorption efficiency of the condenser for the chlorides and the oxides of nitrogen was determined by assuming that all of the chlorides and nitrates in the off-gas downstream of the condenser were absorbable. By dividing the total moles of chlorides and nitrates in the condensate by the total moles in the condenser (summation of the condensate and off-gas streams analyses), the efficiency can be determined. The off-gas rates were assumed to be 0.1 cubic foot per minute and the condensate rate 60 milliliters per minute.

The overall efficiencies determined by the above method ranged between 43.5 and 99.5 per cent and were dependent upon the vigorousness of the reaction. The smoothest operation was achieved in Run 12, i.e., uniform gentle boiling after the initial surge. This run also had the lowest chloride, nitrate, and acid losses, and, consequently, the best condenser efficiency (greater than 99 per cent). Table 4 presents these data.

TABLE 3
CONDENSATE ANALYSES FOR BATCH DISSOLUTION
OF NICHROME-URANIUM ALLOY

Downdraft condenser 1.37 ft ² transfer surface Cooling water flow - 5 gpm Condensate temperature - 65° F				
Run No. and Sample	Time, Hrs from Beginning of Run	Cl ⁻ (mg/ml)	NO ₃ ⁻ (N)	H ⁺ (N)
4-1	0.85	1.59	0.247	0.32
-2	3.5	0.026	0.022	0.02
6-1	1.0	12.50	0.68	0.75
7-1	3.5	19.75	1.21	1.28
-2	4.5	4.30	0.31	0.17
8-1	1.2	1.046	0.245	0.31
-2	2.3	0.599	0.066	0.08
-3	3.5	0.418	0.066	0.10
9-1	1.25	2.00	0.358	0.45
-2	1.75	0.420	0.127	0.163
-3	2.0	0.345	0.076	0.06
-4	2.5	0.235	0.046	0.01*
-5	3.1	0.0185	0.048	0.02*
-6	3.75	0.160	0.03*	0.02
10-1	0.5	0.47	0.175	0.19
-2	1.0	0.175	0.060	0.07
-3	1.5	0.090	0.034	0.06
-4	2.25	0.053	0.048	0.05
-5	3.0	0.075	0.052	0.05
-6	3.75	0.052	0.055	0.06
12-1	1.5	22.00	1.039	1.72
-2	3.25	13.50	1.193	1.55
-3	4.5	14.00	0.96	1.56

*Denotes Normality of Basic Solutions

TABLE 4.
ABSORPTION EFFICIENCIES OF DOWNDRAFT CONDENSER
USED IN THE BATCH DISSOLUTION STUDIES

Heat transfer surface 1.37 ft ² Gas - mixtures of oxides of nitrogen and chlorides Cooling water temperature - inlet 48° F, outlet 52° F Gas flow - 0.1 ft ³ /minute at standard conditions Condensate flow - 60 cc/minute Condensate discharge temperature - 65° F								
Run No.	Time, Hrs After Beginning of Run	Off-Gas		Condensate		Per Cent Absorption ⁽¹⁾		
		Cl ⁻ Moles x 10 ³ (2)	NO ₂ Moles x 10 ³ (2)	Cl ⁻ Moles x 10 ³ (2)	NO ₂ Moles x 10 ³ (2)	Cl	NO ₂	Overall
8	1.1	0.170	1.73	1.77	14.7	91.2	89.5	89.7
	3.5	0.137	2.107	0.706	3.96	83.7	65.3	67.5
9	1.25	0.573	1.775	3.38	2.15	85.5	54.8	70.2
	2.0	0.515	1.45	0.58	4.66	53.1	75.9	72.4
	4.0	1.23	0.797	0.284	2.04	18.8	71.9	53.4
10	0.25	0.552	1.794	0.794	10.5	59.0	85.4	82.8
	3.5	0.069	2.23	0.152	3.24	63.1	59.0	54.3
12	3.25	0.19	0.39	23.0	72.0	99.2	99.5	99.4
	4.5	0.05	0.421	23.66	57.6	100.0	99.3	99.5

(1) Based on the assumption of complete solubility of all compounds of chlorine and nitrogen.

(2) Moles per minute.

II. CHLORIDE REMOVAL

A. Objectives

Early work ⁽²⁾ indicated that the chloride ion could easily be removed from solutions having a high nitric acid content by a batch boil-down procedure. Pilot plant studies were conducted to determine the number of additions of concentrated nitric acid and boildowns required to reduce the chloride content to an acceptable limit (25 ppm).

B. Equipment and Procedures

The chloride removal was conducted in the dissolver proper. In the chloride removal step, a volume of 70 per cent nitric acid equal to the original volume of dissolvent (two liters in most cases) was added to the dissolver and the resultant volume evaporated down to the original volume. The process was then repeated for the second boildown step. In neither step was the condensate returned to the dissolver.

C. Results

Three exploratory runs were made to determine the number of batch boildowns required to obtain a product which could be processed in the stainless extraction equipment without excessive corrosion. The first two runs had two chloride removal boildowns and the third run had a single boildown. Table 5 summarizes the data from the three runs.

The data would seem to indicate that two batch boildowns would be required to remove the chloride down to an acceptable level of 25 ppm ($\sim 0.001M$). The data for Runs 1 and 3 indicate that some conditions might exist which would prevent removing sufficient chloride in a single boildown step.

Further work on the batch method for chloride removal was abandoned in favor of a continuous method. The studies of a continuous process are discussed in Section II of Part II.

TABLE 5
SUMMARY OF BATCH CHLORIDE REMOVAL DATA

Dissolvent - 2 liters, 5M HNO ₃ , 2.5M HCl Fuel - 276.4 gms per run Temperature - boiling								
Run No.	Composition M	Dissolver Product	Product After Chloride Removal-1	Boil-Off Condensate-1	Product After Chloride Removal-2	Boil-Off Condensate-2	Absorber Product During Dissolution	Absorber Product During Chloride Removal
Chm-1	H ⁺	----	8.22	6.69	6.21	13.77	----	----
	Cl ⁻	----	0.0006	0.23	0.0004	0.0008	----	----
	NO ₃ ⁻	----	11.62	6.56	13.72	13.55	----	----
Chm-2	H ⁺	0.61	----	4.90	8.39	8.87	----	----
	Cl ⁻	2.06	----	0.85	0.0005	0.009	----	----
	NO ₃ ⁻	3.42	----	4.13	12.81	8.72	----	----
Chm-3	H ⁺	0.32	7.15	5.03	----	----	0.20	0.30
	Cl ⁻	----	0.0047	0.0047	----	----	0.0038	0.615
	NO ₃ ⁻	----	11.33	4.14	----	----	0.010	0.045

III. CONCLUSIONS

The following conclusions are based on the data obtained from the batch studies.

1. Loss of chloride and nitrate during the dissolution step can be greatly minimized by using proper dissolution techniques such as controlling the dissolution rate, i.e., by temperature control, to minimize gas evolution.
2. Dissolver off-gas rate averaged $0.1 \text{ ft}^3/(\text{min})(\text{ft}^2 \text{ of fuel surface})$.
3. Off-gas composition varied widely and was dependent on rate of gas evolution in the dissolver.
4. Two batch boildowns using 70 per cent nitric acid are probably necessary to remove chloride down to a level of $< 25 \text{ ppm}$.

PART II
CONTINUOUS STUDIES

PART II

CONTINUOUS STUDIES

The major portion of this program was devoted to developing a suitable continuous process both for the dissolution and the chloride removal steps. Information gained in the batch studies of Part I was used in designing the necessary equipment for the continuous process studies.

The following sections discuss the equipment and present the pertinent results of the dissolution, chloride removal, and solvent extraction studies. Two flowsheets based on these results are presented, and areas of the flowsheets in which further information is necessary are discussed.

I. DISSOLUTION

A. Objective

The objective of this portion of the study was to determine the behavior of a continuous dissolver during dissolution of nichrome fuel. Dissolution rate data(7) indicated that sufficient dissolution could be obtained using less concentrated dissolvent (1.0M HCl - 6.0M HNO₃) than that used in the batch process (2.5M HCl - 5.0M HNO₃). Therefore, the dissolution studies of the continuous process used the more dilute dissolvent.

Variables investigated were the effect of acid feed rate and fuel element height on dissolution rate, off-gas rate and composition, and type of solids produced. Also determined was the efficiency of the downdraft condenser for chloride absorption.

B. Equipment

The dissolution system can be divided into four sections: (1) feed, (2) dissolver, (3) product, and (4) off-gas. Figure 2 is a schematic equipment flowsheet of the continuous dissolution equipment. These sections are described below:

1. Feed

The feed section consisted of polyethylene makeup and feed tanks, a metering burette, feed pump, and feed preheater. Polyethylene was chosen for the feed system because of its corrosion resistance to the mixed HCl-HNO₃ solutions under consideration and of its low cost. Small batches of feed were generally made up in 25-liter bottles, and large batches of feed (up to 175 liters) were made in 55-gallon polyethylene drums. The feed could be transferred from the drum to the small feed tank (a 25-liter bottle) by pressuring the drum. A 2000-cc

Figure 2

Schematic Equipment Flowsheet of Continuous Dissolution Equipment

burette, connected in parallel with the small feed tank, was used to check the feed rate.

Steam flow to the preheater was controlled by a motor valve-temperature controller system with the controller receiving its signal from a thermocouple located 18 inches above the bottom of the dissolver.

A peristaltic pump with speed reducer was used in the feed system. Most of the lines handling thermally cold solutions were either Saran plastic tubing or PVC pipe.

2. Dissolver

The dissolver was made of two-inch-diameter Pyrex glass pipe eight feet long with the product take-off line located six feet above the bottom of the dissolver. Attached to the top of the dissolver was the slug charging section consisting of a two-foot length of two-inch-diameter Tygon tubing. Two large pinch clamps were used to close off the tubing. The bottom plate of the dissolver was made of 1/4-inch titanium plate. Two 1/2-inch-diameter tubing lines passed through this plate - one the feed line and the other the condensate return line. Above the plate (separated by a 1/4-inch-thick Teflon gasket) was a perforated 20-gauge titanium sheet. Its 1/16-inch-diameter holes prevented small pieces of fuel elements from plugging the feed and condensate lines.

3. Product

The product section consisted of a product cooler made of approximately 20 feet of 1/2-inch-diameter titanium tubing coiled inside a stainless steel shell. The shell side cooling water was manually controlled through a rotameter with a maximum flow of 6 gpm.

After cooling, the product passed through a specific gravity measuring pot made of two-inch-diameter Pyrex glass pipe 2.5 feet in length. The product was collected in four-liter cylinders for rate checks and then dumped into either 50-liter polyethylene jugs or one of the 55-gallon drums.

4. Off-Gas

The off-gas section consisted of a downdraft condenser, gas-liquid separation pot, rotameter for measuring off-gas rate, and a peristaltic pump for returning condensate to the bottom of the dissolver.

A 1/2-inch-diameter line (made of 0.035-inch wall, titanium tubing) carried the off-gas from the dissolver to the top of the condenser. This line was wrapped with heating tape and insulated with mineral wool; heating was necessary to prevent condensation ahead of the downdraft condenser. This was necessary for evaluation of the performance of the condenser. The temperature of the off-gas passing through this line was maintained at approximately 200°F. The downdraft condenser consisted of a jacketed 1/2-inch-diameter, 0.035-inch wall, titanium tube 92.5 inches in length. The condenser had three separate jacketed sections with condensate and off-gas sampling positions between the jacketed sections, as shown in Figure 3. The use of several sampling positions was necessary for evaluation of the performance of the downdraft condenser, since there were some run conditions where the available surface area was greater than needed. Each condenser section had a cooled length of 25 inches (0.27 ft²). Cooling water passed into the bottom jacketed section and out the top section. The water to the condenser was manually controlled by adjustment of a 0-5 gpm flow regulator. Thermocouples were located between each jacketed section and on the inlet and outlet cooling water lines to measure temperatures needed for determining condenser performance.

The off-gas and condensate first passed into a separation pot (a four- to one-inch glass reducer). The gas then passed through a section of Pyrex glass pipe packed with glass wool (to trap entrained liquid), and then through a rotameter having a maximum capacity of 0.32 scfm of air at 14.7 psia and 70°F. The condensate from the separation pot was pumped to the bottom of the dissolver through a Saran line using a peristaltic pump. With the pump off, the condensate rate was determined by timing the rise of the liquid level in the pot between two marks.

C. Results

The major criterion for correlating the data obtained from the variety of conditions imposed on the system is the ratio of acid feed rate to the surface area of fuel present in the dissolvers. This ratio, designated as FSR, with the units of cm/min, is analogous to space velocity. The variables and their respective ranges were acid feed rate - 3, 6, and 12 liters per hour - and surface area of fuel - 8300, 13,000, 14,800, and 23,300 cm². Two types of fuel were used --- nichrome V containing uranium (described in Appendix A) and 1/8-inch-thick nichrome V plate. Complete data for the dissolution experiments are presented in Table 13 in Appendix B.

Figure 3

Schematic Diagram of Downdraft
Condenser Used in the
Continuous Dissolution Studies

1. Effect of FSR on Dissolution Rate

The main difficulty in calculating the ratio of acid feed rate to surface area of fuel (FSR) was the determination of the actual amount of surface area present in the dissolver during a run. The value used for the surface area was determined as a function of bed height prior to the start of a run; several measurements were made to obtain an average value. Obviously, the total actual surface area present during any individual run could vary widely. However, visual observation of the dissolver during dissolution showed that the configuration of the fuel did not change drastically, and it is felt that the basis used for determining surface area was valid.

The values of dissolution rates at constant FSR were averaged and the results are shown in Figure 4. The data indicate an almost linear relationship between dissolution rate and FSR over the range of conditions studied. No appreciable difference in dissolution rate was observed between the two types of fuel studied.

2. Effect of FSR on Product Metal Concentration

The product metal concentration was also determined as a function of FSR. The results are shown in Figure 5. Since there was a large spread in the data, the metal concentration was averaged at a constant FSR.

Figure 4.

Effect of Acid Feed Rate and Fuel Surface Area
on Dissolution Rate

Figure 5

Effect of Acid Feed Rate and Fuel Surface Area
on Product Metal Concentration

Product metal concentration and product acidity are shown as a function of product specific gravity in Figures 6 and 7. These correlations should be useful for rapidly checking dissolver operation during a run.

3. Effect of FSR on Dissolver Off-Gas Composition and Rates

a. Method of Analysis

A new analytical procedure was developed late in this program for analyzing the off-gas stream produced in this system. This new procedure still had a limitation - it was not possible to determine what species of chlorine might be present. It could, however, determine nitrosyl chloride (NOCl) which the previous method was unable to do.

The following procedure was used to determine the species present in the off-gas streams:

(1) Duplicate samples were collected by passing through a hot (200°F) bed of anhydrous magnesium perchlorate (Anhydron) to remove water vapor. (Previous studies indicated that negligible quantities of the other constituents would be adsorbed.)

Figure 6

Effect of Product Acidity
on Product Specific Gravity

Figure 7

Effect of Product Metal Concentration
on Product Specific Gravity

(2) One sample was analyzed by both infra-red and mass spectrometry for NOCl and the compounds of nitrogen.

(3) The remaining sample was contacted with caustic to determine total chlorides present.

(4) The difference in chloride between the two samples reflects other compounds of chlorine (HCl, Cl₂, etc.).

b. Composition

Experimental work on this project was terminated shortly after the above procedure was developed. Therefore, the off-gas sampling program was very limited.

Samples were taken both ahead of and after the downdraft condenser. Three sets of samples (3 runs) appeared to be quite satisfactory for analyzing the performance of the condenser. The raw data were corrected to an air-free basis and all the NO₂ was assumed to come from NO. Table 6 presents these data on an air-free basis, and also includes the calculated values of the condenser efficiency for chloride removal.

TABLE 6
ANALYSIS OF OFF-GAS PRODUCED IN THE CONTINUOUS DISSOLVER SYSTEM

Component mole Per Cent	Run No.					
	31-33		37		41	
	B(1)	A(2)	B(1)	A(2)	B(1)	A(2)
NO	12.5	81	71.4	73.1	87.5	92
N ₂ O	25	4	0.5	2.4	8.3	5.4
NOCl	62.5	9	28.1	24.4	4.2	2.7
Off-gas rate (scfm)	0.25		0.32		0.10	
Condensate rate (l/min)	0.210		0.025		0.040	
Chloride Conc. (M)	0.60		0.67		0.182	
Chloride Absorbed (%)	84.9		15.4		89.5	

(1) Sample taken ahead of condenser.
(2) Sample taken after condenser.

The chloride absorption efficiency is shown in Figure 8, as a function of the ratio of off-gas rate and condensate rate. Holding the off-gas rate constant and increasing the condensate rate, i.e., by higher boilup rate, increases the efficiency of chloride absorption. Alternate methods for improving the efficiency of the condenser would be (1) to add the acid feed through the top of the condenser, or (2) to fill the condenser with packing to greatly increase the contact of gas and liquid, thereby increasing the amount of chloride transferred.

The results from the total chloride analysis indicate that essentially all (>99%) of the chloride in the off-gas is present as nitrosyl chloride. Thus, design of a downdraft condenser should be based on all the chloride present as NOCl .

c. Off-Gas Rates

The flow rate of the final off-gas stream is also plotted as a function of FSR, and the data are shown in Figure 9. There is almost a linear increase in the flow of off-gas as the FSR increases. Temperature of this final off-gas stream averaged $70 \pm 5^\circ\text{F}$ for the data shown. Figure 10 shows the relationship of FSR and the off-gas/product metal concentration ratio (ft^3/mole).

4. Effect of FSR on Acid Consumption

The acid consumption (moles of acid consumed per mole of fuel dissolved) increased from approximately 2.2 to 3.2 moles as the FSR increased from 0.004 to 0.024 cm/min . Results are shown in Figure 11.

5. Effect of Temperature on Dissolution and Off-Gas Rates

Dissolution temperatures ranged from 180° to 200°F . There was no apparent effect of temperature on the dissolution rate or on the total quantity of off-gas produced.

Figure 8

Effect of the Off-Gas
to Condensate Ratio on
Chloride Removal Efficiency
of the Downdraft Condenser

CPP-S-2069

Figure 9

Effect of Acid Feed Rate and Fuel Surface Area
on Dissolver Off-Gas Rates

CPP-S-2070

Figure 10

Effect of Acid Feed Rate and Fuel Surface Area
on Off-Gas/Product Metal Ratio

CPP - S - 2071

Figure 11

Effect of Acid Feed Rate and Fuel Surface Area
on Acid Consumption

CPP - S - 2073

Figure 12

Effect of Dissolver Off-Gas Rate
on Downdraft Condenser Performance

6. Downdraft Condenser Evaluation

The downdraft condenser functions as a condenser-absorber in that it removes the water vapor and at the same time absorbs some of the components in the off-gas stream, specifically compounds containing chlorine. Complete absorption of the various compounds of chlorine was not accomplished in most cases, indicating that a more efficient absorber is necessary to absorb completely the chlorine compounds.

Temperatures were measured between each of the cooled sections, and these results are presented in Figure 12 as a function of the non-condensable off-gas rate. With the higher off-gas rates (> 0.20 scfm), sufficient cooling of the off-gas could not be accomplished in two feet of 1/2-inch-diameter tubing (0.27 ft^2). In most cases, the off-gas condensate stream was cooled to at least 80°F in four feet of 1/2-inch-diameter tubing (0.54 ft^2).

Analysis of the heat transfer performance of the condenser is complicated by the simultaneous mass and heat transfer that occurs. However, a simplified approach which does give an indication of the performance is the effect of the condensate rate on the overall heat transfer coefficient. This coefficient increased from approximately 200 to 1200 $\text{BTU}/(\text{hr})(\text{ft}^2)(^\circ\text{F})$ as the condensate rate increased from 20 to 210 cc/min . These results are presented in Figure 13.

CPP-S-2072

Figure 13

Effect of Condensate Rate on the
Overall Heat Transfer Coefficient
in Downdraft Condenser

7. Solids

Solids produced during the dissolution of nichrome fuel range in size from less than one micron up to 1/2-inch or slightly larger. Of course, the largest pieces remain in the dissolver and are eventually reduced in size. The dissolver effluent generally contains particles less than 1000 microns in size. Composition of the residue includes oxides of chromium, nickel, niobium, and silicon. Niobium is present in the cladding and the silicon is contained in the braze metal.

A thorough study of the characteristics of the solid residue produced from dissolving niobium-free nichrome in the four-inch-diameter pilot plant continuous dissolver has been reported by Roberts⁽⁶⁾. The major constituents of the solids was Cr_2O_3 , with smaller amounts of nickel and silicon oxides. Negligible quantities of uranium were detected. The residue of solids averaged approximately two per cent of the dissolved metal.

Roberts⁽⁶⁾ used an Andreason pipette⁽⁸⁾ for determining the size distribution of the particles. Table 7 presents these data. As can be seen, most of the solids average 35 microns. Average composition of these solids was 55 per cent chromium, 8 per cent nickel, and 6 per cent silicon.

TABLE 7
PARTICLE SIZE DISTRIBUTION OF SOLIDS PRESENT IN
DISSOLVER EFFLUENT

<u>Effective Average Diameter, μ</u>	<u>Weight Per Cent</u>
875	3
475	5
412	27
35	65

Further solids characterization studies were made on material produced when dissolving 1/16-inch-thick nichrome V containing one per cent niobium and from scrap niobium-free fuel. The solids loading of the dissolver effluent and the composition and quantity of the residues are recorded in Table 8.

When dissolving fuel with and without niobium present, the concentration of solids in the dissolver effluents averaged 1.5 and 0.7 per cent of the total dissolved material, respectively.

TABLE 8

CONTINUOUS DISSOLVER EFFLUENT RESIDUE
FROM NICHROME FUEL

Run No.	Quantity		Constituents		
	g/l	Per Cent of Total Dissolved Material	Major	Minor	Trace
21*	2.3	1.4	Nb, Si	-----	-----
23*	2.8	1.7	-----	-----	-----
31*	0.8	1.4	Nb, Si	Ni, Cr	-----
33*	1.5	1.5	Nb, Si	Ni, Cr	-----
39	0.2	0.4	Cr	Fe, Ni, Si	Nb
41	0.6	0.8	-----	-----	-----
42	0.5	0.6	-----	-----	-----
43	1.0	0.9	-----	-----	-----
44	0.8	0.4	Cr, Ni	None	Nb, U

*Fuel contained one per cent niobium.

Reheating of some dissolver effluents caused the formation of a gelatinous material, later analyzed and found to contain niobium. Solution stability studies were made with niobium-containing solutions produced in two separate runs to determine the percentage of niobium which would remain in solution immediately after dissolution and that which would precipitate on reheating (necessary for the chloride removal step). The results are presented in Table 9.

The results indicate that approximately twenty per cent of the niobium remains in solution after the dissolution step and precipitates as a gelatinous mass on reheating. The data above also indicate that some silicon must also stay in solution for an unknown length of time before precipitating.

A post-precipitation experiment with dissolver effluent from a run using niobium-free fuel showed that approximately 2.5 per cent of the silicon was in the filtrate. It appears that the prior history of a particular solution has a considerable effect on the distribution and the behavior of niobium and silicon upon reheating the dissolver effluents.

TABLE 9

POST-PRECIPITATION RESULTS OF CONTINUOUS DISSOLVER EFFLUENTS

Fuel Dissolved: Nichrome - One Per Cent Niobium

	(mg)	Niobium (mg/ml)	(w/o) ⁽¹⁾	Niobium Residues (mg)	(w/o Nb)
Filtrate	5.9	0.12	19.9	28.9 ⁽²⁾	20.4
Solid	23.8	-	80.1	44.1 ⁽³⁾	53.9

(1) Based on total niobium present in dissolver effluent.

(2) Gelatinous material produced by boiling filtrate for 8 hours with total reflux.

(3) Material obtained by heating solids present in effluent to 1000°C, then fusing.

8. Effect of Aluminum on Dissolution Rate

Since some of the HTRE fuel elements were to be dissected and then put into aluminum cans, information was needed as to the effect of aluminum on the overall dissolution rate. Product solutions containing up to 50 grams aluminum per liter (30 grams nichrome per liter) had no appreciable effect on the overall dissolution rate. Results are shown in Figure 4. It is possible to produce acid-deficient solutions when dissolving a mixture of nichrome and aluminum.

Off-gas rates, when co-dissolving both metals, were essentially the same as when dissolving nichrome only. However, a considerable quantity of hydrogen was produced (15 to 25 volume per cent). Additional studies are required to more clearly define dissolver operation when co-dissolving nichrome and aluminum.

D. Conclusions

The following conclusions are based on the data obtained from the continuous dissolution study.

1. The effects of increasing the FSR from 0.004 to 0.024 cm/min were found to be as follows:

- a. Dissolution rate increased from 0.66 to 1.8 mg/(cm²)(min).
- b. Metal concentration of the dissolver effluent decreased from 185 to approximately 50 grams per liter.
- c. Off-gas rate increased from 0.07 to 0.3 scfm.
- d. Acid consumption increased from 2.2 to 3.2 moles per mole fuel.

2. Dissolver off-gas contained nitrogen oxides and nitrosyl chloride. Essentially all the chloride was present as nitrosyl chloride.

3. The chloride absorption efficiency of the downdraft condenser (0.81 ft^2 of surface area) increased from 15.4 to 84.9 per cent as the ratio of off-gas to condensate (ft^3/l) decreased from 13 to 1.

4. Residues in the effluent consisted of the oxides of chromium, nickel, niobium, and silicon up to total concentrations of 2.8 grams per liter.

5. Chemical stability studies on dissolver effluents indicated that thirty per cent of the niobium remained in solution during the dissolution step but precipitated out on reheating of the solution.

6. Dissolution temperatures from 180 to 200°F had little effect on either the dissolution rate or the off-gas rate.

II. CHLORIDE REMOVAL

A. Objectives

Although it had been shown previously (Part I) that satisfactory batchwise removal of the chloride ion from the dissolver effluent could be attained, it became apparent that the chloride ion could be readily removed by stripping with nitric acid vapors in a stripping column. Crooks, et al. (9), present equilibrium data for a system of nitric acid containing small quantities of chloride ion. Their data show that in solutions containing greater than 5.5M nitric acid, the vapor/liquid distribution coefficient for chloride would be greater than one. Additional equilibrium data are presented by Roberts, et al. (6), for the system studied here. Good agreement was found between the two sets of data. An acid concentration of 7.5M was chosen for the stripper column feed because it required a minimum amount of nitric acid to be added to the dissolver effluent and still did not require an excessive number of stages for the desired chloride removal. Therefore, dissolver effluents were adjusted to approximately 7.5M acid for the chloride removal studies.

B. Equipment

Three columns were used in the chloride stripping studies - 1-, 2-, and 4-inch-diameter Pyrex glass columns. The 1-inch-diameter column had packed heights of 4 and 6.3 feet; packing used was 1/8-inch glass helices. The 2-inch-diameter column had a packed height of 9.5 feet; two types of packing were used - 1/4-inch porcelain Berl saddles and 3/16-inch glass helices. The 4-inch-diameter column had a packed height of 7.5 feet; packing used was 1/4-inch porcelain Berl saddles.

The experimental setups for the three columns contained the same essential items: feed system, feed preheater, column proper, product cooler, and a downdraft condenser for the overheads. The following description applies to the system for the four-inch-diameter column; this was the only system which included automatic column control based on pressure drop across the column. This pressure drop controlled the boilup rate and hence the overhead rate.

1. Feed System

The feed system was identical to that used in the dissolver feed system (described in Section IB, Part II). The feed preheater for the stripping column consisted of 1/2-inch-diameter, 0.035-inch wall, titanium tubing 8 feet long with a 7.5-foot-long stainless steel jacket (0.98 square foot of heat transfer surface). The amount of preheat was manually controlled by a 0 - 100 psig steam regulator.

2. Stripper System

The stripper system consisted of the column, column head, packing, and a reboiler section.

The column was a four-inch-diameter Pyrex glass pipe ten feet in length. At the top of this section was a 4" x 1" x 1" Pyrex glass cross. The one-inch lines were for the feed inlet to the column and the overhead outlet to the downdraft condenser. The column was insulated with mineral wool.

The packing support, made of titanium, was cone-shaped and had a free area equal to the cross-sectional area of the column (12.6 in²).

The reboiler consisted of a 6" x 6" Pyrex glass tee containing a titanium coil constructed of 1/2-inch-diameter tubing having approximately 1.3 square feet of surface area. The vapor space above the coil consisted of a straight section of a 6-inch-diameter glass pipe 6 inches in length and a 6" x 4" Pyrex reducer. A calming section twelve inches long was added to the reboiler section and separated from the coil section by a baffle plate. The plate had a 1/32-inch-diameter weep hole to allow for complete drainage of the column following shutdown. The liquid level in the reboiler was maintained by a control valve which received its signal from probes located in the calming section.

3. Downdraft Condenser System

The condenser was of a shell-and-tube design made by the Corning Glass Works and had 13-1/2 square feet of heat transfer area. It was so constructed that corrosive solutions could be used in either the shell side or the tube side. For these studies, the corrosive stream passed through the tubes (seven 3/4-inch-diameter tubes). Cooling water to the condenser was manually controlled by a rotameter which had a capacity of 3.86 liters per minute. The condensed liquid was separated from the off-gas in a liquid seal at the bottom of the condenser with the off-gas passing through a rotameter to the vent. The liquid was collected and measured.

4. Product System

The product system consisted of a cooler and its control valve. The cooler was made of a stainless-jacketed titanium tube (1/2-inch in diameter) 7 feet long (0.92 ft² heat transfer surface area). Cooling water for the jacket was manually controlled by a rotameter having a capacity of three liters per minute. A 3/4-inch motor valve was used for controlling the liquid level in the reboiler. Figure 14 is a schematic equipment flowsheet of the stripping column equipment.

C. Results

Several variables were investigated to determine their effect on the removal of chloride ion from nitric acid-spiked dissolver effluent. These variables were vapor/liquid ratio, column diameter, type of packing, and packing height. All the dissolver effluents, as well as all the synthetic feeds, were adjusted to a nominal total acid concentration of 7.5M. The actual acidity of the column feeds ranged from 7.1 to 8.8M; chloride concentration ranged between 16 and 36 grams per liter, with the average being approximately 21 grams per liter. Complete data for the stripping experiments are presented in Table 14 in Appendix B.

A run was made in the one-inch-diameter column using chlorine-35 tracer⁽¹⁰⁾. The objective was to check the accuracy of the wet chemical method for chloride determination. Good agreement in the analytical results was found between the two methods.

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Figure 14

Schematic Equipment Flowsheet of Stripping Column System

1. Effect of Vapor/Liquid Ratio (V/L)

The molar vapor/liquid ratio was varied between 0.256 and 0.667 for these studies. Results obtained generally showed that increasing the vapor/liquid ratio decreases the quantity of chloride remaining in the kettle product. Figures 15, 16, and 17 present the data obtained in the three columns.

A problem of major concern in this process is the disposition of the column overhead stream. The most logical use for this stream is as fuel dissolvent. Examination of the data indicates that this stream is reusable, i.e., contains no excess water, if the chloride concentration is at least 1.4 molar. The maximum V/L at which this concentration occurs is approximately 0.4. Therefore, further consideration of a flowsheet assumes the use of a molar V/L of 0.4 in the stripping column.

2. Chloride Removal Performance

The average chloride concentration in the feed used for the chloride stripping studies was 20.7 grams per liter (16,000 ppm). It is necessary to limit the maximum chloride concentration to 100 ppm in the extraction column feed stream (extraction columns are constructed of type 348 stainless steel). This requires a chloride removal performance (concentration of chloride in feed/concentration of chloride in kettle) in the stripping column of at least 160. Values obtained in this work increased from 3 to 11,000 as the molar V/L increased from 0.28 to 0.667. Figures 18, 19, and 20 present these data.

Figure 15

Effect of Vapor/Liquid Ratio on
Product Chloride Concentration
One-Inch Diameter Column

Figure 16

Effect of Vapor/Liquid Ratio on
Product Chloride Concentration
Two-Inch Diameter Column

Figure 17

Effect of Vapor/Liquid Ratio on
Product Chloride Concentration
Four-Inch Diameter Column

Figure 18

Effect of Vapor/Liquid Ratio
on Chloride Removal Performance
One-Inch Diameter Column

Figure 19

Effect of Vapor/Liquid Ratio
on Chloride Removal Performance
Two-Inch Diameter Column

Figure 20

Effect of Vapor/Liquid Ratio
on Chloride Removal Performance
Four-Inch Diameter Column

3. Effect of Type of Packing

Although several types of packing were used, comparison can be made only between Berl saddles and glass helices. These two types of packing were used in the 2-inch-diameter column with a packed height of 9.5 feet in each case. The data shown in Figure 16 are inconclusive, but indicate that the glass helices may be somewhat more efficient at lower vapor/liquid ratios for the removal of chloride than the Berl saddles. Glass helices would be expected to be somewhat more efficient because of their smaller void space which should promote better mixing.

4. Effect of Packing Height

Assuming that a linear relationship exists between chloride removal performance and packing height (at least for V/L of 0.4), the data presented in Figure 21 indicate that approximately 8.1 feet of packing is necessary to obtain a chloride removal performance of 160. A packing height greater than 8.1 feet would undoubtedly be used in a plant column to allow for maloperation of equipment; a height of twelve feet is recommended. This would give a chloride removal performance of at least 1300, or produce approximately 12 ppm chloride in the kettle product.

5. Height of a Transfer Unit

Equilibrium studies reported by Roberts, et al.⁽⁶⁾, had shown that decomposition or oxidation was the controlling factor for chloride removal in solutions greater than 0.3M in hydrochloric acid. These mechanisms should be greatly enhanced by preheating the stripper column feed; this feed generally contains approximately 0.5M HCl. For stage calculational purposes, it was assumed that the chloride concentration of solution entering the packed section was 0.3M. The validity of this assumption is borne out by the results of Runs 24 and 25, which indicated that additional stages would be required to obtain the desired chloride removal when the feed is not preheated.

Calculation of the number of transfer units (NTU) required to obtain 25 ppm chloride in the kettle bottoms indicated that approximately four stages would be necessary. Results for the height of a transfer unit (HTU) were determined by dividing the height of packing by the NTU's. These results are presented

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Figure 21

Effect of Packing Height
on Chloride Removal Performance

in Figure 22 and show that the HTU decreases as the vapor/liquid ratio increases. An HTU of approximately 2.2 to 2.8 feet is produced with V/L of 0.4. This requires a packing height of approximately 9 to 11 feet to obtain the desired chloride removal. A sample calculation of HTU is given in Appendix C.

6. Stripper Off-Gas

The rate of the final off-gas stream from the liquid-gas separation pot below the downdraft condenser ranged from essentially zero to a maximum of 0.12 scfm. Reliable off-gas rates were obtained only on runs made in the four-inch-diameter column.

The results of the few measurements of off-gas rate are presented in Figure 23. Assuming a straight line function of overhead product and off-gas rate, it was calculated that approximately 1.1 ft³ of off-gas (presumably non-condensables) was produced per liter of overhead product.

Samples of this final off-gas stream were analyzed, and the results obtained indicate that essentially all the chloride remaining in the final off-gas stream was present as NOCl.

Average analysis (air-free basis) of the stripper off-gas was as follows:

NO	62.8 mole per cent
NOCl	30.7
N ₂ O	6.5

CPP-S-2082

Figure 22

Effect of Vapor/Liquid Ratio
on Height of a Transfer Unit

Figure 23

CPP-S-2083

Effect of Stripper Overhead Rate
on the Non-Condensable Off-Gas Rate

The chloride loss in the final off-gas stream amounted to 25 per cent of the total chloride in the system, indicating that the downdraft condenser used (13.5 ft² of surface area) was not very efficient in removing the chloride ion from the off-gas. However, consideration of chloride equilibrium shows that additional removal of the compounds of chlorine from the off-gas is not possible when in contact with the overhead stream. A separate piece of equipment, such as a caustic scrubber, would be necessary to remove the chlorides remaining in the off-gas.

7. Column Capacity

Comparison of measured flooding capacities with those calculated using the correlation of Lobo, et al. (11), is presented in Figure 24 for the two- and four-inch-diameter columns. Good agreement was obtained only with the data obtained in the four-inch-diameter column.

The difference in flooding rates between the two columns could possibly be due to the way in which the columns were initially loaded with packing, since the void fraction of the packing exerts a large effect on the flooding rate.

8. Column Control

The method for controlling the boilup rate (hence the overhead rate) in the four-inch-diameter stripping column was by pressure drop across the packing. Satisfactory control of the column was obtained using this method.

Figure 24

Capacities of
Packed Stripping Columns

D. Conclusions

The following conclusions are based on the data obtained from the chloride removal study.

1. The effects of increasing the molar vapor/liquid ratio from 0.27 to 0.667 on the following factors were observed.

- a. Chloride concentration in the uranium-bearing stream decreased from 16,000 to 2 ppm.
- b. Chloride removal performance increased from 5 to 12,500.
- c. Height of a transfer unit decreased from 3.2 to 1.0 feet.

2. Increasing the height of packing from 4.0 to 9.5 feet increased the chloride removal performance from 30 to 350 at a V/L of 0.4. A vapor/liquid ratio of less than 0.4 is required to produce an overhead product usable directly as a dissolvent.

3. Essentially no difference in column performance was observed when using either 1/4-inch porcelain Berl saddles or 3/16-inch glass helices.

4. Increasing the stripper overhead product rate from 25 to 110 cc/min increased the off-gas rate (non-condensables) from 0.05 to 0.12 scfm.

5. Off-gas contained nitrogen oxides and nitrosyl chloride.

6. The chloride absorption efficiency of the downdraft condenser (13.5 ft² of surface area) was not sufficient to remove all compounds of chlorine from the off-gas; equilibrium considerations indicate the need for a dilute acid or caustic scrubber for complete removal.

7. Flooding capacity of the column agreed with experimental data obtained with other systems.

III. SOLVENT EXTRACTION

A. Objectives

Initial work reported by Rohde, et al.⁽⁷⁾, had indicated that adequate recovery (99.99 per cent) of the uranium from solutions containing 0.01M uranium, 0.19M nickel, and 0.05M chromium could be attained using 5 volume per cent TBP in Amsco 125-90W. Five extraction runs were made in the pilot plant extraction system using this extractant. Both synthetic feed and feed produced from dissolver effluents (after chloride removal) were used; also, some feeds contained aluminum. Capacity of the extraction column was also determined. Results of the extraction data are summarized in Appendix B (Table 15).

B. Equipment

The solvent extraction system consisted of three glass pulsed columns, arranged for cascade flow of solvent between columns, and the necessary auxiliaries. These included pulse generators, feed and receiver tanks, pumps, control instruments, and solvent washing equipment.

The columns were assembled from precision-bore Pyrex glass pipe with an inside diameter of 2.00 inches. The plates used were made of perforated stainless steel, with 1/8-inch holes spaced on 1/4-inch centers to give twenty-three per cent free area, and spaced on two-inch centers in the column. This is the standard cartridge described by Sege and Woodfield (12). The cartridge length in the extraction column was 15.2 feet. A settling zone was provided at both ends of the column. A Lapp "Pulsafeeder" was used as the extraction column pulse generator.

C. Results

1. Column Capacity

Initial tests conducted in the system were for the purpose of determining a flooding curve using synthetic feed. The results of these tests are presented in Table 10 and shown in Figure 25. Flooding point checks were made at the end of three runs, and these results are also presented in Table 10 and shown in Figure 25.

2. Uranium Losses

Uranium waste losses ranged from 0.02 to 0.15 per cent. The high waste losses were due to an insufficient number of stages in the column. Addition of aluminum (7 grams/liter) to the column feed increased the uranium waste loss from 0.02 to 0.06 per cent. Increasing the aluminum concentration in the feed from 7 to 37 grams per liter increased the uranium waste losses from 0.06 to 0.15 per cent.

TABLE 10
EXTRACTION COLUMN FLOODING DATA

Column: 2" x 15.2' standard cartridge
Solvent: 5% TBP (by vol) in Amsco 125-90W
Feed: 5.7N HNO₃
20.8 g/l Nickel
5.1 g/l Chromium
Aqueous-to-Organic
Ratio (A/O): 1.0
Pulse Amplitude: 1.0 inch

Run No.	Volume Velocity gal/(hr) (ft ²)	Flooding Points Frequency cycles/minute
A	493	82
B	635	70
C	747	60
D	890	48
E	1045	15, 35
F	1200	inoperable
1	500	70
3	510	77
5	489	77

Figure 25

Effect of Pulse Frequency on Extraction Column Capacity

3. Height of a Transfer Unit

The height of a transfer unit (HTU) ranged from 1.5 to 2.0 feet. The number of transfer units ranged from 7.7 to 10.2. These data indicate that approximately eleven stages are necessary to obtain satisfactory uranium losses, and that stage heights on the order of two feet will be obtained. Since the plant columns have a cartridge section of at least 21 feet, satisfactory uranium recovery (99.99 per cent) should be obtainable.

Additional studies reported by Roberts ⁽⁶⁾ include equilibrium data for solvents containing up to 30 volume per cent TBP. Using the higher TBP concentration gives higher equilibrium values for the uranium distribution between solvent and aqueous feed, and would therefore require fewer stages to obtain the same recovery. The flowsheet discussed in Section IV recommends the use of 15 volume per cent TBP.

4. Column Operation

Satisfactory operation of the extraction column was obtained in all the runs except one. At no time were emulsions encountered either in the plate section or at the interface, even with feed solutions produced from dissolver effluents. Considerable difficulty in column operation was encountered during Run 5 which had 37 g/l of aluminum in the feed. The original plan was to run with a pulse frequency of 70 cycles per minute; however, the column kept flooding until the pulse frequency was lowered to 57 cycles per minute. Additional work would be necessary to further define the stability area with solutions containing aluminum.

D. Conclusions

The following conclusions are based on the data obtained from the solvent extraction study.

1. Satisfactory extraction column performance can be obtained when processing nichrome solutions.

2. Uranium waste losses are somewhat high when using five volume per cent TBP in Amsco 125-90W (0.02 per cent in 10.2 stages).

3. Presence of high concentrations of aluminum in the feed has an adverse effect on uranium waste losses and column capacity.

IV. PROCESS FLOWSHEETS

An integrated process combining dissolution, chloride removal, and solvent extraction is discussed in this section. Two flowsheets are presented for the dissolution step. One process would produce a dissolver effluent containing approximately 55 grams of dissolved metal per liter, and the other process would produce a dissolver effluent containing 195 grams of dissolved metal per liter.

The major criterion for Flowsheet No. 1 was criticality; therefore, the uranium concentration in any stream did not exceed 10 g/l. In the extraction column feed, the uranium concentration was set at 6 g/l and an aqueous-to-organic ratio of 1.0 was specified. Use of this flowsheet would allow the processing of 16.3 kilograms of U per day if sufficient extraction column capacity were available. Present SIR extraction system, if used, would limit the uranium capacity to approximately five kilograms per day. Table 11 presents details of Flowsheet No. 1.

TABLE 11
CONTINUOUS PROCESS FOR RECOVERING 16.3 KILOGRAMS OF URANIUM PER DAY
FLOWSHEET NO. 1

		Dissolver				Stripping Column				Extraction Column		
		<u>Fuel</u>	<u>Dissolvent</u>	<u>Product</u>	<u>Off-Gas</u>	<u>HNO₃ Spike</u>	<u>Feed</u>	<u>Product</u>	<u>Overheads</u>	<u>Off-Gas</u>	<u>H₂O Spike</u>	<u>Feed</u>
Weight	(kg)	5.5										
Volume	(L)	--	100	100	68.0	45	145	102.6	42.4	1320	22.4	125
Acidity	(N)	--	7.0	5.0	--	13.05	7.5	9.6	5.0		--	7.80
Chloride	(M)	--	1.0	1.0	0.1 mole	--	0.69	0	1.91	18 moles	--	--
Nitrate	(M)	--	6.0	5.8	3 moles	13.05	8.05	11.2	3.1	42 moles	--	9.19
Nickel	(M)	3.6	--	0.613	--	--	0.423	0.596	--	--	--	0.49
Chromium	(M)	0.9	--	0.173	--	--	0.119	0.169	--	--	--	0.130
Uranium	(M)	1.0	--	0.0425	--	--	0.0293	0.0414	--	--	--	0.0338
Metal	(g/l)	--	--	55.0	--	--	37.9	53.6	--	--	--	44

*Flowsheet values based on one kilogram of uranium.

One method for controlling the operation of the dissolver is to consume completely the acid in the feed. The nichrome dilute $\text{HCl-NH}_4\text{NO}_3$ system cannot become acid deficient; therefore, running the dissolver under the proper conditions (i.e., complete acid consumption) should produce a fairly constant product. Flowsheet No. 2 is based on this premise. This flowsheet, however, requires equipment that is critically safe, geometry-wise. Again, the uranium concentration in the extraction column feed is limited to 6 grams per liter. The uranium capacity would be 7.5 kilograms per day if extraction capacity were not limiting. A molar V/L of approximately 0.3 in the stripping column is necessary to produce a reusable overhead. This requires a stripping column with a packed height of approximately twelve feet to produce a suitable product. Table 12 presents details of Flowsheet No. 2.

TABLE 12
CONTINUOUS PROCESS FOR RECOVERING 7.5 KILOGRAMS OF URANIUM PER DAY
FLOWSHEET NO. 2

		Dissolver				Stripping Column				Extraction Column		
		Fuel*	Dissolvent	Product	Off-Gas	HNO ₃ Spike	Feed	Product	Overheads	Off-Gas	H ₂ O Spike	Feed
Weight	(kg)	5.5										
Volume	(L)		30.2	30.2	62.0	40.8	71.0	56	15	626	69	125
Acidity	(N)		7.0	0		13.05	7.5	9.0	4.0		--	4.03
Chloride	(M)		1.0	1.0	0.1 mole	--	0.426	0	1.5	8 moles	--	0
Nitrate	(M)		6.0	5.9	2.8 moles	13.05	10.0	12.4	2.5	20 moles	--	5.56
Nickel	(M)	3.6 kg	--	2.00		--	0.851	1.08	--		--	0.48
Chromium	(M)	0.9 kg	--	0.55		--	0.23	0.292	--		--	0.131
Uranium	(M)	1.0 kg	--	0.14		--	0.060	0.076	--		--	0.0340
Metal	(g/l)	--	--	182.1		--	77.5	98.3	--		--	44

* Flowsheet values based on one kilogram of uranium.

V. OPERATIONAL PROBLEMS

The work reported here was performed using scrap fuel obtained from General Electric in 1958. This scrap, the type used for HTRE No. 1 fuel elements, did not contain the small quantity of niobium in the cladding that later fuel elements contained. Therefore, niobium in small quantities was added to the dissolver charges and subsequently sent through the stripping and extraction columns. No troubles were encountered in the operation of the dissolver, the stripping column, or the extraction system.

Some unirradiated HTRE No. 3 fuel elements were received early in 1960, and additional tests were scheduled for proving the flowsheet that had been developed using the scrap material. The results of the dissolutions made with the HTRE No. 3 fuel agreed very well with the previous results. Serious trouble was encountered, however, in the operation of the stripping column. Within 1/2-hour after starting feed to the column, troubles were encountered by buildup of a gelatinous material in the packed section. Within five hours, the stripping operation had to be stopped because the column had become completely plugged. Several attempts were made to operate the stripper without much success. Clean-out of the column was easily accomplished with a boiling, ten per cent caustic solution.

The stripper bottoms were collected, diluted with water, and then sent through the extraction system. No problems were encountered in the extraction column operation.

Observation of duplicate dissolver effluent samples showed no gelatinous material present after six months' storage. Evidently the formation of the gelatinous material, hydrated silica and niobia, was enhanced by the reheating of dissolver effluents following the dissolution step. Additional studies are necessary to develop satisfactory means for handling silica and niobia solids.

VI. ACKNOWLEDGEMENTS

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VIII. APPENDIXES

APPENDIX A

FUEL DESCRIPTION

Nichrome fuel has been used in all the major GE-ANP reactor experiments. In HTRE-1 and HTRE-3, it is the only fuel to be used; in HTRE-2, it is the parent core. There are variations in the design and gross composition of the various reactor cores, but the variations are small in the designs of fuel element assemblies. For brevity, only the HTRE-1 core A2, the first operational model, is described here.

The core contains 37 fuel element assemblies in a hexagonal arrangement.

Each assembly is a right circular cylinder. It is made up of 18 elements, spaced 0.125 inch between surfaces, supported by four longitudinal stainless steel rails about $\frac{3}{8}$ inch by $\frac{1}{32}$ inch. A complete assembly is 29.125 inches long and has a maximum diameter of 3.557 inches.

Each element is made up of 14 or 16 concentric cylindrical ribbons and is 1.49 inches long and 3.432 inches in diameter. Other models may have from 11 to 20 ribbons. The spacings of the ribbons in 14-ribbon elements are:

5 inner spaces: 0.091 inch
8 outer spaces: 0.102 inch;

the inside radius is 0.186 inch. The spacing of the ribbons in 16-ribbon elements is 0.080 inch; the inside radius is 0.241 inch. There are eleven 14-ribbon elements and seven 16-ribbon elements per assembly.

The ribbons are sandwiches of an enriched UO_2 -nichrome matrix, made by powder metallurgical techniques, 0.004 inch to 0.013 inch thick, clad on both sides by 0.004-inch-thick layers of niobium-modified nichrome. The uranium is enriched to 93.4 per cent U^{235} ; the nominal nichrome composition is 80 Ni-20 Cr; and niobium content of the cladding is 1 per cent. The UO_2 is 42 per cent of the fuel matrix, which has a density of 9.344 grams per cubic centimeter.

The gross composition of a fuel element assembly, calculated from the above data, is given below.

	<u>Weight (grams)</u>	<u>Weight (fraction)</u>
UO_2	1528.9	0.2472
Ni	3782.9	0.5869
Cr	974.6	0.1512
Fe	132.9	0.0206
Nb	26.6	0.0041

For the calculations, the average ribbon thickness of 0.018 inch was used.

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APPENDIX B
TABLE 13
SUMMARY OF CONTINUOUS DISSOLUTION EXPERIMENTAL
DATA OF NICHROME FUEL (HTRE)

Run No.	1	2	3	4	5	8	12	13	14	15	16	17	18	19	20	21
Run Conditions																
Feed rate (cc/min.)	52.3	50.0	100.0	50.0	50.4	47.2	100.	50.	100.	100.	50.	100.	104.	100.	100.	92.0
Product rate (cc/min.)	50.0	52.3	100.0	50.0	51.7	48.0	100.	54.	120.	100.	46.	110.	111.	85.	96.	67.
Diss. Temp. (°F)	188.	180.	172.	190.	186.	190.	178.	187.	183.	180.	182.	178.	190.	190.	194.	200.
Fuel element Ht. (ft)	-----	-----	-----	-----	-----	-----	-----	-----	-----	-----	-----	-----	-----	3.5	3.5	3.5
Surface area of fuel (cm ²)	-----	-----	-----	-----	-----	-----	-----	-----	-----	-----	-----	-----	-----	14,820.	14,820.	14,820.
Fuel addtn. rate (gm/min.)	1.72	4.0	6.0	4.0	4.0	4.0	8.	4.	4.	8.	4.	4.	4.	4.53	11.6	11.1
Run duration (hrs)	22.5	13.	10.	13.	6.	7.	8.	4.	4.	4.	4.	4.	4.	4.	4.	3.
Stream Analysis																
Feed																
H ⁺ (N)	7.31	7.59	6.87	7.0	6.97	6.25	6.73	7.27	7.24	7.29	7.26	7.39	7.17	6.79	7.34	7.16
Cl ⁻ (g/l)	36.92	43.26	42.30	36.0	44.47	40.9	46.6	38.9	40.9	42.6	38.9	40.5	37.1	34.6	32.5	79.0
NO ₃ (M)	5.85	6.33	5.81	5.90	4.84	5.93	5.8	5.88	5.61	5.80	5.66	5.77	5.80	5.51	6.94	4.57
sp. gr.	1.206	1.218	1.194	1.207	1.193	1.206	1.203	1.209	1.209	1.210	1.209	1.212	1.215	1.198	1.213	1.187
Product																
H ⁺ (N)	5.20	4.43	4.77	3.15	2.59	3.29	3.91	3.56	5.06	3.50	3.59	4.45	5.14	5.29	2.97	0.15
Cl ⁻ (g/l)	34.50	33.88	30.64	34.0	49.3	33.5	43.7	39.6	36.3	32.6	32.6	30.7	31.2	30.2	34.8	85.0
NO ₃ (M)	5.34	5.87	5.46	5.12	4.77	5.06	5.23	5.22	5.57	5.14	5.18	5.18	5.30	5.13	5.55	4.8
Ni ³ (g/l)	18.50	52.88	28.40	60.0	63.85	50.15	52.3	52.6	26.6	44.2	46.2	32.5	23.6	20.5	68.5	125.
Cr (g/l)	5.52	11.26	6.80	11.76	14.63	11.27	5.51	12.4	6.44	10.1	11.0	8.04	6.25	5.70	15.5	26.
U (g/l)	-----	-----	-----	-----	-----	-----	-----	-----	-----	-----	-----	-----	-----	-----	12.50	12.5
Al (g/l)	-----	-----	-----	-----	-----	-----	-----	-----	-----	-----	-----	-----	-----	-----	-----	-----
sp. gr.	1.238	1.303	1.245	1.287	1.302	1.267	1.280	1.248	1.268	1.276	1.290	1.251	1.236	1.230	1.326	1.40
Results																
Diss. rate [mg/(cm ²)(min.)]	-----	-----	-----	-----	-----	-----	-----	-----	-----	-----	-----	-----	-----	0.20	0.70	1.0
Off-gas rate (scfm)	0.12	0.10	0.10	0.07	0.10	0.12	0.10	0.13	0.15	0.15	0.13	-----	0.05	-----	-----	0.16
Ft ³ off-gas/mole fuel diss.	0.26	0.09	0.15	0.06	0.07	0.11	0.09	0.21	0.24	0.19	0.25	-----	0.09	-----	-----	0.06
Acid consumption (moles/mole)	4.55	2.35	3.13	2.88	2.84	0.88	2.69	2.55	1.56	4.79	3.98	2.71	2.88	4.67	2.80	2.6
Chloride consumption (moles/mole)	0.15	0.18	0.49	0.03	-----	0.06	0.08	0.14	0.55	0.35	0.50	0.20	0.19	0.53	-----	-----

APPENDIX B
TABLE 13 (continued)

22	23	24	31	32	33	34	35	36	37	38	39	41	42	43	44
100.	200.	100.	200.	98.	200.	200.	100.	200.	202.	100.	100.	200.	100.	200.	50.
95.	200.	98.	195.	98.	200.	190.	80.1	-----	180.	100.	102.	200.	101.	201.	48.
196.	197.	187.	185.	192.	188.	195.	193.	195.	186.	183.	188.	186.	184.	190.	192.
3.5	5.5	3.5	5.5	5.5	5.5	5.5	5.5	5.5	3.5	3.5	5.5	3.5	5.5	5.5	5.5
14,820.	23,270.	14,820.	13,000.	13,000.	13,000.	13,000.	13,000.	13,000.	8,250.	8,250.	13,000.	8,250.	13,000.	13,000.	13,000.
20.	22.5	7.9	13.5	10.2	21.6	12.0	13.3	10.3	13.1	9.5	-----	-----	-----	-----	-----
3.	3.	8.	3.	3.	3.	4.	3.	3.	3.5	3.	8.	3.	4.	4.	3.
7.15	6.94	7.10	7.20	7.20	7.03	7.02	7.14	7.17	7.02	6.48	7.03	7.27	7.29	7.27	7.18
72.91	67.7	33.5	37.2	37.2	32.7	37.2	37.3	37.4	37.8	33.3	34.8	34.2	34.8	34.5	34.9
4.88	4.55	6.05	5.86	5.86	5.92	-----	-----	-----	5.64	4.43	5.40	5.48	5.50	5.96	6.16
1.190	1.186	1.205	1.208	1.208	1.210	1.206	1.207	1.209	1.200	1.189	1.206	1.211	1.212	1.210	1.209
0.05	0.36	0.40	4.65	2.65	2.85	1.20N ^B	1.82N ^B	1.00N ^B	3.19	4.19	4.33	3.79	3.49	2.67	0.0
92.5	86.0	36.7	30.6	37.7	31.4	38.3	38.7	-----	31.3	29.4	29.1	29.3	30.3	31.0	37.1
5.6	4.22	5.47	5.27	-----	5.02	-----	-----	-----	4.92	4.83	5.0	5.24	5.14	5.38	4.72
143.5	113.	112.5	38.5	72.4	68.7	22.0	50.2	-----	49.6	34.5	36.4	49.3	53.8	73.1	126.7
31.2	28.2	22.5	9.28	-----	15.9	-----	-----	-----	-----	-----	-----	11.4	12.9	17.3	31.1
-----	-----	17.7	-----	-----	-----	-----	-----	-----	12.3	6.4	11.2	13.2	12.8	16.6	26.1
-----	-----	-----	-----	-----	-----	55.3	50.9	-----	-----	-----	-----	-----	-----	-----	-----
1.448	1.386	1.391	1.261	1.322	1.308	1.343	1.374	1.331	1.290	1.243	1.264	1.285	1.293	1.337	1.435
1.3	1.3	1.0	0.75	0.70	1.55	1.3	0.9	-----	1.82	0.60	0.44	1.79	0.61	1.65	0.71
0.17	0.20	0.10	0.20	0.12	0.20	0.21	0.10	0.25	0.32	0.08	0.07	0.11	0.10	0.20	0.10
0.05	0.04	0.04	0.21	0.06	0.12	0.08	0.03	-----	0.26	0.10	0.07	0.20	0.08	0.11	0.03
2.2	2.54	2.58	3.2	3.02	2.97	3.3	3.06	-----	3.72	2.78	2.86	2.83	2.86	2.58	2.23
-----	-----	-----	0.23	0.08	0.03	-----	-----	-----	0.18	0.13	0.17	0.11	0.10	0.06	-----

PAGES 60 to 61
WERE INTENTIONALLY
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APPENDIX B
TABLE 14
SUMMARY OF CHLORIDE REMOVAL EXPERIMENTAL DATA

Run No.	1	2	3	4	6	7	8a	8b	10a	10b	11	12	13	14	15	16	17a
Run Conditions																	
Feed Rate (cc/min)	63.5	63.3	57.4	100.5	58.0	45.0	54.0	55.0	43.0	50.0	40.0	40.0	41.0	40.0	40.0	40.0	65.0
Product Rate (cc/min)	35.3	39.7	35.3	59.2	38.0	27.0	40.0	31.5	25.0	25.0	22.0	22.5	25.0	25.0	28.0	23.0	43.0
Overhead Rate (cc/min)	28.2	23.6	22.1	41.3	20.0	18.0	14.0	23.5	18.0	25.0	17.5	17.5	15.0	15.0	12.0	19.0	22.0
Column Dia. (inch)	2.0	2.0	2.0	2.0	2.0	2.0	2.0	2.0	2.0	2.0	2.0	2.0	2.0	2.0	2.0	2.0	2.0
Type of Packing	1/4-inch Berl Saddles								1/4-inch Berl Saddles								
Packing Height (ft)	9.5	9.5	9.5	9.5	9.5	9.5	9.5	9.5	9.5	9.5	9.5	9.5	9.5	9.5	9.5	9.5	9.5
Run Duration (hr)	7.0	5.0	11.0	5.0	20.0	8.0	11.0	11.0	13.0	13.0	17.5	7.0	15.0	7.0	7.0	8.0	5.0
Stream Analysis																	
Feed																	
H ⁺ (N)	7.44	7.44	7.62	7.72	7.49	7.50	7.45	7.45	7.52	7.52	8.08	8.01	6.91	7.75	7.99	7.97	7.27
Cl ⁻ (g/l)	22.5	22.5	25.4	23.8	18.3	18.0	18.5	18.5	20.0	20.0	18.9	17.6	18.6	18.0	21.3	20.9	27.5
NO ₃ ⁻ (M)	6.9	7.34	7.48	7.88	7.82	7.88	7.70	7.70	8.27	8.27	8.80	8.47	7.82	7.76	8.2	8.49	7.99
Ni (g/l)	15.8	14.7	15.8	24.9	18.2	14.0	17.2	17.2	30.7	30.7	2.71	2.27	29.9	26.2	25.7	26.3	30.0
Cr (g/l)	4.1	3.8	4.0	5.6	4.57	4.16	4.25	4.25	7.03	7.03	6.33	5.77	6.50	5.88	6.06	5.98	6.2
Sp. Gr.	1.265	1.276	1.280	1.307	1.291	1.293	1.290	1.290	1.326	1.326	1.337	1.327	1.311	1.316	1.318	1.316	1.316
Product																	
H ⁺ (N)	9.87	10.7	10.1	10.5	10.3	10.7	9.53	9.83	9.90	10.4	10.4	10.0	9.01	8.61	9.85	10.7	9.16
Cl ⁻ (g/l)	0.18	0.079	0.109	0.051	0.042	0.0025	0.028	0.005	0.005	0.005	0.0025	0.0047	0.157	0.13	0.007	0.007	0.057
NO ₃ ⁻ (M)	11.1	11.5	11.2	11.6	11.7	12.0	10.5	10.9	12.2	12.4	12.5	11.2	11.4	9.38	10.1	12.5	11.4
Ni (g/l)	26.5	30.2	27.9	28.4	28.5	28.0	27.2	31.8	52.7	50.6	37.6	51.7	50.0	33.0	48.4	55.0	49.1
Cr (g/l)	6.4	7.0	6.8	8.32	7.43	7.16	6.20	6.65	12.1	12.1	10.2	7.76	10.5	7.48	9.70	10.5	10.7
Sp. Gr.	1.380	1.345	1.384	1.416	1.410	1.417	1.382	1.388	1.457	1.464	1.455	1.426	1.435	1.368	1.410	1.447	1.427
Overhead																	
H ⁺ (N)	3.71	3.78	3.85	3.96	3.18	3.22	5.13	3.43	4.71	3.52	4.31	4.39	3.64	4.59	4.94	3.83	5.11
Cl ⁻ (g/l)	49.5	58.8	57.3	48.4	40.1	45.0	82.5	47.0	58.1	35.0	52.7	56.0	41.9	59.7	65.6	42.4	71.2
NO ₃ ⁻ (M)	2.2	2.33	2.33	2.51	1.88	1.80	2.69	2.02	2.6	2.35	2.8	2.82	2.52	3.11	2.80	2.62	3.08
Sp. Gr.	1.094	1.097	1.102	1.120	1.087	1.084	1.126	1.089	1.121	1.096	1.120	1.119	1.094	1.125	1.126	1.106	1.135
Material Balance (Overall Per cent)																	
H ⁺ (N)	96.0	108.9	101.1	101.3	104.6	102.4	112.7	95.4	103.1	92.6	94.7	91.6	98.9	91.6	104.7	100.0	107.0
Cl ⁻ (g/l)	98.3	98.1	87.1	83.7	75.9	100.0	115.7	108.5	121.5	87.4	125.7	123.3	82.8	124.7	92.6	96.8	87.7
NO ₃ ⁻ (M)	97.4	110.1	104.0	99.7	106.4	100.3	110.1	92.2	98.9	89.1	92.3	87.3	100.6	91.0	96.6	99.1	107.5
Ni (g/l)	100.0	128.6	108.7	67.2	102.7	120.0	117.1	105.7	101.8	85.8	93.5	93.3	102.0	78.7	131.4	120.2	108.3
Cr (g/l)	93.8	115.4	104.3	88.4	106.4	103.2	107.8	89.3	100.3	86.1	80.6	75.8	98.5	79.6	112.4	100.4	114.1
Mass	98.3	100.6	99.6	99.0	100.6	99.3	102.0	96.3	99.3	96.5	91.5	93.1	97.3	97.0	100.5	103.1	100.9
Results																	
Mole % Acid in Feed	14.5	14.5	15.0	15.1	14.8	14.7	14.6	14.6	14.7	14.7	14.7	15.8	13.9	13.9	15.2	16.1	14.5
Molar Vapor/Liquid Ratio (V/L)	0.5	0.39	0.41	0.44	0.39	0.43	0.27	0.45	0.44	0.52	0.47	0.47	0.39	0.39	0.32	0.52	0.35
Number of Transfer Units	2.72	3.78	3.54	3.70	4.0	5.0	6.55	4.66	4.73	3.99	4.52	3.51	3.42	3.83	6.0	3.97	4.35
Height of a Transfer Unit (ft.)	3.50	2.51	2.69	2.56	2.37	1.9	1.45	2.04	2.01	2.38	2.10	2.70	2.78	2.49	1.58	2.39	2.18
Chloride DF	125.0	285.0	233.0	467.0	436.0	7200.0	661.0	3700.0	4000.0	4000.0	7560.0	374.0	118.0	138.0	3040.0	2980.0	----

APPENDIX B
TABLE 14 (continued)

17b	18	19a	19b	21	22a	22b	23	24	25	26	27	28	29	30	31	32	33	34	35
60.0	60.0	60.0	60.0	80.0	60.0	60.0	60.0	50.0	50.0	101.0	101.9	98.0	98.9	100.0	212.0	202.0	200.0	200.0	307.0
35.0	34.6	23.2	25.0	52.5	43.0	38.0	33.0	32.5	39.0	33.8	43.5	49.3	63.4	68.2	108.5	115.9	125.0	150.0	206.0
25.0	25.4	36.8	35.0	27.5	17.0	22.0	27.0	18.1	12.0	67.6	59.1	47.3	35.6	26.7	108.2	80.4	80.0	48.0	108.0
2.0	2.0	2.0	2.0	2.0	2.0	2.0	2.0	2.0	2.0	4.0	4.0	4.0	4.0	4.0	4.0	4.0	4.0	4.0	4.0
3/16-inch glass helices										1/4-inch Berl Saddles									
9.5	9.5	9.5	9.5	9.5	9.5	9.5	9.5	9.5	9.5	7.5	7.5	7.5	7.5	7.5	7.5	7.5	7.5	7.5	7.5
5.0	5.0	5.0	5.0	6.0	7.0	7.0	7.0	5.0	6.0	4.0	4.0	4.0	4.0	4.0	7.0	5.0	4.0	4.0	5.5
5.30	7.23	4.52	4.78	4.48	7.21	7.25	7.72	7.10	7.52	8.83	8.55	8.24	8.34	8.36	8.72	7.55	7.52	7.61	7.62
37.5	24.1	36.3	33.3	26.8	20.3	20.0	20.4	16.0	16.0	19.2	22.0	21.49	21.1	22.4	3.56	1.74	18.0	17.1	18.6
6.30	7.88	5.58	5.53	6.19	8.14	8.14	8.41	7.18	7.82	9.62	9.05	0.36	8.77	9.00	9.33	8.28	9.26	8.54	8.20
39.4	22.6	37.4	37.9	48.8	27.5	26.6	25.0	19.4	22.8	30.4	31.6	34.4	35.8	34.4	19.9	25.6	12.8	36.9	25.9
8.73	5.24	8.60	8.56	15.2	6.0	6.0	5.64	6.20	7.44	6.52	7.0	7.97	7.45	7.67	4.87	5.25	21.5(A1)	8.26(U)	6.38
1.293	1.304	1.268	1.268	1.305	1.315	1.315	1.316	1.278	1.310	1.349	1.355	1.358	1.358	1.362	1.329	1.317	1.372	1.345	1.312
9.16	9.00	8.13	7.68	8.75	7.74	7.87	10.4	8.34	7.98	11.3	11.0	10.5	9.94	9.47	12.5	11.6	9.10	8.12	9.34
0.068	0.050	0.435	0.560	0.085	0.470	0.940	0.014	1.97	0.69	<0.005	0.008	0.015	0.079	0.047	<0.005	<0.005	0.010	0.125	0.348
11.3	11.6	11.7	11.7	11.9	10.9	10.7	12.3	8.97	9.05	13.6	13.7	13.4	11.9	11.3	14.0	13.2	10.62	9.87	11.0
46.9	46.9	68.6	79.8	66.8	58.0	51.0	45.1	29.7	28.8	79.1	61.7	61.3	53.3	48.9	44.3	42.4	19.9	48.8	41.8
10.7	11.3	17.1	18.8	10.7	14.0	12.8	8.96	7.81	9.36	16.2	13.6	13.9	12.3	11.4	9.59	9.4	8.0(A1)	11.0(U)	9.98
1.429	1.433	1.505	1.505	1.486	1.440	1.431	1.442	1.345	1.345	1.536	1.509	1.503	1.469	1.441	1.478	1.459	1.381	1.406	1.401
3.27	4.31	3.24	4.59	4.53	4.32	3.91	3.68	2.80	4.81	7.78	5.81	4.57	4.86	5.33	3.98	2.14	4.45	4.64	4.23
46.8	56.1	26.9	52.2	65.9	56.1	45.0	45.4	40.9	73.1	26.4	38.5	43.7	50.3	63.0	72.1	46.0	51.2	50.0	43.6
2.16	2.61	1.80	1.78	2.37	2.69	2.58	2.67	1.57	2.95	6.8	4.58	3.78	3.11	3.42	3.4	1.93	4.24	3.08	2.82
1.088	1.110	1.106	1.108	1.112	1.110	1.106	1.106	1.072	1.119	1.228	1.164	1.134	1.131	1.141	1.119	1.066	1.130	1.123	1.115
126.7	96.8	113.7	123.0	163.1	93.8	88.5	95.7	90.7	98.1	101.6	86.7	91.0	85.2	94.3	96.4	99.4	100.0	94.7	101.8
52.1	98.6	46.0	92.1	84.9	80.2	85.5	100.6	100.6	113.3	92.0	101.8	98.0	86.1	75.3	103.3	105.4	114.0	70.8	94.9
119.0	98.9	100.9	106.9	139.2	105.7	95.1	94.9	89.1	99.2	94.7	93.8	91.6	99.8	95.8	76.9	100.7	90.0	95.3	101.7
69.5	119.8	70.9	83.8	89.8	151.2	121.4	99.1	99.6	98.5	87.0	83.2	89.6	95.5	96.8	113.9	94.9	97.1	99.3	108.7
71.6	124.5	76.7	91.4	46.2	167.2	135.0	87.6	81.9	117.7	83.2	83.1	87.7	105.7	101.3	100.9	102.6	23.3(A1)	101.0(U)	105.1
99.5	99.4	97.2	100.4	104.0	112.5	99.7	98.1	98.7	102.1	99.0	98.8	95.9	99.3	94.6	99.9	95.8	95.8	98.5	101.3
10.1	14.7	8.6	8.8	7.8	14.4	14.4	15.4	13.7	13.7	18.0	16.9	16.5	16.3	16.4	17.6	14.7	14.5	15.0	15.0
0.42	0.44	0.61	0.60	0.33	0.27	0.39	0.48	0.38	0.24	0.64	0.60	0.50	0.37	0.28	0.55	0.42	0.39	0.25	0.37
3.67	3.67	1.89	1.77	4.74	4.43	1.96	3.97	2.27	----	3.89	3.83	3.82	3.89	5.99	3.0	3.0	4.84	5.71	3.0
2.58	2.58	5.01	5.36	2.0	2.15	4.85	2.39	4.18	----	1.92	1.96	1.96	1.92	1.25	2.5	2.5	1.55	1.31	2.5
----	----	----	----	----	----	----	----	----	----	3840.0	2750.0	1470.0	270.0	370.0	710.0	350.0	1800.0	140.0	54.0

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APPENDIX B
TABLE 15
SUMMARY OF SOLVENT EXTRACTION EXPERIMENTAL DATA
Two Inch Diameter Column
Two-Inch Plate Spacing-Standard Cartridge
Plate Section-15.2 Feet

Run No.	1	2	3	4	5
Run Conditions					
Aqueous Feed Rate (cc/min.)	330.	341.	333.	330.	340.
Organic Feed Rate (cc/min.)	340.	330.	350.	325.	333.
Volume Velocity (GHSF)	500.	500.	510.	489.	503.
Aqueous to Organic Ratio (A/O)	0.97	1.03	0.95	1.02	1.02
TBP Conc. (Vol %)	5.	5.	5.	5.	5.
Pulse Frequency (cycles/min.)	55.	55.	55.	57.	55.
Pulse Amplitude (in.)	1.0	1.0	1.0	1.0	1.0
Stream Analysis					
Aqueous Feed					
H ⁺ (N)	4.65	5.25	3.80	2.99	3.09
U (g/l)	4.15	4.64	4.99	9.67	3.66
Ni (g/l)	18.9	19.9	19.2	13.3	16.0
Cr (g/l)	4.5	4.53	5.21	3.4	4.1
Al (g/l)	-----	6.75	7.73	37.2	6.4
sp. gr.	1.259	1.285	1.251	1.362	1.197
Raffinate					
U (g/l)	0.00075	0.0031	0.0020	0.0045	0.0019
sp. gr.	1.250	1.281	-----	1.355	-----
Organic Prod.					
U (g/l)	4.13	4.86	4.16	9.73	3.65
Material Balance					
U (%)	103.	101.	87.7	99.1	98.2
Results					
Uranium Waste Loss (%)	0.015	0.067	0.04	0.15	0.06
Number of Transfer Units (NTU)	10.22	8.79	9.29	7.70	8.83
Height of a Transfer Unit (HTU) in ft.	1.49	1.73	1.64	1.97	1.72
Flooding Frequency (cycles/min.)	70.	-----	77.	77.	-----

APPENDIX C

Calculations of the height of a transfer unit (HTU) were performed in the following manner:

1. The operating line was determined by a material balance around the column. For run S-28, the operating line is

$$y = 1.98 x - 0.0008 \quad (1)$$

where x = conc. of chloride in liquid ($\underline{M}$)
 y = conc. of chloride in vapor ($\underline{M}$)

2. The equilibrium line used was that completed by Roberts, et al. (6).

3. The equilibrium and operating lines were plotted on log-log paper and stages stepped off as shown in Figure 26. Number of stages were 3.8.

4. The height of a transfer unit (HTU) was determined by dividing the height of packing used in the column by the NTU's; this gave the HTU, i.e., 1.97 feet for run S-28.

An identical procedure was used to determine the HTU's in the solvent extraction studies.

Figure 26

Equilibrium Relationship of
Chloride-Nichrome Solutions

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