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UNITED STATES ATOMIC ENERGY COMMISSION

ZIRCONIUM-PRECIPITATION PILOT PLANT

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MASSACHUSETTS INSTITUTE OF TECHNOLOGY
ENGINEERING PRACTICE SCHOOL
CARBIDE AND CARBON CHEMICALS DIVISION
Union Carbide and Carbon Corporation

MEMORANDUM

(KT-86)

EPS-Y111

September 30, 1950

TO: T. H. Pigford
FROM: G. T. Parish (Leader), R. Bakal, E. I. Goodman, H. A. Larsen
SUBJECT: Zirconium-Precipitation Pilot Plant

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INTRODUCTION

Salicylic acid is currently being used to precipitate zirconium salicylate from an aqueous solution of zirconium chloride. The precipitate is removed by filtration and calcined to form zirconium oxide, with a consequent loss of the salicylate used. The oxide is then reduced to zirconium metal.

The method used in the present investigation precipitates the zirconium as zirconyl phthalate which is, in turn, converted to zirconium hydroxide by leaching with concentrated ammonium hydroxide. The hydroxide is filtered, and the phthalate may be recovered in the filtrate and recycled. The chemical reactions involved are

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Laboratory investigations by Googin, Sprague, and McLeod show that as much as 80% of the phthalic acid may be recovered in the filtrate and reused (1).

The object of this investigation is twofold: First, to improve the design of an existing pilot plant to allow better control of operating variables than was attained by previous investigators (1), and second, to determine the effect of leaching time on phthalate recovery.

This investigation began September 13, 1950, and terminated September 29, 1950.

RESULTS

The pertinent results are presented below.

TABLE I

Run No.	Hold-up Time in Leaching Tank 57 (Min)	Leach Slurry pH	N*	% Phthalate Recovered
1	57		0	28
2	57	4.9	-0.01	26
3	33	5.3	-0.01	29
4	12	7.2	-0.01	29
5	12	9.3	0.13	58
6	30		0.16	73

N* Excess ammonia in no. 2 filtrate (equivalents/liter)

DISCUSSION OF RESULTS

Insufficient data have been taken to definitely establish the effect of ammonium hydroxide concentration and hold-up time during the leaching process. During the first four runs the ammonium hydroxide added during the leach was less than the stoichiometric value instead of the desired 5% excess. The data for these runs is not significant because the reaction could not be completed regardless of hold-up time.

From Run 5 to Run 6 the percent phthalate recovery increased by 26%; the percent excess ammonia increased by 23%, and the hold-up time increased by 150%.

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These changes seem to indicate that the percent phthalate recovery is insensitive to the hold-up time but changes more rapidly with the excess ammonia in the leaching solution. Future investigations of the phthalate recovery in this system should focus on the effect of the percent excess ammonia used during leaching and the concentration of the ammonium hydroxide used. The effect of hold-up time could be determined after the establishment of the optimum conditions for the leaching solution.

If the percent phthalate recovery is really insensitive to changes in hold-up time as Runs 5 and 6 indicate, then the percent phthalate recovery might be determined indirectly by a measurement of the excess ammonia of the leached slurry as it goes to the No. 2 filter. The most rapid determination of this excess would be a measurement of the pH of a diluted sample of the slurry.

Material Balances

Zirconium material balances (see Appendix) reveal that, due to poor functioning of the filters, approximately 35% of the feed zirconium went into the filtrates as solids. The balances also leave a large portion of zirconium unaccounted for. It is possible that this zirconium was in solution in one or both of the filtrates. These filtrates were not analyzed for zirconium in solution because previous work had indicated that very little zirconium was lost in the filtrates (1).

The wide scatter of the material balances for both zirconium and phthalate probably reflect the erratic filter operation. It was necessary to adjust the vacuum often and sometimes to run the wash water only intermittently. For these reasons it was difficult to obtain representative samples of the cakes and filtrates.

Filters

As high as 3% solids was observed in the filtrates. The filter masking was removed in two steps, as described in the section (A) on Equipment Modifications, but no trend was observed to indicate that high filtration rates were the cause of the solids passing through the filters. The solids may have passed through the observed leaks at the edges of the filters. These leaks must be repaired before studying filtration characteristics quantitatively.

The filter cakes produced contained from 60% to 75% moisture. Positive pressure type filters, such as plate-and-frame filters, would be more appropriate for filtering these finely divided precipitates (3).

Foaming in No. 1 Hold-up Tank

Previous workers suggested the use of additives to prevent the excessive foaming found in the No. 1 hold-up tank when the temperature was allowed to go above 95°C (1). The present investigation showed that the temperature in the hold-up tank could be readily maintained at the proper level, and that foaming occurred only

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when the solution actually boiled. Temperature control should be adequate to eliminate this difficulty, making the addition of anti-foamants unnecessary.

EQUIPMENT MODIFICATIONS

The group which operated the pilot plant just prior to the present investigation reported a great deal of operating difficulty (1). Consequently, several equipment modifications were made in an effort to eliminate these difficulties, making the control of operating variables more dependable.

Flow Measurements

As the system was originally designed, flow rates were to be controlled by means of calibrated finger pumps. Previous work has shown these finger pumps to be unreliable for metering, and it was therefore recommended that rotameters be installed in the principle flow lines. Rotameters were inserted in the zirconium feed, ammonium phthalate, leach solution, and the two wash solution lines.

Because of the difficulty in priming the finger pumps and because they were no longer required for metering, the ammonium phthalate feed and the two wash solutions were fed by gravity from tanks on the balcony, and their respective pumps were removed from the system. As the liquid level in the ammonium phthalate tank decreased the maximum flow rate obtainable decreased below the desired value of ten gallons per hour. This condition can be corrected by using larger tubing in the feed line or by using a second feed barrel in parallel with the first.

Finger pumps were still used to supply the zirconium feed and leach solution. In order that flows of ten gallons per hour could be maintained without running these pumps at excessive speeds, two rubber tubes, one on each side of the fingers, were placed in parallel through the pumps instead of the customary single tube. This arrangement worked satisfactorily and resulted in a steady flow which did not cause bothersome surging of the rotameter bobs.

Originally the hold-up tank levels were controlled by gravity legs. The previous group found that these legs became clogged with solids. These gravity legs were removed and the flow from hold-up tanks to their respective filters was controlled by finger pumps. The use of small tubing (to give higher flow velocities) and the pulsing action of the pumps prevented any settling of solids in these lines.

Pre-heater

Laboratory data have shown that it is desirable to pre-heat the zirconium chloride feed to approximately 90°C before reacting it with the phthalate. The pilot plant was designed for a feed rate of ten gallons per hour, but the pre-heater was inadequate to heat more than seven gallons per hour to the desired temperature. A second heating coil was added to double the heating area.

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ZIRCONIUM PRECIPITATION PILOT PLANT

DATE 11-20-50	DRAWN BY G.T.P.	FILE NO. EPS Y-III	FIG. 1
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Mixing in Hold-up Tank No. 1

It had been recommended that additional agitation be applied to the zirconyl phthalate slurry (1) (2). Accordingly, a centrifugal pump had been set up so that zirconium chloride and ammonium phthalate were introduced at the pump inlet and so that the mixture was continuously recycled through the pump during the hold-up time. This arrangement was used for Run 1, but difficulty was experienced in filtering the thick foam produced. The original mixing equipment was used for all subsequent runs, and it functioned satisfactorily.

Masking of Oliver Vacuum Filters

Laboratory data indicated that the installed filters were considerably larger than was necessary (1). Therefore, approximately 80% of the effective area of each filter was masked with a vinylite sheet. In actual operation it was found necessary to remove some of this masking from both filters to achieve satisfactory filtration. Eventually all of the masking was removed.

Control of Vacuum on Oliver Filters

It was possible to control the vacuum within the filters by bleeding air into a hole in the distribution plate of each filter. On filter No. 1 this hole was tapped for $\frac{1}{4}$ -inch pipe; on No. 2, $\frac{1}{8}$ -inch pipe. Valves were installed on each filter at these holes, but were found to be too small to permit the proper range of vacuum control. The valves were removed, and notched rubber stoppers were used to control the vacuum. Large bleeder valves are currently being installed in the main vacuum lines between each filter and its respective filtrate tank.

CONCLUSIONS AND RECOMMENDATIONS

1. Both the excess amount of leach ammonia and the leaching time are important variables in the conversion of zirconyl phthalate to zirconium hydroxide; the effect of the excess ammonia being greater than that of the hold-up time.
2. Additional investigations should be carried out on the modified zirconium precipitation pilot plant with particular emphasis on the factors which control the leaching step. These variables are, in order of importance: excess ammonia in the leaching solution, concentration of ammonia in the leaching solution, and reaction time. A simultaneous study should be made of the possible use of the pH of the slurry coming from the reaction tank as a measure of the excess leach ammonia.
3. Future analyses for carbon in the zirconium hydroxide cake should be made by means of a carbon train. (See Appendix C.)
4. Provision should be made for stabilizing the ammonium-phthalate flow rate by maintaining a sufficient gravity head from the feed barrel.

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5. Bleeder valves should be installed on vacuum lines to the Oliver filters.
6. Drains should be installed in the premixer, the ammonium-phthalate recycle drum, and the catch basin under the No. 1 filter to facilitate draining and cleaning.

PROCEDURE

Equipment

A flow sheet of the process and the pilot plant equipment is shown in Fig. 1.

Starting up

All solutions were made up according to the specifications given below. The ammonium phthalate was brought up to approximately 90°C by means of a steam coil in an agitated, 55-gallon, stainless steel drum. Zirconium chloride feed was pumped at a rate of ten gallons per hour into an agitated preheater where the temperature was maintained at 80°C to 85°C. As soon as the zirconium feed overflowed into the premixer, the hot ammonium phthalate was fed by gravity into the premixer, and the agitator was started. Overflow from the premixer was stirred and allowed to build up to 8.6 gallons in the No. 1 hold-up tank and was then pumped to the first Oliver rotary filter. Sufficient steam was admitted to the jacket of No. 1 hold-up tank to maintain a temperature of 85°C to 90°C. The first filter was started, using a differential pressure of 2 to 5 inches of mercury, and after a cake had been built up to the point where it was removed by the doctor knife, the filter wash-water and the air blow-back were started. The ammonium hydroxide leaching solution was added to the cake in the re-slurrying trough, and the slurry flowed by gravity to the No. 2 hold-up tank. The level in No. 2 hold-up tank was controlled to maintain the desired reaction time for each run. When this level was reached, the slurry was pumped to the No. 2 Oliver filter. The filter was started using a differential pressure of 3 to 10 inches of mercury. Ammonium hydroxide wash and air blow-back were turned on after sufficient cake had been built up. During the present investigation the filtrate from the second filter was not recycled. The filtrates were collected, sampled, and discarded.

Normal Operation

The following data were recorded every fifteen minutes:

<u>Temperature</u>	<u>Flow Rate</u>	<u>Vacuum</u>	<u>Others</u>
Ammonium-phthalate Storage Tank	Ammonium phthalate	No. 1 Filter	Level in No. 1 Hold-up Tank

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Normal Operation (cont'd)

<u>Temperature</u>	<u>Flow Rate</u>	<u>Vacuum</u>	<u>Others</u>
Preheater	Zirconium chloride feed	No. 2 Filter	Level in No. 2 Hold-up Tank
No. 1 Hold-up Tank	Ammonia Leach Solution		Line Steam Pressure
No. 2 Hold-up Tank			

Although they were not recorded, the flow rates of the two filter-wash solutions were checked every fifteen minutes. Adjustments were made whenever any of the above-mentioned conditions varied from the pre-selected values.

In all runs except Nos. 1 and 6 two sets of samples were taken at each hold-up time of tank No. 2. The system was allowed to operate for 30 to 60 minutes after any change of conditions before samples were taken. The samples taken and the analyses made were as follows:

<u>Sample</u>	<u>Analysis</u>
No. 1 Filter Cake	Moisture (g/g)
No. 2 Filter Cake	Zirconium (g/g), Carbon (g/g), Moisture (g/g)
No. 1 Filtrate	Solids (g/g), Phthalate (lb/gal), Excess Ammonia (eq/l)
No. 2 Filtrate	Solids (g/g), Phthalate (lb/gal), Excess Ammonia (eq/l)
Zirconium Chloride Feed (two/batch)	Zirconium (lb/gal)
Ammonium Phthalate (two/batch)	Phthalate (lb/gal)

Control measurements of pH on the two slurries fed to the filters were also taken every 30 minutes. The two batches of leach solution used were analyzed for ammonia.

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Shutting Down

The zirconium chloride and ammonium phthalate feeds were turned off, and the preheater and premixer were turned off. The system was allowed to empty itself at the normal operating rates, and individual pieces of equipment were shut down as they became empty. The filters, particularly the filter drum and cloth, were washed and drained. All lines and tanks containing solids were flushed and drained to prevent plugging.

Preparation of Solutions

The analysis of the raw zirconium chloride feed was obtained at the time the feed was taken from the tank truck. Before use, the feed was diluted down to 0.6 lb/gal of zirconium. The pH was not adjusted because pH of the feed was found to have little effect on the first precipitation step.

The ammonium phthalate was made up in 40-gal. batches. For a 40-gal. batch the amounts used were 40.8 lb phthalic anhydride, 133.8 liters water, and 16.8 liters of 28 to 30% aqueous ammonia. The pH was measured but not adjusted.

The ammonium hydroxide concentration in the leach solution was 5.7% ammonium hydroxide by weight. Solution was made up in 40-gal. batches and required the following quantities: 134 liters of water and 17.5 liters of 28 to 30% aqueous ammonia.

The No. 1 Wash Solution was plant water.

The No. 2 Wash Solution was 1.5% ammonium hydroxide, and was made up in 40-gallon batches using 134 liters of water and 4.0 liters of 28 to 30% aqueous ammonia.

LOCATION OF ORIGINAL DATA

The original data are located in data books 1557 and 1565 and Zirconium Pilot Plant Sample Log (300 - 14U).

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APPENDIX

A. Operating Conditions and Data

TABLE I-A

Summary of Operating Conditions and Data

Run No.	1	2	3	4	5	6
1. Rates (gal/hr)						
Zirconium chloride	-----	10	-----	-----	-----	6.0
Ammonium Phthalate	-----	10	-----	-----	-----	5.5
Leach Solution	-----	10	-----	8	-----	7.5
No. 1 Filter Wash	-----	5	-----	-----	-----	-----
No. 2 Filter Wash	-----	2	-----	-----	-----	-----
2. Temperature (°C)						
Amn. Phthalate Feed Tank		78		87	89	72
Zirconium Chloride Preheater		83		81	84	85
No. 1 Hold-up Tank	70	88		88	89	89
No. 2 Hold-up Tank	28	29	29	34	33	29

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TABLE I-A (Cont't)

Run No.	1	2	3	4	5	6
3. Analyses*						
Ammonium Phthalate						
a. Phthalate conc. (lb/gal)	0.596	0.602	- - - -	0.587	- - - - -	- - - - -
Zirconium Chloride						
a. Zirconium conc. (lb/gal)	0.634	- - - -	0.735 batch diluted to 0.600			
No. 1 Filter Cake						
a. Moisture Content (%)	64.6	61.6	60.5	64.0	70.2	69.1
No. 2 Filter Cake						
a. Moisture Content (%)	68.3	69.6	70.7	71.4	69.7	72.8
b. Zirconium Content (% dry cake)	36.8	38.3	38.7	39.8	49.1	54.6
c. Carbon Content (% dry cake)	24.2	29.8	28.8	28.7	21.6	15.4
No. 1 Filtrate						
a. Solids (wt %)	1.13	0.21	0.17	1.94	0.14	1.09
b. Phthalate conc. (lb/gal)	0.0351	0.0900	0.132	0.0217	0.130	0.0400
No. 2 Filtrate						
a. Solids (wt %)	3.74				0.86	2.28
b. Phthalate conc. (lb/gal)	0.111	0.053	0.105	0.097	0.530	0.602

*Analyses recorded for Runs No. 1 and 6 were based on one set of samples while the other four runs used the average results from two samples.

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TABLE I-A (Cont'd)

Run No.	1	2	3	4	5	6
4. NH_4OH Concentration						
Leach Solution (Normality)	.4	.4	.4	1.0	2.7	2.7
No. 2 Wash Solution (% NH_4OH)	1.54	1.54	1.54	1.54	1.54	1.54
5. pH						
Slurry to No. 1 Filter		1.3	1.4	0.2	0.6	
Slurry to No. 2 Filter		4.9	5.3	7.2	9.3	
6. Operating Conditions						
No. 1 Hold-up Time (min)	52	52	52	52	52	52
No. 2 Hold-up Time (min)	57	57	33	12	12	30
No. 1 Filter Drum Rate (rpm)	0.7	0.7	0.7	0.7	0.7	0.7
No. 2 Filter Drum Rate (rpm)	0.8	0.8	0.45	0.26	0.26	0.26
No. 1 Filter Vacuum (in.Hg)	5	5	5	2.5	2	2
No. 2 Filter Vacuum (in.Hg)	10	9	9	8	8	2
7. Zirconium Hydroxide Product						
Total Wet Weight (lbs)	33.7	27.1	9.8	20.8	16.6	8.31
Product Rate (lbs/hr)	8.4	13.5	9.8	11.0	11.0	10.3

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TABLE I-A (Cont'd)

Run No.	1	2	3	4	5	6
8. Material Balance						
Zirconium in (lb/hr)	6.34	6.00	6.00	6.00	6.00	3.60
Zirconium out (lb/hr)						
Filtrate No. 1	.79	.15	.12	1.35	.10	.51
Filtrate No. 2	1.37				0.42	0.98
Cake	0.98	1.57	1.10	1.25	1.64	1.53
% Zirconium Accounted for	51	71	80	57	64	16
Phthalate in (lb/hr)	4.89	4.94	4.82	4.82	4.82	2.65
Phthalate out (lb/hr)						
Filtrate No. 1						
Solids	1.42	0.26	0.21	2.44	1.76	0.91
Solution	0.88	2.25	3.30	0.54	3.25	0.66
Filtrate No. 2						
Solids	1.55				0.32	0.48
Solution	1.33	0.64	1.26	0.97	6.36	5.72
Cake	1.10	2.09	1.41	1.55	1.23	0.74
% Phthalate Accounted for	128	106	128	114	269	321

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B. Sample Calculations

Composition of "hydroxide" cake

The weight of carbon in one mole of zirconyl phthalate is $(8)(12) = 96$ g; the molecular weight of zirconyl phthalate is 272. The molecular weight of zirconium hydroxide is 160.

For Run 1 the laboratory analysis gave 36.76% zirconium and 24.18% carbon in the dry cake. These data provide a basis for two independent calculations of the composition of the dry cake.

On a basis of 100 g dry cake there is

$$(272)(24.2)/96 = 68.6 \text{ g zirconyl phthalate.}$$

The zirconium hydroxide is then 31.4 g by difference.

The second method is based upon the total zirconium concentration. Let P be the weight of zirconyl phthalate in 100 g dry cake and $(100-P)$ be the weight of hydroxide. Let M be the total moles of zirconium present in 100 g dry cake.

$$M = 36.76/92 = 0.399$$

Then $P/272 + (100-P)/160 = M$. By rearranging the terms,

$$P = 388(0.625 - M)$$

$$P = 87.6 \text{ g and } (100-P) = 12.4 \text{ g}$$

From these two methods the average analysis of the dry cake is

zirconyl phthalate 78.1%

zirconium hydroxide 21.9%

The percent of the phthalate lost in the filter cake is then

$$(78.1 \times 100)/(0.399 \times 272) = 72.1\%$$

C. Error Analysis

The laboratory analysis of "hydroxide" cake checked within 2% for carbon analysis and within 3% for zirconium analysis on two determinations for each sample. These deviations lead to errors of 1% in the % phthalate as calculated from the carbon train analysis, and 3% as calculated from the zirconium analysis. From these two methods the error in the average analysis of the dry cake is 3%.

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D. Methods of Analysis

Hydroxide Cake from Filter No. 2

The hydroxide cake is first dried to remove the moisture. It is analyzed for carbon by slowly burning a sample in a carbon train. The carbon dioxide evolved is then adsorbed and weighed. The use of 0.1 gm samples for cakes high in carbon content eliminates the problem previously encountered of fouling the train with incompletely burned carbon. Another dry sample is converted to ZrO_2 by ignition at $1000^\circ C$. These analyses give two independent methods for calculating the amount of zirconyl phthalate and zirconium hydroxide present in the dry cake, assuming that no other solids are present.

An alternate method of analyzing for phthalate and hydroxide suggested by Lea et al, (1), was tried unsuccessfully. The suggestion consisted of dissolving the zirconium hydroxide from the phthalate with 1 N hydrochloric acid. The zirconyl phthalate was to remain undissolved, and it could be filtered, dried, and weighed. Difficulty was encountered in dissolving the hydroxide from the cake. Samples known to contain zirconium hydroxide were analyzed by this method, and no hydroxide was detected.

Excess Ammonia

In the analysis for excess ammonia a known amount of hydrochloric acid is added to a sample. This acid is then back titrated to a methyl orange end point with standardized sodium hydroxide.

Zirconium

Zirconium in solution is analyzed gravimetrically by precipitation in a solution of phenylarsenic acid.

Phthalate

Phthalate is analyzed gravimetrically by precipitation with lead acetate.

Moisture

The amount of moisture in the filter cakes is determined by the weight loss after heating for one hour at $250^\circ C$.

E. Literature Citations

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