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PILOT PLANTS SECTION

REPORT FOR MAY, 1950

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ORNL-741
Chemistry-Separation
Processes for Plutonium
and Uranium

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CHEMICAL TECHNOLOGY DIVISION

PILOT PLANTS SECTION REPORT FOR MAY, 1950

D. G. Reid

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0.0 ABSTRACT

The monthly report for the Pilot Plants Section of the ORNL Chemical Technology Division for the month of May is presented. The information presented includes:

1. Processing information and data on the first half of the Chalk River Separations Program being done by means of the Redox process.
2. Evaluation of the remote radio-frequency foam level indicator and the conductivity recorder as operational aids for the ORNL Chemical Waste evaporator.
3. Progress and program for the pilot planting of the Purex Process and the design of the ORNL Waste Uranium Recovery Unit.

CHALK RIVER SEPARATION

Technical - Rigstad, Gresky, Hylton, Sadowski, Winget

Non-Technical - Benson, Groover, Jennings, Ledbetter, Lockmiller, McLellan, Shields, Sexton, Shipwash, Strader, Summers, Thomas, Thompson, Wiggins, Purkey, Spangler

1.0 CHALK RIVER SEPARATION

1.1 Summary

The first three batches of highly irradiated uranium from Chalk River have been processed thru two plutonium cycles and concentrated for transfer to the laboratory for final processing.

Overall product losses thru the two extraction cycles and product concentration were in the order of 1.0 - 2.0%. Material balances ranged between 98-108%. Plutonium beta decontamination was in the order of 0.5 to 1.2 x 10^6 and gamma decontamination in the order of 5 to 8 x 10^5. The products contained unusual quantities of uranium due to mechanical difficulties during column operations making it necessary to include a partitioning cycle in the laboratory purification step.

The fourth batch of material was completed thru one extraction cycle and the fifth batch of slugs were charged to the dissolver. Complete data on all six runs of the Chalk River material will be made available in the monthly report for June.

1.2 First Cycle Operations

First cycle operations during the first two runs were normal and without incident. During the first portion of the third run difficulties were encountered in operation of the IA Extraction Column resulting in an undue loss (4-11%) of product in the IAW stream. The difficulties were determined to be

caused by the presence of water in the Solvent Feed Tank. Pumping of the water into the column resulted in dilution of the salting agent and high IAW loss. These difficulties were corrected by draining the water from the solvent feed tank and increasing the solvent flow rate.

A high loss (0.7%) was encountered during recycling of the Run 3 IAW solution because of low salting strength of the feed and flashing of solvent from the original IAW. This IAW material was combined with the "clean out" portion of the run and a adjusted ORNL-Modified Flowsheet run made. The raffinate from this re-worked material was not recycled.

1.3 First Cycle Results

Plutonium losses during the first cycle of Runs 1-3 were 0.13, 0.37, and 0.85% respectively. IAW flowing stream equilibrium losses were 0.04% for Run 1 and 0.10% for Run 2. High IAW losses were encountered in Run 3, apparently as a result of recycling the IAW which had been thru the solvent flashing procedure in preparation for transfer to the waste storage system.

Unusually poor separation from uranium occurred in the IB column apparently as a result of temporary instrument difficulties during operation. Air and steam blow-down of partially plugged instrument lines apparently caused some uranium to pass out with the IBP stream, resulting in uranium contamination of the IBP solution.

Beta and gamma decontamination factors for plutonium varied during sections of the first cycle runs from 5×10^3 to 1.5×10^5. Beta and gamma decontamination factors for uranium varied from $1\text{-}3.5 \times 10^4$. Additional data is given in Tables 1 and 2.

1.4 Second Cycle Operation

An attempt to use the KAPL-Modified Flowsheet in the first section of Run 1 resulted in plugging of the IIA column and coincidental losses of product. The IIAW's were recycled with the "clean-out" section of the run using the flowsheet developed at ORNL for the Redox IBP plutonium recovery process.

Runs 2 and 3 were made with the latter acidic flowsheet successfully, except for an occasional partial plugging of the instrument probes in the extraction column.

1.5 Second Cycle Results

Plutonium losses in the second cycle of Runs 1-3 were 0.26, 0.55, and 1.15% respectively. IIAW flowing stream losses were 0.04, 0.3, and 1.1% respectively. The high loss in Run 3 was apparently as a result of incomplete plutonium oxidation.

Beta and gamma decontamination factors ranged from 50 to 100. Ruthenium was the principal contaminant in the product. Additional data is given in Tables 1 and 2.

1.6 Evaporator Results and Operation

Due to the difficulty of flow control in the operation of the continuous two-stage evaporator and resultant losses in the second stage, product concentration was carried out in two batch operations.

Product losses in this step for Runs 1-3 were 0.51, 0.09, and 0.05% respectively. Recycling of condensates was found necessary in both Runs 1 and 2 which were made prior to conversion of the evaporators to a batch operation procedure. The high loss in Run 1 was due to the high aluminum concentration in the final product concentrate.

Products from the first three runs were concentrated to about two liters each, including evaporator washes.

1.7 Miscellaneous

The products from Runs 1-3 have been analyzed and are ready for final laboratory purification. Unduly high uranium content of the product will necessitate a partition cycle in the first processing. Analysis of the products are recorded in Table 3.

IAW's from Runs 1 and 2 were transferred to storage for the Isotope Development Department. The remaining IAW's will be transferred to the Fission Product Chemistry Group for the recovery of Np^{237}, 43, 61, and other elements.

Inadvertent delivery and pre-treatment of a batch of Carbide and Carbon Chemicals Division methyl isobutyl ketone led to re-evaluation of this material for possible use in Redox processings. Analysis of this batch of pre-treated material indicated that it would fall within process specifications as shown below. The use of the Carbide solvent had no noticeable adverse affect on decontamination as occurred when the material from this vendor was tested previously.

	Red. Norm.	HAc %	Color	Mesityl Ox.	R.I.
Pretreated Shell MIBK	0.000-0.024	<0.001-0.002	<1 γ/ml	--	--
Pretreated Carbide MIBK	0.017	0.002	<1 γ/ml	--	--
Raw Carbide MIBK	0.022*	--	--	0.1%	1.395

* MIBC = 0.11 %/wt.

1.8 Program

Pilot Plant processing of the six batches of highly irradiated uranium from Chalk River will be completed about June 15, after which processing of HEW irradiated uranium will begin. It is hoped that all processings will be completed in the canyon by July 1 and in the laboratory by August 1.

TABLE 1

205 Pilot Plant

Chalk River Separations Process: Product Losses in Runs 1-3

Run No.	LAMS 995 Batch No.	Slug No.	Section of Rods	Wt. U Kg.	Gms. Pu in Process			Mat. Bal. %	% Loss in Various Streams					
					Feed	Product	Waste		IAW	ICU	ICW	IIAW	IIBW	Evap.
1	6	40	1,13	133	34.1	32.9	0.5	98	0.08	0.05	<.001	0.55	0.25	0.51
2	5	40	2,12	137	65.0	64.5	0.65	100	0.21	0.16	0.002	0.52	0.02	0.09
3	4	40	3,11	142	92.4	98.1	1.8	108	0.71	0.13	0.011	1.14	0.01	0.05

TABLE 2

205 Pilot Plant

Chalk River Separations Process

Plutonium Decontamination in Runs 1-3

Run	D.F.'s ($\times 10^{-4}$) in First Cycle						Overall D.F.'s ($\times 10^{-6}$) in Two Cycles					
	β	γ	Ru	Zr	Cb	Ce	β	γ	Ru	Zr	Cb	Ce
1	9.0	3.0	0.6	3.0	10.0	400.0	1.0	0.8	0.3	--	--	--
2	1.0	1.0	0.1	1.0	2.0	55.0	0.5	0.6	0.09	0.7	1.0	100.0
3	0.8	0.4	0.1	0.4	0.3	4.0	0.4	0.5	0.06	--	--	--

TABLE 3

205 Pilot Plant

Chalk River Separations Process

Analysis of Feeds and Final Product in Runs 1-3

Run	FEED				PRODUCT			
	Tot. Pu (1AF)	Pu/U Ratio	β/Pu Ratio	γ/Pu Ratio	Tot. Pu (IIBP)	Pu/U Ratio	β/Pu Ratio	γ/Pu Ratio
	Gms.	Gm./Kg.	c/m/gm	c/m/gm	Gms. (approx.)	Gm/gm	c/m/gm	c/m/gm
1	34.1	0.256	2.43×10^{13}	4.86×10^{10}	32.9	0.2	1.63×10^{7}	2.76×10^{4}
2	65.0	0.473	2.01×10^{13}	4.55×10^{10}	64.5	1.2	3.8×10^{7}	5.33×10^{4}
3	92.4	0.650	1.82×10^{13}	3.98×10^{10}	98.1	4.0	4.0×10^{7}	4.6×10^{4}

RADIOCHEMICAL WASTE PROCESSING

Technical - Shank

Non-Technical - Davis, Grizzell

2.0 RADIOCHEMICAL WASTE PROCESSING DEVELOPMENT

2.1 Evaporator Operating Summary

The ORNL Radiochemical Waste Evaporator processed 167,000 gallons of mixed wastes with an average overall beta decontamination factor of 7.7×10^3 and an average condensate discharge activity of 1.3×10^{-9} curies/cc. The decontamination was higher than usual because of a lower processing rate; however, increased activity of the feed yielded an evaporator condensate with higher activity than normal.

2.2 Development Progress

2.21 Foam Characteristics of Evaporator Feed

A second attempt at investigating evaporator feed foam characteristics was made this month using a redesigned heating coil and metal waste supernate solution. Sufficient foam was produced to furnish the information required for a adequate evaluation of the proposed radio-frequency foam level indicator installation as mentioned in the Pilot Plants February Report, ORNL-655. The results and conclusions are listed below:

Results

(1) The foam level within a 6" pipe was not equal to that in the main tank with the difference being inconsistent in both magnitude and direction.

(2) The magnitude of deviation of the foam in the pipe was dependent on the position of the test pipe with respect to the heating elements.

(3) The conductivity of the foam was twice that of 0.05 M H_2SO_4 solution.

Conclusions

(1) The irregular deviations of foam rise within the test pipe indicated the inability of the proposed instrument installation to indicate foam level accurately.

(2) The conductivity of the foam is sufficient for such an installation.

2.22 Resin Capacity for Foam-over

Condensate samples taken after evaporator foam-overs were analyzed for sodium and aluminum content to determine the possible effectiveness of using resin columns to retain evaporator foam-overs without displacing previously adsorbed condensate activity. The capacity of the cation resin, (IRA-400) as established in the laboratory, is approximately 60 grams sodium/liter of resin. Concentrations of sodium in moderate foam-overs were as high as 56 grams/liter permitting only 40 gallons of foam-over solution to be processed before saturating the resin column containing 5 cubic feet of resin. This represents six minutes of operation at 400 gph to saturate the cation resin and permit a large amount of prior adsorbed activity to be elutriated.

2.23 Evaluation of Condensate Conductivity Cells for Evaporator Foam-Over Indication

Evaluation of the evaporator condensate conductivity cell installed last month (ORNL-702) was completed this month with the following results and suggestions.

Results

(1) All foam-overs, as determined visually, resulted in a full scale deflection of the recording instrument.

(2) High entrainment containing beta activity of approximately 10^3 cts/min/ml resulted in 1/4 to 1/2 scale deflection.

(3) The pre-indication of foam-overs was not consistent, but pen movement proved useful as an operational control and reduced the number of foam-overs.

Suggestions

(1) The alarm point should be set at a scale reading of 15 on a L & N #307 chart when using a 0.1 L & N conductivity probe.

(2) Mounting of the recording instrument should be on the panel board to facilitate its use as an operational aid.

(3) All condensate having a conductivity above the alarm point should be recycled.

One particular inconsistency in the conductivity cell operation was observed. At evaporation rates above 300 gph a high average scale reading was obtained. Possible causes of this will be investigated.

2.24 Evaporator Condensate Demineralizer

Purchase of the Evaporator Condensate Demineralizer has been delayed.

2.3 Program

The program for the coming period is as follows:

(1) Obtaining of the installed cost of a 5' diameter, 5 plate bubble-cap evaporator vapor-scrubber will be made since work at other sites indicates the feasibility of such an installation for de-entrainment. Column vapor velocities between 0.6 and 1.9 ft/sec will be encountered at evaporation rates of 200 to 600 gph.

(2) Purchase and fabrication of an eight-point electronic probe assembly for foam level indication in the Waste Evaporator. Actual installation is scheduled for August 1. It is anticipated that the instrument will permit automatic shutdown if the evaporator foams excessively.

(3) Further investigation of the temporary condensate conductivity cell installation (Section 2.23) followed by the permanent installation of the recorder on the panel board.

ORNL WASTE URANIUM RECOVERY UNIT

Technical - Stewart

3.0 ORNL METAL RECOVERY UNIT

The program is expected to continue at its present tempo and design stage for two months. Revisions upward of the Change Recommendation in both time and money are to be made as a result of the request from the AEC that a breakdown of the CR be made to provide for a lump sum contractor to construct the building. The revisions and subsequent delays are expected to postpone the start of operation of the Recovery Unit six months or until the latter part of March 1951.

Preliminary purchase inquiries have been made for additional process equipment pieces including pumps and process vessels, and tentative specifications for the instrumentation are nearly completed. The design of all process pieces should be completed during the coming period. Structural plans for the building are 85% complete. The electrical details constitute the major part of the remaining work.

PUREX PROCESS

Technical - Jackson, Harrington, Landry, Lewis

4.0 PUREX PROCESS

4.1 Summary

The object of the Purex Development Program at ORNL is to develop the Purex Process through the Pilot Plant phase to obtain a firm basis for comparison with other proposed processes for uranium and plutonium recovery from neutron irradiated natural uranium metal. Laboratory development work at ORNL has demonstrated the feasibility of the process, and has resulted in the establishment of a chemical flowsheet which is adequate for the preparation of a pilot plant equipment flowsheet. A Change Recommendation covering the conversion of the pilot plants used for Redox and 23 Process Development has been submitted to AEC, with modifications to these plants scheduled to be started during June. The cost of equipment changes and additions to the two pilot plants will be approximately $150,000. The first runs using irradiated metal will be made in early fall of this year and the experimental program will continue for eight to twelve months. Final demonstration runs will be made at full Hanford activity level.

4.2 General Description of the Process

The general features of the Purex process are similar to those of the Redox Process. In both solvent extraction processes, uranium and plutonium are first separated from fission products and then from each other by adjusting the valence state of plutonium. In the Purex process, tributyl phosphate is used as the solvent, and nitric acid as the salting and scrubbing agent. This process is promising primarily because the waste volumes are greatly reduced by the evaporation and recovery of the nitric acid.

The contemplated pilot plant operations are described briefly as follows:

First Cycle

Irradiated metal will be dissolved, and plutonium adjusted to the IV valence state. Uranium and plutonium will be extracted from the bulk of the fission products in the IA column, uranium and plutonium separated in the IB column, and uranium stripped in the IC column. Both uranium and plutonium product streams (ICU and IBP) will be passed through stripping columns to remove small amounts of dissolved TBP. The volumes of the plutonium and uranium product streams will be reduced by evaporation.

Second Uranium Cycle

The uranium product stream from the first cycle will be extracted in the ID column and stripped in the IE column. This will complete the decontamination of uranium.

Second Cycle Plutonium

The first cycle plutonium product stream (IBP), after volume reduction, will be transferred to the Bldg. 706-HB pilot plant, formerly used for 23 process development. Plutonium will be extracted in the IIA column and stripped in the IIB column. Plutonium from this cycle will be transferred to the laboratory for additional processing.

Nitric Acid Recovery

Nitric acid from the aqueous waste streams of the first cycle and second uranium cycle will be evaporated and concentrated, reducing the waste volume by a factor of 10 to 100.

4.3 Program

The modifications and additions to the Pilot Plant equipment will begin in June, and the conversion effort will reach maximum intensity shortly after the termination of the Chalk River program, about July 1.

"23" PILOT PLANT DEVELOPMENT

Technical - Jackson, Ferguson, Nicholson

5.0 23 PROCESS DEVELOPMENT

A summary report, ORNL-715, will be issued during the coming period.

6.0 MISCELLANEOUS

J. W. Ullman and R. B. Lindauer, Chemical Engineers, entered the Pilot Plants Section as new hires on May 1 and May 24, 1950 respectively.

Permanentization of the 205 Bldg. is approximately 35% complete. The exterior concrete block work is approximately 70% complete and installation of the penthouse roofing has started. The floors for the Control, Analytical and Pilot Plants Warm Laboratory have been poured and installation of the interior tile should start next period. The roofing, siding, and exterior masonry should be completed during the month of June.

During the past period the following Pilot Plant sample requests have been fulfilled:

Sample	Sample Size	Batch No.	Shipped to
Dissolver Solution (IMd)	4-5 ml	LAMS 995, No. 3	LA
IBP	100 ml	LAMS 995, No. 4	KAPL
IIBP concentrate	2165 ml	Redox #5	HEW