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**MONTHLY TECHNICAL ACTIVITIES REPORT**

**THROUGH SEPTEMBER 15, 1955**

BY

D. L. SCOTT, J. F. EICHELBERGER, AND F. C. MEAD, JR.

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**POLONIUM-210 OPERATIONS**

*This program is concerned with the production of polonium sources and the disposal of radioactive waste. Work is being done to improve the present methods and processes.*

One polonium-lithium, six polonium-beryllium neutron sources and two alpha sources were made and shipped in August. All were for other AEC sites except one alpha source and one polonium-beryllium neutron source.

A sample containing a small percentage of hydrogen is being tested to determine the possibility of analyses for hydrogen by the use of neutron sources. The number of thermal neutrons obtained was not sufficient to give a detectable number of 2.2 mev. gamma rays which result from the capture of a thermal neutron by hydrogen. A graphite block will be used to give an increased flux of thermal neutrons. If necessary, a large sodium iodide crystal will be used to increase the sensitivity for detecting gamma rays.

A new design for alpha sources has been made to permit evacuation without flexing of the windows. In previous alpha sources, the window was not in direct contact with the polonium. Stainless steel has been placed in direct contact with the polonium without polonium diffusing through it. This fact makes it possible to place a window in contact with polonium deposited on a support, thereby eliminating the air space. The window may be held in place by a sleeve or may be sealed in place with a coat of nickel.

Data for the Waste Disposal Operations are shown in Table I.

**TABLE I**

DISCHARGE VOLUME	-	506,000	GALLONS
TOTAL ALPHA ACTIVITY	-	2.1	MILLICURIES
TOTAL BETA ACTIVITY	-	28.6	MILLICURIES
ACTIVITY DENSITY			
ALPHA	-	1	C/MIN/ML
BETA	-	17	C/MIN/ML

## LIVERMORE PROJECT

The Livermore program is concerned with production of lithium deuteride blocks, pressed and canned to meet specifications of a Livermore Laboratory research project.

### INTRODUCTION

All processing work was completed on the Livermore Project by the end of August 1955, and the last shipment was sent to the University of California Radiation Laboratory on September 13, 1955. Approximately 34 kilograms of normal salt was weighed and pressed into blocks, and 81.9 kilograms of blocks were sealed into cans of the three different sizes. The disassembly, crating, and shipment of equipment has been coordinated under the jurisdiction of the Engineering Division. Since this project has been completed, this is the final monthly report.

### OPERATIONS

**DRY-BOX ATMOSPHERE.** The dew point remained well below  $-77^{\circ}\text{F}$  during all the remaining canning operations, and the large capacity of the Lectrodryer units required very few change-overs from unit to unit.

**PRESSING, CURING, SEALING, AND FINAL OPERATIONS.** Thirty-one curing batches of normal salt, each containing approximately 144 pressed 2-inch blocks, were pressed, cured and sealed into aluminum cans. This material was completely processed, gauged, and inspected. It was sent to Livermore as shipment number 6, which consisted of the following:

- 253 size 6-inch standard cans
- 69 size 2-inch dimple cans
- 63 size 4-inch dimple cans
- 65 size 6-inch dimple cans
- 74 size 2-inch standard cans (as samples for analysis).

The following is a summary of all the cans and shipments to Livermore containing type A and type B material:

## TYPE A (ENRICHED SALT) MATERIAL

SHIPMENT NUMBER	*2-INCH	DIMPLES			STANDARD		
		2-in.	4-in.	6-in.	2-in.	4-in.	6-in.
3	25					139	28
4	20		64			72	44
5	<u>60</u>	<u>65</u>	—	<u>66</u>	<u>260</u>	—	<u>181</u>
Total Shipped	105	65	64	66	260	261	253
Required	—	<u>63</u>	<u>63</u>	<u>63</u>	<u>252</u>	<u>252</u>	<u>252</u>
Excessed Shipped	-	2	1	3	8	9	1

\*Sample for Analysis

## TYPE B (NORMAL SALT) MATERIAL

1					252		
2	22					252	
6	<u>74</u>	<u>69</u>	<u>63</u>	<u>65</u>	—	—	<u>253</u>
Total Shipped	96	69	63	65	252	252	253
Required	—	<u>63</u>	<u>63</u>	<u>63</u>	<u>252</u>	<u>252</u>	<u>252</u>
Excessed Shipped	-	6	0	2	0	0	1

RECORD OF INVENTORY. The following shows the amounts of materials received, processed, and shipped; the remainder of salt to be shipped in drums to Livermore; and the processing losses:

## TYPE A MATERIAL

Received	138,015 grams
Shipped to ORNL for Analysis	45 grams
Shipped in Sealed Cans	104,309 grams
Remaining Salt in Drums (uncanned) to be Shipped to Livermore	<u>33,390</u> grams
Total Processed and Unprocessed	<u>137,744</u> grams
Processing Loss (Based upon 115,960 grams handled)	271 grams or 0.23 per cent

## TYPE B MATERIAL

Received		149,732 grams
Shipped to Livermore, Raw Salt	6,213 grams	
Shipped in Sealed Cans	113,525 grams	
Remaining Salt in Drums (uncanned) to be Shipped to Livermore	<u>29,677</u> grams	
Total Processed and Unprocessed		<u>149,415</u> grams
Processing Loss (Based upon 133,436 grams handled)		317 grams 0.24 per cent

The last phase of the operations was completed very rapidly and within the estimated time. The dismantling of the equipment was begun immediately under the jurisdiction of the Engineering Division. All equipment earmarked for shipment will be crated and marked adequately for both content and identification. All operating personnel were disbanded from the project on or about September 12, 1955.

## CONCLUSIONS

1. For maintaining dry-box atmospheres at very low moisture levels, the Lectrodryer unit was found to be very successful and easy to use.
2. Little or no correlation of moisture content with respect to hydroxide analysis (by the method of Frazer of UCRL) could be found. If any swelling of the sealed cans were to occur in a relatively short time, the swelling would be detected within 48 hours after sealing, and all such cans would be immediately rejected.
3. Complying with chemical purity specifications was very difficult because Mound Laboratory had virtually no control over the purity of the raw material.
4. From knowledge gained from this work, it is probable that pressed compounds of hygroscopic materials can be sealed into containers and stored for long periods of time.

## THORIUM PROJECT

The thorium project is directed toward development and operation of a process suitable for extraction of thorium from Brazilian monazite sludge and AEC waste materials. The process must produce a thorium salt suitable for the preparation of metallic thorium of high purity.

### INTRODUCTION

Since the thorium project was cancelled, work has continued on the development of a flow sheet for the extraction of thorium from Brazilian hydroxide sludge. The pilot plant solvent extraction work has continued, but all other work on this project has stopped.

### SOLVENT EXTRACTION

After studying all of the previously collected data from pilot plant runs, the conditions for run number 14 were established. All five columns were operating in this run. Run number 14 was the first successful operation with the fifth column. The column conditions are shown at the end of this section in the form of a flow sheet (Figure 1).

Table I is the data that were collected after all five columns were "on stream" about ten hours. The first two columns had been "on stream" about 14 hours at this time.

No rare earth analyses are available as the spectrograph is being repaired. Thorium loss at this time was only 0.3 per cent in the raffinate and 0.3 per cent from the top of column four in the organic phase.

All of the available slurry feed has been sent through the columns. It is believed that slurry feed has been behaving so well that it is needless to experiment with clarified feed in the columns.

This report concludes the work on the thorium pilot plant, and it is believed that flow sheet number 14 gives the optimum conditions for the operation of a thorium refinery on slurry feed prepared from Brazilian hydroxide sludge.




TABLE I  
ANALYTICAL RESULTS RUN NUMBER 14

	THORIUM (gm/l)	URANIUM (gm/l)	NITRIC ACID (N)	RATIO (Th/U)
Organic from 1st Column	47.16	0.84	0.754	56.2
Organic from 2nd Column	43.74	0.87	0.401	50.3
Organic from 3rd Column	26.77	0.32	0.228	83.6
*Organic from 4th Column	0.09	0.49	0.040	0.18
Organic from 5th Column	0.50	0.04	0.010	12.5
Raffinate from 1st Column	0.42	0.17	0.107	-
TNT Product from 3rd Column	37.61	0.0025**	0.560	15,050.0
U Product from 5th Column	0.35	3.57	Basic	-
Reflux 2nd to 1st Column	8.12	0.31	3.089	-
Reflux 4th to 3rd Column	44.71	0.15	0.557	-

\*This sample was taken one hour earlier

\*\*By Fluorimeter



## FLOWSHEET RUN 14

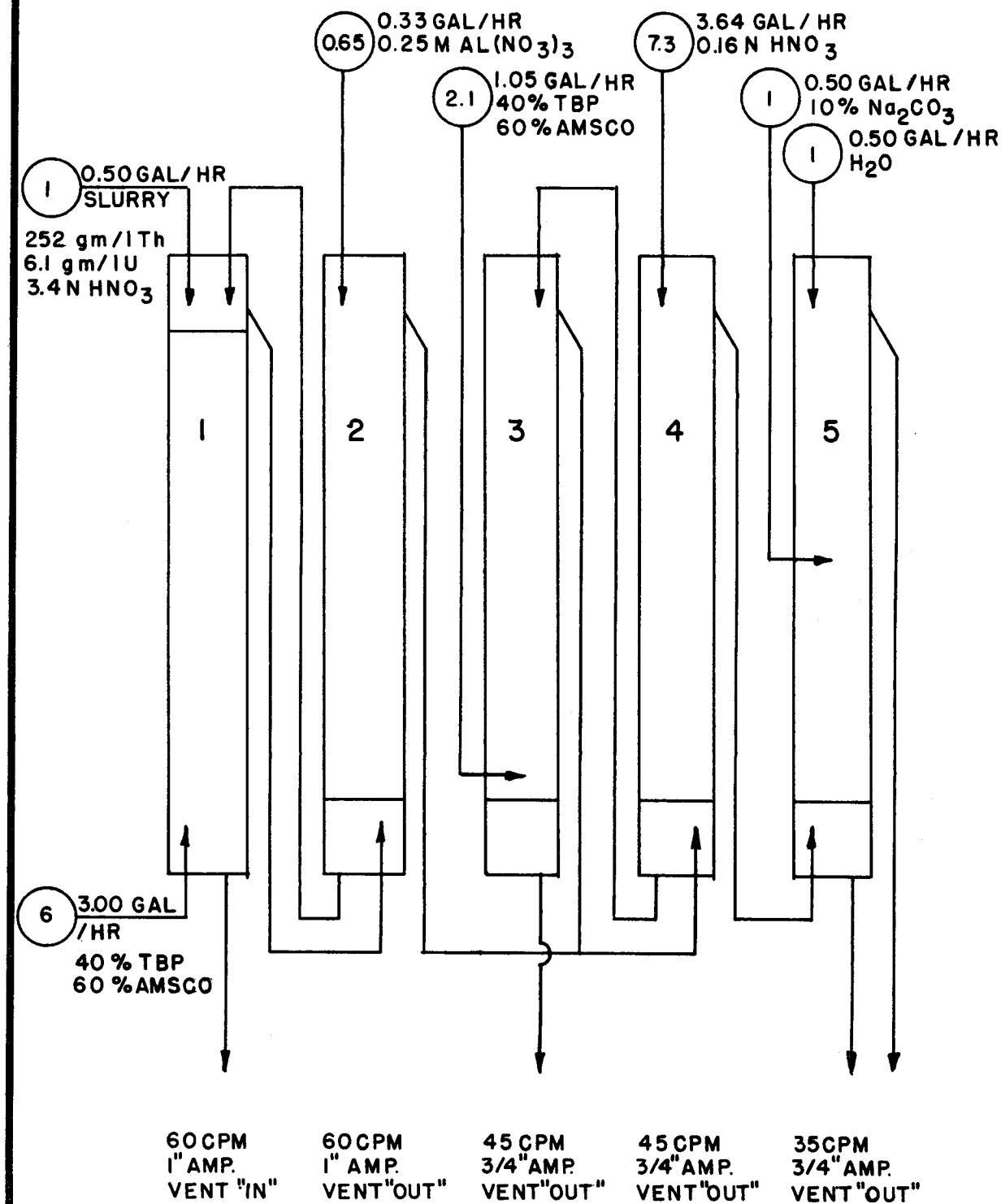


FIGURE 1

5898

**IONIUM PROJECT**

The ionium project is directed towards the development and operation of a process for the extraction of ionium from partially processed raffinates from the Mallinckrodt uranium refinery. The process must produce ionium suitable for use as a tracer for use by Los Alamos.

The present laboratory method for the isolation of ionium (thorium-230) from the partially processed raffinates is a laborious process involving extraction with diethyl ether and pentaether using separatory funnels.

Process development work has been started to establish a flow sheet for the extraction of ionium using continuous flow equipment. Discussions with personnel at Argonne National Laboratory and Mallinckrodt have indicated that a flow sheet using a tributyl phosphate solvent extraction appears promising as a solution to this problem.

A study with batch countercurrent extraction equipment, using thorium-232 spiked with small amounts of uranium and rare earth elements to represent the material to be received from Mallinckrodt, has been started. Since ionium is an alpha emitter and a thorium isotope, it is thought that it would be better to do as much work as possible on simulated cold solutions, due to the contamination problem using hot solutions, and also due to the fact that no hot material will be available from Mallinckrodt until at least February 1956. Tributyl phosphate with Amsco is being used as the organic phase.

A study of liquid-liquid contacting equipment in which to carry out the continuous extraction has indicated that mixer-settlers would possibly be the type of equipment to use. A miniature mixer-settler is to be obtained to carry out the initial work on cold solutions.

A request has been sent to Los Alamos for the specifications of the product ionium to be sent to them. It is possible that if the specifications are very low as to the amount of impurities allowable in the product, analytical methods will have to be developed for the detection of those impurities in the range desired.

## REACTOR PROGRAM

## FUSED SALTS RESEARCH PROJECT

The Aircraft Nuclear Propulsion Project is considering the use of a fused-salt fuel system. Mound Laboratory has been assigned the problem of determining the phase relationships and physical properties of the components of some of the proposed fuel systems.

## PHASE STUDIES

The compositions investigated by differential thermal analysis and petrography were made up by accurately weighing and mixing the purified starting components, NaF, BeF<sub>2</sub>, and UF<sub>4</sub>. The samples examined petrographically were held at 800°C for 2-1/2 hours and then cooled slowly. The data are interpreted in conjunction with Table I and Figure 1. In Figure 1, E stands for eutectic and P for peritectic.

TABLE I

RESULTS OF PETROGRAPHIC AND DIFFERENTIAL-THERMAL-ANALYSIS  
STUDIES IN THE NaF-BeF<sub>2</sub>-UF<sub>4</sub> SYSTEM

COMPOSITION NUMBER	COMPOSITION MOLE PER CENT			LIQUIDUS TEMPERATURE (°C)		PHASES
	UF <sub>4</sub>	BeF <sub>2</sub>	NaF			
1	30.0	35.0	35.0	*	*	
2	22.0	26.0	52.0	*		NaBeF <sub>3</sub> and Na <sub>7</sub> U <sub>6</sub> F <sub>31</sub>
3	36.3	21.3	42.4	655		NaBeF <sub>3</sub> , Na <sub>7</sub> U <sub>6</sub> F <sub>31</sub> and unidentified crystal
4	31.5	15.7	52.8	670		NaBeF <sub>3</sub> and Na <sub>7</sub> U <sub>6</sub> F <sub>31</sub>
5	10.0	30.0	60.0	525		NaBeF <sub>3</sub> , Na <sub>2</sub> BeF <sub>4</sub> and Na <sub>7</sub> U <sub>6</sub> F <sub>31</sub>
6	17.1	31.4	51.5	*		NaBeF <sub>3</sub> and Na <sub>7</sub> U <sub>6</sub> F <sub>31</sub>
7	20.0	40.0	40.0	600		NaBeF <sub>3</sub> , Na <sub>7</sub> U <sub>6</sub> F <sub>31</sub> and an unidentified crystal
8	22.2	11.1	66.7	635		Na <sub>2</sub> UF <sub>6</sub> , Na <sub>2</sub> BeF <sub>4</sub> and NaF
9	27.1	9.2	63.7	630	*	
10	27.3	13.6	59.1	*	*	
11	10.0	20.0	70.0	*		Na <sub>2</sub> UF <sub>6</sub> , Na <sub>2</sub> BeF <sub>4</sub> and NaF

\* Data insufficient

~~SECRET~~



A quasi-binary was definitely established on the join between  $\text{Na}_2\text{BeF}_4$  and  $\text{Na}_3\text{UF}_7$  (see composition 11). Another quasi-binary was definitely established on the join between  $\text{NaBeF}_3$  and  $\text{Na}_7\text{U}_6\text{F}_{31}$  (see compositions 2, 4, and 6). The investigations of compositions 9 and 10 are not yet complete. However, all other compositions investigated indicate the absence of any additional quasi-binaries.

Examinations of compositions 1, 3, and 7 indicated that the samples were not completely homogeneous. Additional samples were held at  $1000^\circ\text{C}$  for thirty minutes and then slowly cooled. An unidentified crystalline phase in composition 7 is green, pleochroic, biaxial negative,  $\alpha \approx 1.521$ , and  $\beta \approx 1.565$ , and  $\gamma \approx 1.581$ . This suggested the possibility of a ternary compound. However, the unidentified crystalline phase in composition 3 differed in that its refractive indices were intermediate between the indices above and those of  $\text{UF}_4$ . The crystals observed in both cases may have been  $\text{UF}_4$  in which the refractive indices were modified by solid-solution formation. X-ray diffraction data on these samples are inconclusive. At present, studies are being made to determine whether or not a ternary compound exists.

#### PHYSICAL PROPERTIES

The viscosities of six ternary mixtures of  $\text{NaF}$ ,  $\text{BeF}_2$  and  $\text{UF}_4$  and two binary mixtures of  $\text{NaF}$  and  $\text{BeF}_2$  were measured over the temperature range  $500^\circ$  to  $900^\circ\text{C}$  in a series of ten runs. All of the ternary mixtures contained 8 mole per cent uranium fluoride. The viscosities and densities at  $600^\circ\text{C}$  and  $800^\circ\text{C}$  are given in Table II. Plots of the logarithm of the viscosity as a function of the reciprocal of the absolute temperature gave linear curves similar to those obtained from other mixtures of sodium, beryllium and uranium fluorides.

The densities of twelve ternary mixtures, in addition to those given in Table II, were measured this month. The compositions varied from 76 mole per cent  $\text{NaF}$ , 18 mole per cent  $\text{BeF}_2$ , 6 mole per cent  $\text{UF}_4$  to 46 mole per cent  $\text{NaF}$ , 50 mole per cent  $\text{BeF}_2$ , 4 mole per cent  $\text{UF}_4$ . Densities of each mixture were measured between their liquidus point and approximately  $900^\circ\text{C}$  at  $50^\circ\text{C}$  intervals. The density values obtained at  $600^\circ\text{C}$  and  $800^\circ\text{C}$  are given in Table III. Reruns were made on the 52  $\text{NaF}$ , 42  $\text{BeF}_2$ , 6  $\text{UF}_4$ ; 46  $\text{NaF}$ , 48  $\text{BeF}_2$ , 6  $\text{UF}_4$ ; and the 76  $\text{NaF}$ , 20  $\text{BeF}_2$ , 4  $\text{UF}_4$  runs because the data on these runs did not seem to be consistent with the data of the other members of the same series. Although the data on these reruns was not significantly different than the original runs, these mixes are being investigated further.

TABLE II

## VISCOSITY AND DENSITY OF FUSED SALTS

Composition (Mole Per cent)	600°C			800°C		
	$\eta$ (Centipoise)	$\rho$ (gm/cm <sup>3</sup> )	$\eta/\rho$	$\eta$ (Centipoise)	$\rho$ (gm/cm <sup>3</sup> )	$\eta/\rho$
76 NaF 16 BeF <sub>2</sub> 8 UF <sub>4</sub>	10.0	2.918	3.43	4.5	2.738	1.64
70 NaF 22 BeF <sub>2</sub> 8 UF <sub>4</sub>	9.2	2.850	3.23	2.78	2.720	1.02
64 NaF 28 BeF <sub>2</sub> 8 UF <sub>4</sub>	8.6	2.820	3.05	2.99	2.687	1.11
58 NaF 34 BeF <sub>2</sub> 8 UF <sub>4</sub>	10.1	2.779	3.63	2.85	2.651	1.08
52 NaF 40 BeF <sub>2</sub> 8 UF <sub>4</sub>	11.7	2.769	4.23	3.20	2.645	1.21
46 NaF 46 BeF <sub>2</sub> 8 UF <sub>4</sub>	14.0	2.752	5.09	4.00	2.636	1.51
21.9 NaF 78.1 BeF <sub>2</sub>	~900	2.05*	~450	58.0	1.956	29.7
42.7 NaF 57.3 BeF <sub>2</sub>	--	--	--	6.4	1.982	3.23

\* Estimated

TABLE III  
DENSITY OF FUSED SALTS

Composition (Mole) (Per cent)		Density at 600°C (gm/cm <sup>3</sup> )	Density at 800°C (gm/cm <sup>3</sup> )
76 NaF 18 BeF <sub>2</sub> 6 UF <sub>4</sub>		2.663	2.543
70 NaF 24 BeF <sub>2</sub> 6 UF <sub>4</sub>		2.633	2.502
64 NaF 30 BeF <sub>2</sub> 6 UF <sub>4</sub>		2.600	2.475
58 NaF 36 BeF <sub>2</sub> 6 UF <sub>4</sub>		2.560	2.442
52 NaF 42 BeF <sub>2</sub> 6 UF <sub>4</sub>	1st run 2nd run 3rd run	2.603 2.600 2.658	2.481 2.471 2.535
46 NaF 48 BeF <sub>2</sub> 6 UF <sub>4</sub>	1st run 2nd run	2.560 2.586	2.459 2.466
76 NaF 20 BeF <sub>2</sub> 4 UF <sub>4</sub>	1st run 2nd run 3rd run	2.430 2.438 2.433	2.339 2.326 2.326
70 NaF 26 BeF <sub>2</sub> 4 UF <sub>4</sub>		2.476	2.345
64 NaF 32 BeF <sub>2</sub> 4 UF <sub>4</sub>		2.468	2.349
58 NaF 38 BeF <sub>2</sub> 4 UF <sub>4</sub>		2.422	2.308
52 NaF 44 BeF <sub>2</sub> 4 UF <sub>4</sub>		2.398	2.280
46 NaF 50 BeF <sub>2</sub> 4 UF <sub>4</sub>		2.381	2.276

## ANALYTICAL METHODS

Work is underway to improve the precision and accuracy of the X-ray fluorescence method. Among the work done are studies on the general scattering curves of solutions, the installation of improved scaling circuits, and the addition of proportional and scintillation counters.

Scattering curves were constructed from point-by-point counts made with the X-ray spectrometer, using distilled water and Lucite samples. A marked similarity was noted in the shapes of the curves from these two materials, indicating that the curves are primarily a function of the exciting radiation and of the characteristics of the detectors, and are only secondarily a function of the sample material. Such data will be useful in applying more exact background corrections, one of the uncertainties in high precision work.

Most of the time was devoted in assembling and reconditioning the electronic apparatus. The General Electric 2SPG preamplifier has been modified to feed into an Atomic Instrument, 204-B linear amplifier, which in turn feeds an Atomic Instrument, 1060-A decade scaler with a one microsecond input.

Preliminary results using a General Electric Krypton proportional counter are encouraging, indicating that pulse-height analysis may effect an even greater improvement in reducing background and interference from undesired elements. A scintillation counter tube (DuMont #6291 tube with thallium activated NaI crystal) has been installed and is being tested.

Some thought is being given toward a restudy of the effects of the presence of sodium ion upon the plating of uranium and upon the precipitation of beryllium. Previous work reported in July, showed that the presence of sodium ion caused a 1.4 per cent deficiency in the plating of uranium.

## FILTRATION STUDIES

Last month's report gave a tentative chart of the "valley" from the ternary eutectic [located near 12 mole per cent  $\text{UF}_4$ , 16 mole per cent  $\text{BeF}_2$ , 72 mole per cent NaF] toward the NaF- $\text{BeF}_2$  side of the phase diagram. The location of the "valley" was further confirmed when a sample with the composition 2 mole per cent  $\text{UF}_4$ , 30 mole per cent  $\text{BeF}_2$ , 68 mole per cent NaF was filtered at  $550^\circ\text{C}$ . At this temperature, the filtrate had the composition 4.1 mole per cent  $\text{UF}_4$ , 26.7 mole per cent  $\text{BeF}_2$ , 67.2 mole per cent NaF, which places it exactly in the previously described "valley" in regards to both composition and temperature.

Several other compositions were filtered at various temperatures, but their filtrates did not follow the valley described above. These experiments are given in Table IV.



TABLE IV  
LIQUIDUS COMPOSITIONS BY FILTRATION

Starting Compositions Mole Per Cent			Filtration Temperature (°C)	Filtrate Compositions Mole Per Cent		
UF <sub>4</sub>	BeF <sub>2</sub>	NaF		UF <sub>4</sub>	BeF <sub>2</sub>	NaF
2	32	66	556	4.2	31.8	64.0
2	32	66	575	2.6	33.6	63.8
5	28	67	534	8.1	25.7	66.2

Sufficient data have not been collected to show whether these points form a new "valley". The importance, thus far, is it tends to prove that the compounds Na<sub>3</sub>UF<sub>7</sub> and Na<sub>2</sub>BeF<sub>4</sub> form a quasi-binary. All of the starting compositions which lie within the triangle formed by the compounds NaF-Na<sub>3</sub>UF<sub>7</sub> - NaBeF<sub>4</sub> have had filtrates which follow along the first described "valley". The latter compositions which do not lie within this triangle appear to follow a different liquidus route.

## PROTACTINIUM SEPARATION PROJECT

A program has been undertaken to isolate and purify a gram of protactinium-231. This material is important since it will provide a relatively stable isotope to study the physical and chemical properties of the 27-day protactinium-233 which will be created in the Th-232 → Pa-233 → U-233 sequence in thorium-breeder blankets.

## PROCESSING

The first run in the protactinium separation plant has been completed. Drum number 20, containing 288 pounds of raffidue, was used as the charge. The previously described HCl process was employed with modifications suggested by recent development work.

The raffidue was dissolved in 2 N muriatic acid, using 10 milliequivalents of HCl per gram of cake. Solution was complete in two hours. In the carrier precipitation step, approximately 1.88 grams of salt per gram of dissolved raffidue was added to the solution. The liquid was boiled for two hours. After separation of the carrier, the latter was washed, first with 0.5 N muriatic acid, then with 12 per cent sodium hydroxide, and finally with water. All phase separations were accomplished by overnight settling.

The distribution of protactinium between the carrier product and the various waste fractions is shown in Table I. The results, based on the 27 kev peak counting rate after suitable radiochemical separations, indicate a recovery of about 82 per cent of the protactinium. The principal loss was in the solution step. This might be reduced by reprocessing, by better phase separation, or by both.

TABLE I

## ACTIVITY INVENTORY FOR PLANT RUN WITH DRUM NUMBER 20

Source	27 kev Activity (cpm)	% of Total
Solids from HCl Digestion Residue	6,215,000	5.98
Liquid from HCl Digestion Residue	7,710,000	7.40
Carrier Supernate	2,075,000	2.00
HCl Carrier Wash	2,660,000	2.56
NaOH Carrier Wash	none	none
H <sub>2</sub> O Carrier Wash	none	none
Final Washed Carrier	<u>85,360,000</u>	<u>82.06</u>
TOTAL	104,020,000	100.00

During the run, two pumps failed to operate satisfactorily. One, a diaphragm slurry pump, needed an external source of vacuum on the intake side in order to operate. A vacuum pump was secured and installed. The slurry pump then operated satisfactorily with water feed.

The other pump was a large Haveg pump for pulling waste supernates from three vessels to the neutralization tanks. This pump could not be primed without disturbing the settled slurry in the kettles. The piping on the suction side was simplified, and several leaks were corrected. An aspirator connection was installed at the discharge side of the pump to assist in priming.

The venting system for the kettles proved inadequate to handle the vapor from a boiling solution in Pfaudler kettle number 2. A spray chamber was installed over the vent of this Pfaudler kettle. A 10 per cent sodium hydroxide solution was used in the spray. All condensate plus the caustic solution was returned to a reservoir for recycling. This spray chamber operated satisfactorily during a plant run.

The services of a chemical operator were secured on September 12, and a second plant run, utilizing drum number 19 as charge, was started on September 13. The solution and carrier precipitation steps have been completed.

An attempt was made further to process the product slurry from the first plant run. Five-hundred milliliters of slurry were dissolved in hydrochloric acid and the normality was adjusted to about 9 N. This solution was started through a 9-centimeter diameter anion exchange column, using Dowex-1 resin, but the sorption step was unsuccessful because the resin bed was disturbed and "floated" by the solution.

#### EXPERIMENTAL DEVELOPMENT

Small-scale ion-exchange-column experiments are in progress to determine suitable operating conditions for final purification of the protactinium. Two one-centimeter-diameter columns were filled to a depth of two centimeters ( $D = D_1$ ) and two others to a depth of four centimeters ( $D = D_2$ ), all with Dowex-1 anion exchange resin. This gave a column volume of 1.56 milliliter for each of the first two columns and 3.12 milliliter for the last two columns.

To obtain a suitable column feed, the slurry product of the plant run with drum number 20 was stirred with hydrochloric acid and the insoluble residue was filtered off. The filtrate, with concentration denoted by  $F = F_2$ , was approximately 9 N in HCl and gave a gamma count of 3322 counts per minute per milliliter at the 27 kev peak. Sixteen milliliters of this

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solution were employed as feed in each of the last two columns. This feed solution was diluted with an equal volume of 9 N HCl to produce a second feed ( $F = F_1$ ), having one-half the concentration of the original feed. Thirty-two milliliters of this solution were employed as feed in each of the first two columns.

Flow rates throughout the experiments were maintained at approximately 0.4 milliliter per minute per square centimeter. After sorption, each column was washed with five column volumes of 9 N HCl. The eluent was 9 N in HCl and 0.05 N in HF, and fifteen column volumes were used. After elution, each column was stripped with ten column volumes of 1 N HCl.

The distribution of the 27 kev activity between the various effluents is shown in Table II. The 27 kev counting rate appears to be approximately additive with respect to the various fractions. A significant protactinium break-through evidently occurred in the sorption step when the more concentrated feed was employed with the two-centimeter column. The 27 kev counts in the strip effluents evidently represent primarily activity other than protactinium, since no 27 kev peaks could be detected. When the four-centimeter columns were used, the amount of eluent required to remove the protactinium appeared to be independent of the feed concentration.

The gamma spectra of the feed and of the elution effluents suggested a rather high degree of radioactive purity. The elution effluents are being analyzed by X-ray fluorescence to detect the presence of nonradioactive impurities.

In the meantime, experiments are being run with six-centimeter bed heights, since the appearance of the two-centimeter and four-centimeter beds, after elution, indicated that cobalt or nickel break-throughs may have occurred.



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TABLE II

DISTRIBUTION OF 27 KEV GAMMA ACTIVITY  
IN ION EXCHANGE COLUMN EFFLUENTS

(PER CENT OF TOTAL FEED)

<u>COLUMN 1</u> <u>(D<sub>1</sub>F<sub>1</sub>)</u>	<u>COLUMN 2</u> <u>(D<sub>2</sub>F<sub>1</sub>)</u>	<u>COLUMN 3</u> <u>(D<sub>1</sub>F<sub>2</sub>)</u>	<u>COLUMN 4</u> <u>(D<sub>2</sub>F<sub>2</sub>)</u>
<u>FEED EFFLUENT</u>			
1.8	0.97	2.80	0.60
<u>WASH EFFLUENT</u>			
0.25	0.18	2.8	0.34
<u>ELUTION EFFLUENT</u>			
(Accumulative percentages for successive 4 ml fractions)			
0.57	0.03	1.0	0.03
36.4	0.05	40.0	0.05
73.2	12.0	75.3	8.0
90.9	40.4	85.1	47.4
96.8	70.0	90.7	76.7
100.1	86.6	94.1	88.8
	93.4		95.0
	96.7		98.0
	98.5		99.5
	99.4		100.4
	100.2		101.2
	100.8		101.5
<u>STRIP EFFLUENT*</u>			
5.4	2.5	4.2	2.4

\*These values probably not associated with protactinium.

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