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Central File No. 56-5-45

MONTHLY TECHNICAL ACTIVITIES REPORT
THROUGH MAY 15, 1956

BY

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PRODUCTION PROGRAM

These projects are concerned with the production of alpha and neutron sources, and with the disposal of radioactive wastes. Work is being done to improve present methods and processes.

A thorough investigation is in progress to find a pliable glove which will resist deterioration when in contact with acids containing radioactivity. Many different manufacturer's types of rubber and neoprene gloves were tested and were found to be unsuitable. Up to the present time, the best results obtained were from gloves made from Unichrome which were fabricated in the T-Building. Further work is in progress to increase the thickness of this type of glove to reduce the migration of radioactivity through it.

Thirteen polonium-beryllium neutron sources and three alpha sources were shipped in April, 1956. Seven neutron sources and two of the alpha sources were for other AEC sites

Total Alpha activity discharged to the river remained about the same as for the previous report period. Total Beta activity discharged to the river was about 75 per cent greater than the previous report period Data for the Waste Disposal Operations are given in Table I.

TABLE I

DISCHARGE VOLUME	516,000	GALLONS
TOTAL ALPHA ACTIVITY	5.4	MILLICURIES
TOTAL BETA ACTIVITY	25.3	MILLICURIES
ACTIVITY DENSITY		
ALPHA	3	CT/MIN/ML
BETA	14	CT/MIN/ML

DEVELOPMENT PROGRAM

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 by Los Alamos as a tracer.

Three "hot" runs have been processed through the ionium pilot plant since April 15, 1956. The first two runs used feed shipped from the Mallinckrodt Chemical Works, while the third was a reprocessing run on the aqueous raffinates from the first two runs plus the sulfate solution that was on hand. Variations were made in the flowsheet to obtain better rare earth and common element decontamination; these experiments resulted in better products, but led to very high ionium losses in the raffinate streams.

A comparison of feed and product for these runs is given in Table I.

TABLE I

FEED VERSUS PRODUCT

BATCH NO.	TOTAL THORIUM *(gm/l)*	IONIUM *(gm/l)*	URANIUM *(gm/l)*	NITRIC ACID *(N)*	VOLUME *(liters)*	TOTAL IONIUM *(grams)*
3 FEED	7.28	0.657	0.38	2.158	64.27	42.23
3 PRODUCT	67.09	6.41	0.034	0.793	1.388	8.89
4 FEED	8.50	0.762	0.99	1.964	196.86	150.0
4 PRODUCT	112.22	11.68	0.022	0.191	5.715	66.75
5 FEED	16.85	1.51[a]	0.62	1.967	60.0	90.6[a]
5 PRODUCT	-	-	-	-	-	-

a. THIS SAMPLE WAS NOT ALPHA COUNTED AND THE VALUE IS BASED ON THE AVERAGE OF THE Th-232 + Th-230/Th-230 RATIOS FOR FEEDS NUMBER 1, 3 AND 4. THE RATIO FROM FEED NUMBER 2 WAS OMITTED BECAUSE IT WAS ABNORMALLY HIGH IN Th-232 CONTENT. THIS COULD HAVE BEEN CAUSED BY GETTING SOME CARNOTITE MIXED IN WITH THE AIR-PORT CAKE.

Two shipments of final product have been made from Mound Laboratory since April 6, 1956. The first was on May 1 and consisted of the four gram unit which was short on the April 6 shipment. The second was on May 10 and consisted of five units of two grams each and five units of ten grams each. These units were part of the 200 gram shipment for May 18.

The analytical data on the major components of these two shipments are listed in Table I under batch numbers 3 and 4. Batch numbers 3 and 4 had a weight composition of 5.607×10^{-3} and 9.38×10^{-3} grams of ionium per gram of solution respectively. The shipping data are listed in Table II.

TABLE II

IONIUM SHIPMENTS

BOTTLE CODE	IONIUM CONTENT *(grams)*	SHIPPING DATE
T-3-15-4	4.01	MAY 1
T-4-16-2	1.99	MAY 10
T-4-17-2	2.00	MAY 10
T-4-18-2	2.00	MAY 10
T-4-19-2	1.99	MAY 10
T-4-20-2	2.00	MAY 10
T-4-21-10	10.02	MAY 10
T-4-22-10	10.02	MAY 10
T-4-23-10	10.00	MAY 10
T-4-24-10	10.03	MAY 10
T-4-25-10	10.00	MAY 10
TOTAL	64.06	

A "material balance" of the ionium throughout the process on these three runs is given in Table III.

TABLE III

IONIUM "MATERIAL BALANCE"

DESCRIPTION	IN	OUT
BATCH 3 FEED	42.23	-
BATCH 4 FEED	150.0	-
SULFATE FEED	3.4	-
BATCH 3 PRODUCT	-	8.89
BATCH 4 PRODUCT	-	66.75
BATCH 5 FEED	-	90.6
BATCH 3 ORG. RAFF.	-	0.35
BATCH 4 ORG. RAFF.	-	10.46
EXTRA HOLD-UP IN UNITS + SAMPLES	-	17.58
TOTAL	195.63	195.63

As in the "material balance" previously reported, no analytical data were available on extra hold-up.

The spectrographic analysis of product number 3 appears in Table IV. Products 4 and 5 will be analyzed together to expedite the chemical processing.

TABLE IV

SPECTROGRAPHIC ANALYSIS FOR PRODUCT 3

ELEMENT	PPM (BASED ON IONIUM)	ACCURACY
La	37	±50%
Rh	<60	-50%
Dy	77	±50%
Ce	120	±50%
Gd	22	±50%
Y	3	±50%
Sm	44	±50%
Pr	8	±50%
As	24	UNKNOWN
Ta	<25	±50%
W	70	+100%-50%
Sc	<250	-50%
Eu	<15	-50%

A method has been developed for analyzing thallium and arsenic by X-ray fluorescence using selenium as an internal standard. Both of these elements are too volatile for satisfactory spectrographic analysis. For product number 3, the values of arsenic and thallium were 200 ppm ± 50% and 20 ppm + 100% - 50% respectively, based on ionium.

The 0.7 per cent of the alpha energies that were attributed to Th-227 and its daughters in products 1 and 2 has not decayed as anticipated. This indicates the presence of the parent, Ac-227.

Work has continued on the development of techniques for the preparation of slides for alpha pulse-height analysis. Slides were prepared by plating from a thorium nitrate solution using potassium ferrocyanide as an electrolyte. This resulted in two slides for pulse-height analysis. A third slide, to complete the assay, was prepared by pipetting the solution remaining from the electrolysis onto a fresh slide. The results of the pulse-height analysis for products 3 and 4 are given in Table V.

The supernatants from the head-end concentrations contained a total of 800 mg of ionium. Aluminum nitrate was added in an attempt to carry the ionium down with the aluminum hydroxide flock. Fifty per cent of the ionium was carried down by this method. These supernatants are still held in the high risk corridor and preliminary waste disposal treatment will be carried out on these streams after the May 29 shipment. One organic raffinate that was high in ionium has been partially stripped. The aqueous stream from the strip will be concentrated for reprocessing. It is believed that all of the aqueous raffinate streams may be treated in the ionium processing hoods to bring the ionium down to a level that will permit them to be sent to the waste disposal building. The organic raffinates will be stripped, and then possibly absorbed in vermiculite or other material, and shipped to Oak Ridge for burial.

TABLE V

PULSE HEIGHT ANALYSIS

PRODUCT NUMBER	ALPHA ENERGIES BELOW 4.53 MEV	ALPHAS DUE TO Ac-227 AND DAUGHTERS	ALPHAS DUE TO Th 228
3	3.4%	0.5%	<0.02%
4	2.1%	0.6%	<0.02%

RESEARCH 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.

PETROGRAPHIC STUDIES

Binary compositions containing ten to thirty mole per cent of UF_4 (on the LiF-UF_4 binary system) were studied to verify the binary diagram, and to gain further information on the unidentified green crystalline phase previously reported. The unidentified phase was found in all of the compositions studied. It was found to occur as a quench product in samples quenched from above the liquidus temperature. The unidentified green phase was also formed in samples that were soaked for extended periods in the temperature range between the solidus and the liquidus. On slow-cooling to room temperature, the green phase was unchanged. However, on holding the samples just below the solidus temperature for an extended period, the unidentified phase disappeared in all cases. An attempt is now in progress to produce a pure sample of the phase for identification by leaching out the associated LiF.

A sample of ternary composition, 8 mole per cent UF_4, 23 mole per cent BeF_2, 69 mole per cent LiF, was studied by thermal gradient quenching over the temperature range 395° to 460°C. The liquidus temperature was found to be approximately 430°C, and the solidus temperature was found to be approximately 425°C. This composition is apparently very close to a ternary eutectic. The phases identified were Li_2BeF_4, $Li_7U_6F_{31}$, and a third phase of questionable identity which appeared to be LiF. Additional compositions in this same region are being prepared for examination.

FILTRATION STUDIES

Additional data for the system LiF - BeF_2 - UF_4 is given in Table I. Samples 1 and 2 tend to confirm an earlier suggestion that the UF_4 phase area extends very close to the LiF - BeF_2 edge of this ternary.

TABLE I

LIQUIDUS COMPOSITIONS BY FILTRATION

UF_4 - BeF_2 - LiF TERNARY

SAMPLE NUMBER	STARTING COMPOSITION MOLE PER CENT			FILTRATION TEMPERATURE °C	FILTRATE COMPOSITION MOLE PER CENT		
	UF_4	BeF_2	LiF		UF_4	BeF_2	LiF
1	20	40	40	481	3.1	49.0	47.9
2	20	40	40	528	4.9	48.0	47.1
3	20	28.5	51.5	400	1.6	46.8	51.6
4	20	28.5	51.5	400	1.7	45.9	52.4
5	15	25	60	439	5.8	30.5	63.7
6	15	25	60	440	6.3	30.6	63.1
7	15	25	60	458	7.0	29.5	63.5

Testing work has continued with the high temperature centrifuge mentioned in the April report. The chief difficulty has been that some liquid goes around the filter instead of through it, so that some solid may be included with the filtrate Apparently the amount of solid in the filtrate has been small, since Samples 3 and 4, representing two separate experiments, have nearly identical filtrate compositions. It seems doubtful that the amount of solid in the filtrates would be identical in each experiment. Further support for this reasoning is given by samples 5 and 6 which are also two separate experiments that gave nearly identical filtrate compositions.

DIFFERENTIAL THERMAL ANALYSIS

Investigation by differential thermal analysis of the binary equilibrium phase diagram LiF - BeF_2 will be completed soon. Data in Table II show the melting point of LiF to be 847°C as against 870°C reported by Roy, Roy, and Osborne. Various handbooks give the melting point at 870°C. However, the value found at this laboratory agrees with the value determined by Oak Ridge National Laboratory. A plot of the liquidus values from 72 mole per cent LiF - 28 mole per cent BeF_2 to 100 mole per cent LiF falls on a smooth curve, but some uncertainty in the liquidus curve exists for the area between 65 mole per cent LiF - 35 mole per cent BeF_2 and 72 mole per cent LiF - 28 mole per cent BeF_2. In this latter area, liquidus values taken from cooling curves are nearly 50°C above the values observed by Roy, Roy, and Osborne. Some additional work in this portion of the diagram with the help of X-ray diffraction will complete this investigation.

TABLE II

DIFFERENTIAL THERMAL ANALYSIS

LiF - BeF_2 BINARY

DTA NO.	COMPOSITION MOLE PER CENT LiF	 BeF_2	SOLID TRANSITIONS °C	EUTECTIC °C	LIQUIDUS °C	LIQUIDUS[a] °C
11L	100	0000	-	-	847	870
12L	30	70	218	353	495	488
13L	66.6	33.3	460	455	497 ?	458
14L	68.5	31.5	-	455	507	455
15L	72	28	-	455	555	560
16L	74	26	-	455	620	625
17L	90	10	-	455	797	-
18L	85	15	390	453	756	-

[a] ROY, ROY, AND OSBORNE, "FLUORIDE MODEL SYSTEMS: IV. THE SYSTEMS LiF - BeF_2 AND PbF_2 - BeF_2". J. AMER. CERAM. SOC. *73*, 300-305 (1954).

Ten differential thermal analysis compositions have been run in the ternary system LiF - BeF_2 - UF_4. Complete liquidus values will be given in the next monthly report. The eutectic of the primary phase areas of LiF, Li_2BeF_4, and $Li_7U_6F_{31}$ occurs at 426° ± 5°C. The location of the eutectic has not been definitely determined, but preliminary data would indicate that it lies near 70 mole per cent LiF - 22 mole per cent BeF_2 - 8 mole per cent UF_4.

In concluding a report on a previously investigated system, BeF_2 - UF_4 an X-ray diffraction investigation was made to determine an unexplained transition break occurring between the eutectic and liquidus. The transition, occurring both on heating and cooling curves, ranged in temperature from 760°C to 790°C. Nearly twenty diffraction patterns were made of various compositions held and quenched at several temperatures bracketing the area in question. Only UF_4 and BeF_2 were found to exist in the system and therefore the differential thermal analysis break remains unassigned and unexplained.

PHYSICAL PROPERTIES

A series of 15 density and 15 viscosity runs were made on the ternary mixture of lithium fluoride, beryllium fluoride, and uranium fluoride over the temperature range of 500° to 900°C. Interpolated values at 800°C and where possible at 600°C are given in Table III.

TABLE III

DENSITY AND VISCOSITY OF LiF, BeF_2, UF_4 TERNARY MIXTURES

MOLE PER CENT			600°C			800°C		
LiF	BeF_2	UF_4	η Poise	ρ gm/cm^3	η/ρ Stokes	η Poise	ρ gm/cm^3	η/ρ Stokes
45	55	0	0.2950	1.915	0.1540	0.0619	1.843	0.0335
96	0	4	-	-	-	0.0774	1.989	0.0389
80	16	4	-	-	-	0.0546	2.203	0.0248
80	14	6	-	-	-	0.0402	2.483	0.0162
70	24	6	0.1100	2.549	0.0432	0.0491	2.471	0.0199
60	34	6	0.1240	2.546	0.0487	0.0518	2.442	0.0212
50	44	6	0.2501	2.442	0.1024	0.0617	2.358*	0.0262
92	0	8	-	-	-	0.1358	2.879	0.0472
80	12	8	0.0672	2.864	0.0235	0.0281	2.765	0.0102
70	22	8	0.0740	2.826	0.0262	0.0307	2.726	0.0113
60	32	8	0.1328	2.713	0.0489	0.0518	2.615	0.0198
50	42	8	0.2319	2.722	0.0852	0.0570	2.637	0.0216
88	0	12	-	-	-	0.0632	3.330	0.0190
76	12	12	0.1565	3.325	0.0471	0.0445	3.173	0.0140
70	18	12	-	3.217	-	-	3.128	-
60	28	12	-	3.198	-	-	3.072	-
50	38	12	-	3.042	-	-	2.980	-
80	0	20	-	4.2390	-	-	4.1425	-
70	10	20	-	4.015	-	-	3.854	-

*EXTRAPOLATED

HIGH TEMPERATURE CALORIMETER

The environment for the high temperature calorimeter will be a Leeds and Northrup calibration furnace (Cat. #9004). It has been calibrated and is now temperature controlled by a Foxboro Controller using a Chromel-Alumel thermocouple input. The furnace has a thermal resistance (R) of 1.80, 1.55, 1.39, 1.28, and 1.13 °C-watt^{-1} at temperatures 116°, 280°, 424°, 628°, and 850°C respectively. Its effective heat capacity (C) is about 9000 cal-°C^{-1}, and so the time constant (RC) is about 4.8×10^4 seconds at 628°C. This is sufficiently long for complete on-off control to be practicable. For example, when controlling at 628°C with the on-off power set at twice the steady-state demand of $\frac{(628 - 26)}{1.28} = 470$ watts, the power is on 50 per cent of the time and the temperature rises and falls at a rate of $\Delta T/RC = 602/4.8 \times 10^4 = 0.0125$°C-sec^{-1}. Since the Foxboro Controller has a ten-second period, the temperature could conceivably be maintained to ± 0.13°C if the sensitivity were higher. With a Chromel-Alumel thermocouple, a change of temperature of about 0.5°C is required to actuate the on-off switch. Since the Foxboro was factory calibrated for this thermocouple, and the calibration appears good to within 1°C, the sensitivity will not be increased at this time. The calibration was checked with a platinum resistance thermometer to 435°C and with the standard millivolt calibration chart of Chromel-Alumel to 850°C.

The first control junction and leads were secured to the inner wall of the furnace with Sauereisen No. 63 paste. The Alumel wire became extremely brittle at all points in contact with the cement.

X-RAY DIFFRACTION STUDIES

Twenty-five X-ray diffraction samples were processed during the past month.

A sample of composition $LiBe_2F_5$, initially of glassy composition, was soaked five days at 172°C. The X-ray diffraction pattern consisted of a series of somewhat diffuse lines and another series of sharp lines. It was possible to index the sharp lines as a face-centered tetragonal lattice. The diffuse lines belong to a lattice of lower symmetry as yet undetermined.

It is certain that $LiBe_2F_5$ is not a compound, but is a eutectiod. Definite identification of the phases has not yet been made. The eutectiod point lies between 218°C and 225°C.

A small fragment of Li_4UF_8 (about 0.2 x 0.2 x 0.3 mm.) was found in a thermal gradient quench prepared by N. E. Rogers. This was indicated by Laue photographs to be a single crystal. Precession photographs showed the crystal to be orthorhombic, space group Pnma (or the isomorphous diffraction group Pna). Unit cell dimensions are: $a = 5.98 \pm 0.02$ Å, $b = 9.91 \pm 0.03$ Å, $c = 10.00 \pm 0.02$ Å. Assuming a reasonable density (about 4.5), the unit cell would contain four formula units. An accurate density determination has not yet been made. This data would verify the chemical formula, which has not been rigidly determined.

A number of diffraction samples were run for B. Rhinehammer in an attempt to detect a UF_4 - BeF_2 compound which may be indicated by the differential thermal analysis work. No compound was found. If such a compound exists, it obviously does not exist metastably, and could only be detected by a suitable high-temperature X-ray spectrometer or camera.

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

A total of 130 gallons of the slurry produced in the first step of the protactinium concentration process has been processed through the solvent extraction cycle operation being carried on in 1-B of the GP-Building. Approximately 20 gallons (76 liters) of aqueous strip has been obtained from this operation. The protactinium concentration in this aqueous strip is about 10 milligrams per liter.

The raffinate from the solvent extraction operation is presently being stored. Examination of the clear aqueous phase after settling has indicated that a second extraction of it may yield an appreciable amount of protactinium. If this action is necessary, it can be easily carried out without the use of additional materials. The insoluble material in the raffinate has been separated from the aqueous phase and treated with concentrated sulfuric acid. This treatment has put additional protactinium into a solution which can be extracted with di-isobutyl carbinol. This extraction operation is more readily accomplished than the extraction in which hydrochloric acid is used to dissolve the slurry. Practically no interphase emulsion appears.

Several batches (drums) of the slurry produced by the initial concentration operation contained titanium which had been added as a carrier for the protactinium. It was necessary to treat the slurry with up to twenty per cent of its volume of concentrated sulfuric acid before adding the concentrated hydrochloric acid to complete the solution of the slurry for the preparation of the feed solution for solvent extraction. If the pre-treatment with sulfuric acid was omitted, the amount of the protactinium extracted by the di-isobutyl carbinol system was between fifty and seventy per cent in all cases.

PROCESS DEVELOPMENT

The aqueous strip solution from the first solvent extraction of protactinium is being accumulated, and work is being directed towards the development of the next step in the process.

In last month's report, preliminary experiments exploring the possibility of direct solvent extraction recycling were described. Negative results were reported in contrast to the success achieved with two precipitation methods which separated the protactinium nearly quantitatively from iron.

Further work on the solvent extraction recycle procedure, however, has resulted in the development of conditions under which more than 90 per cent of the protactinium can be extracted, effecting a further volume concentration without previous separation from iron. It is presumed that the iron separation may be carried out in the next stage of the process; either by anion exchange, or by one of the two precipitation methods previously described.

It was found, as expected, that conditions which were effective for the extraction of protactinium from one batch of aqueous strip solution did not give consistent results with other batches. Accordingly, it was decided to combine several batches in order to reduce the amount of development work required.

Composite Strip I consists of about six and one-half gallons of clear solution obtained by mixing seven individual batches of aqueous strip solution. The solution contains approximately seven milligrams of protactinium per liter. Development of extraction conditions was carried out on four-milliliter samples of Composite Strip I to which were added varying quantities of concentrated (98%) sulfuric acid and concentrated (36%) hydrochloric acid. There was no heat applied other than the heat of dilution of the sulfuric acid.

The data reported in Table I show the effect of variation in sulfuric and hydrochloric acid additions. The aqueous mixture was contacted for five minutes, immediately after addition of the reagents, with a mixture consisting of one part of di-isobutyl carbinol and one part of Amsco naphtha. The volume of the organic phase was, in all cases, the same as the volume of Composite Strip I, that is, four milliliters.

TABLE I

EFFECT OF VARIATION OF SULFURIC ACID AND HYDROCHLORIC ACID ADDITIONS ON THE EXTRACTION OF PROTACTINIUM

REAGENTS ADDED		PROTACTINIUM RECOVERED IN ONE EXTRACTION
ml H_2SO_4	ml HCl	(%)
1	4	68.7
2	4	77.9
3	4	70.0[a]
2	6	87.0
2	8	93.1
2	10	93.9

a. VIGOROUS EVOLUTION OF HYDROGEN CHLORIDE OCCURRED WHEN THE SULFURIC ACID WAS ADDED. THE ORGANIC PHASE SPLIT INTO TWO LAYERS. THE UPPER (AMSCO) LAYER BEING ALMOST COLORLESS AND CONTAINING LESS THAN ONE PER CENT OF THE Pa.

The percentages recovered were determined by making a second extraction with the same quantity of organic solution, and counting the aqueous phase before and after the second extraction. Under all conditions tested, the second extraction recovered a higher percentage of protactinium from the depleted aqueous phase than had the first extraction.

The separation of the organic phase into two layers under conditions of high sulfuric acid concentration indicated the need for an investigation of the role of the diluent used in the extraction. Although the di-isobutyl carbinol has a low density (0.82) and will separate from the aqueous phase on centrifugation, its viscosity is so high that spontaneous phase separation is slow. Table II shows the effect of variation in the kind and amount of diluent used.

TABLE II

COMPARISON OF AMSCO NAPHTHA AND BENZENE AS DILUENT FOR DI-ISOBUTYL CARBINOL (DIBC) EXTRACTION OF PROTACTINIUM

H_2SO_4 ml	HCl ml	DIBC ml	DILUENT ml	NATURE OF DILUENT	Pa IN FIRST EXTN. %	QUALITY OF PHASE SEPARATION
2	8	0.5	0.5	AMSCO	51.7	GOOD
2	8	2.0	1.0	AMSCO	88.0	GOOD
2	8	2.0	0.5	AMSCO	87.5	MARGINAL[a]
2	8	2.0	1.0	BENZENE	91.0	GOOD
2	8	2.0	0.5	BENZENE	91.0	SATISFACTORY
3	4	2.0	1.0	AMSCO	66.6	SATISFACTORY

a. ORGANIC PHASE TURBID

Because of time limitations and because the benzene diluent appears to be satisfactory for the purpose at hand, no other diluents were tested. The effect of external heating was tested by making an aqueous mixture consisting of 4.0 milliliters of Composite Strip I, 2.0 milliliters of concentrated sulfuric acid, and 8.0 milliliters of concentrated hydrochloric acid. The mixture was heated for five minutes at 100°C, during which time there was vigorous evolution of hydrogen chloride. Gas evolution ended before the end of the heating period While the mixture was still hot, a one-to-one mixture of DIBC and Amsco was added and the phases were mixed vigorously for five minutes. The organic phase contained 91.7 per cent of the protactinium, a figure which is comparable to the 93.1 per cent value obtained without external heat.

The use of hydrogen peroxide as a complexing agent for titanium, mentioned briefly in last month's report, has not yet been tested extensively. On the basis of qualitative observations on the colors of the aqueous and organic phases, however, it is possible that this reagent may have some value in the solvent extraction purification of protactinium

ION EXCHANGE PROCESS DEVELOPMENT

Experiments have continued on the purification by ion exchange of protactinium in the aqueous strip solution from the solvent extraction operation

REMOVAL OF FLUORIDE FROM HYDROCHLORIC ACID - HYDROFLUORIC ACID EFFLUENT. The last technical activities report described how boric acid or aluminum chloride, when added to the acid effluent containing protactinium, served to neutralize the fluoride and render the protactinium sorbable by a fresh Dowex-1 column. However, final elution of the column with 1-normal HCl only slowly removed the protactinium. Accordingly, effort has been concentrated on increasing this elution rate. The feed used in these experiments was prepared in the manner described in the previous report (c.f. run 59). As before, a Dowex-1 column, 1 centimeter in diameter and 4-centimeters long, was used for the fluoride removal experiments with flow rates maintained at one-half milliliter per minute. Since nearly identical elution curves were obtained with the boric acid and aluminum chloride treated feeds, expansion of the study of the elution characteristics has employed only the boric acid treated feed. As before, the feed was made one-tenth normal in boric acid.

The effect of increasing the concentration of the hydrochloric acid elutriant was studied in one series of experiments. Each of the runs employed 25 milliliters of the boric acid treated feed, followed by 40 milliliters of 9-normal hydrochloric acid wash. Table III gives the accumulated per cent of the protactinium eluted at different elution stages for three values of the acid concentration of the elutriant. A marked improvement in elution rate was observed when the hydrochloric acid concentration of the elutriant was increased from 1-normal to 2-normal. However, an increase to 3-normal failed to give further improvement.

TABLE III

EFFECT OF CONCENTRATION OF HYDROCHLORIC ACID ELUTRIANT ON RATE OF ELUTION OF PROTACTINIUM AFTER SORPTION FROM BORIC ACID TREATED FLUORIDE EFFLUENT: ACCUMULATIVE PER CENT PROTACTINIUM ELUTED FOR VARIOUS EFFLUENT VOLUMES

NORMALITY OF HCl	ACCUMULATED ELUTION EFFLUENT VOLUME (ml) 10	20	30
	ACCUMULATED PROTACTINIUM ELUTED (%)		
1	10	32	37
2	30	40	46
3	28	41	47

In a second series of experiments, the influence of the quantity of feed on elution rate was studied. Table IV gives the results of four runs, employing 15, 25, 40, and 100 milliliters of the boric acid treated feed respectively. In each case, the column was washed with 40 milliliters of 9-normal hydrochloric acid before eluting with 2-normal hydrochloric acid. No breakthrough occurred during sorption and washing, even with the run employing 100 milliliters of feed; thus indicating that the breakthrough capacity has been greatly increased by the removal of the iron in the earlier ion exchange process. Table IV shows that the elution rate steadily increased as the quantity of feed was increased from 15 milliliters, through 25 milliliters, to 40 milliliters. However, when the feed was further increased to 100 milliliters, the elution rate decreased, approximating the value obtained with the 25-milliliter run. The data suggest a maximum elution rate, lying between 40 milliliters of feed and 100 milliliters of feed.

TABLE IV

EFFECT OF QUANTITY OF BORIC ACID TREATED FEED ON RATE OF ELUTION OF PROTACTINIUM WITH 2-NORMAL HYDROCHLORIC ACID: ACCUMULATIVE PER CENT PROTACTINIUM ELUTED FOR VARIOUS EFFLUENT VOLUMES

	ACCUMULATIVE ELUTION EFFLUENT VOLUME (*ml*)			
	10	20	30	40
VOLUME OF FEED (*ml*)	ACCUMULATED PROTACTINIUM ELUTED (%)			
15	22	28	32	36
25	30	40	46	48
40	44	52	56	59
100	29	38	42	45

The effect of substituting 3-normal sulfuric acid for 3-normal hydrochloric acid as elutriant is shown in Table V. The hydrochloric acid gave superior initial elution rates; however, the integral elution curves crossed at about 35 milliliter of effluent.

TABLE V

ELUTION OF PROTACTINIUM WITH 3*N* HYDROCHLORIC ACID AND 3*N* SULFURIC ACID AFTER SORPTION FROM BORIC ACID TREATED FLUORIDE EFFLUENT: ACCUMULATED PER CENT PROTACTINIUM ELUTED FOR VARIOUS EFFLUENT VOLUMES

	ACCUMULATIVE ELUTION EFFLUENT VOLUME (*ml*)			
	10	20	30	40
ELUTRIANT USED	ACCUMULATED PROTACTINIUM ELUTED (%)			
3*N* HCl	28	41	47	51*
3*N* H_2SO_4	11	31	45	54

*OBTAINED BY EXTRAPOLATION

Work was started with this column on the evaluation of the separation process for 50 atomic per cent mixtures of hydrogen and deuterium. For a 0.016 inch diameter molybdenum center wire temperature of 900 °C, the column exhibits an equilibrium separation time of approximately 2½ hours. The optimum initial pressure has not been determined precisely, as yet, but preliminary data indicate that this pressure is in the range of from 0.6 to 0.8 atmosphere. In this pressure region the column exhibits an equilibrium H-D separation factor of approximately 500

The rates of hydrogen and deuterium uptake by the 1.5 kg of uranium in one of the chemical pumps are being determined. With the pump approximately 90 per cent UH_3 and 10 per cent unreacted, the uptake rate for hydrogen averaged 1.2 liters per minute for the first five minutes This pump was then completely exhausted, and the uptake rate for hydrogen was found to average 4.8 liters per minute for the first five minutes.

The uptake rate for deuterium in this pump, completely exhausted, was essentially the same as that for hydrogen in the similar case.

A total of 21 hydrogen-helium gas samples and 149 hydrogen-deuterium gas samples were analyzed on the mass spectrometer this month. This work was done in conjunction with the above experimentation.