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SEPARATION AND PURIFICATION OF A GRAM
OF PROTACTINIUM-233

MASTER

J. W. Coddington, J. R. Berreth,
R. P. Schuman, W. H. Burgus, and R. A. Deal

**PHILLIPS
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Nuclear Technology Branch

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ABSTRACT

One gram (20,000 Curies) of pure protactinium-233 has been separated from irradiated thorium and prepared as a sample for neutron cross section measurements with the MTR Fast Chopper. Development of the process is described. Irradiated thorium was dissolved in nitric-hydrofluoric acid. Separation of protactinium from thorium, uranium and most fission products was obtained by three successive co-precipitations with MnO_2 , each of which carried about 90% of the Pa-233. Manganese was subsequently removed by leaching the final Pa-bearing MnO_2 with dilute nitric acid plus hydrogen peroxide, leaving behind a hydrated Pa-oxide, which was then dissolved by prolonged digestion with sulfuric acid. Final recovery was by addition of iodic acid, which precipitated Pa-iodate. This was dried and fired at 800°C to Pa_2O_5 . Overall Pa-233 recovery was about 50%. Chemical analysis, gamma counting, and neutron absorption measurements showed the absence of thorium, initial uranium, manganese, or other impurities in the final product.

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OF PROTACTINIUM-233

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I INTRODUCTION

When thorium is irradiated with neutrons, U-233 is formed as the ultimate major product. The U-233 is of considerable interest because it possesses a large fission cross section and because it can be produced from a plentiful and inexpensive fertile material. Of perhaps greater importance is that the quantity η , the number of neutrons released per neutron absorbed, is significantly higher for U-233 than for U-235 or Pu-239.⁽¹⁾ Because of the relatively high abundance of thorium, and because U-233 is a relatively efficient neutron producer, the thorium-U-233 system is a promising one from the standpoint of nuclear power production, and indeed may be the only practical thermal reactor system capable of producing more fuel than is burned, i.e., capable of "breeding".

The reactions leading from thorium to U-233 are as follows:

These reactions are analogous to those for U-238, U-239, Np-239 and Pu-239 in the production of plutonium. An important difference between the two systems, however, is that the intermediate isotope Pa-233 in the U-233 production chain has a mean life over eleven times that of the analogous Np-239 in the Pu-239 chain. Therefore, before decaying to the final desired product, Pa-233 may remain in a reactor for a comparatively longer time than does its Np-239 analogue. This circumstance bestows upon Pa-233 a position of considerable consequence in the Th-232-U-233 breeding cycle. Because of its relatively long lifetime, Pa-233 can build up to concentrations where, by parasitic neutron capture, it may effect the neutron economy of the breeder; furthermore (and

worse) the capture results in production of Pa-234 which decays to relatively useless U-234 instead of valuable U-233.

A thorough evaluation of the feasibility of U-233 breeding requires a detailed knowledge of the neutron cross section of Pa-233, both in the thermal energy region and in the resonance region. The Materials Testing Reactor Fast Chopper⁽²⁾ and Radioactive Sample Changer⁽³⁾ are designed to obtain just this sort of information for radioactive isotopes. These instruments have been used previously to measure σ vs. E for Pa-233 induced in samples of neutron-irradiated thorium, without separating the protactinium from the target material.⁽³⁾ Under these circumstances, however, the presence of about an equal amount of U-233 and a hundred times as much Th-232 as Pa-233 tended to make difficult an interpretation of the data, particularly in the thermal region. Much more reliable cross section information could be obtained, however, if the Pa-233 were separated from the interfering impurities Th-232, U-233 and certain high cross section fission products. This paper describes a process, developed and applied at the MTR, for the separation and purification of a gram sample of Pa-233 oxide for cross section measurements with the MTR Fast Chopper. The results of these cross section measurements will be reported separately.⁽⁴⁾

Before describing development of the process, it is worthwhile to mention some of the chemical properties of the relatively unknown element protactinium, which sometimes displays elusive and unreproducible behavior in aqueous solution. Protactinium is only slightly soluble in most aqueous media except sulfuric and hydrofluoric acids, and easily undergoes hydrolysis in the absence of suitable complexing ions even in quite acidic solutions. It exhibits marked tendencies toward polymeric colloid formation, yielding a material not readily redissolved, and often deposits on the walls of glass vessels, remaining adsorbed with remarkable tenacity. Protactinium reveals its most capricious nature in the frequently encountered differences between its tracer level behavior and

its macro-chemistry. In fact, the scale-up of a Pa recovery process based on tracer level data can be a very frustrating experience indeed. In addition to the vexing problems associated with protactinium chemistry, a high radiation level is encountered in separating macro amounts of Pa-233 from Th-232, U-233 and fission products. The radiation is due almost entirely to the beta and gamma radiation from the Pa-233 itself; ~ 20,000 Curies of beta and ~ 10,000 Curies of 0.31 MeV gamma per gram, corresponding to ~ 50 watts of heat. The effects of the radiation and heat complicate already present chemical and precipitate handling problems. Indeed, radiation (and radiolysis) became the major aggravation encountered during the protactinium separation and recovery program at the MTR.

II PROCESS DEVELOPMENT

A. PROCESS OBJECTIVES

Of assistance in the development of any separations procedure is a knowledge of the starting conditions and of the product requirements. In this instance the starting conditions for recovering protactinium were set by the reactor irradiation of thorium, which yields in addition to untransmuted Th-232, the desired product Pa-233, some U-233, small amounts of other heavy isotopes, and some fission products. The composition of irradiated thorium has been calculated for various conditions of flux and nvt.⁽⁵⁾ Based on that work, the anticipated composition was estimated for a 100-gram thorium sample at the end of a high-flux one-cycle irradiation in the Engineering Test Reactor. The calculated composition of the irradiated thorium together with irradiation parameters are shown in Table I. In making the calculations, because of its relatively short half-life Th-233 was considered as decaying immediately to Pa-233.

TABLE I

Calculated Composition of a 100 Gram Thorium Sample
After Pile Irradiation

Thermal Flux = 5×10^{14} n/cm²-sec

Resonance flux = 1/12 thermal flux

nvt = 1.2×10^{21} n/cm² (28 day irradiation)

<u>Material</u>	<u>Amount</u>
Th-232	98.6 grams
Pa-231	~ 1 mg
Pa-233	950 mg
U-233	320 mg
U-234	65 mg
U-235	5 mg
Fission Products	60 mg

As can be seen from Table I, the starting material for Pa-233 separation contains significant amounts of fission products, about one-hundred times as much thorium and about one-third as much uranium as the desired product Pa-233. Severity of the separations problem can now be judged by a consideration of product requirements with respect to purity and amount.

From earlier work⁽³⁾, Pa-233 was known to have a thermal cross section of approximately 50 b; this compares with about 7 b for Th-232 and about 600 b for U-233. Thus the combined thermal neutron absorption by thorium and uranium before separation is about 20 times the effect of the Pa-233. A reasonable separations goal then might be to lower the Th-U effect to less than 5% of the total neutron absorption of the recovered Pa. To accomplish this, a separation factor of > 400 is required for Th+U.

An additional requirement of the final product was that it should be in the form of a thermally stable, stoichiometric compound in order that the Pa-233 content could be determined by weighing. The pentoxide, Pa₂O₅

appeared to satisfy this requirement, and was thus chosen as the final product. Further, it was necessary that the product be very finely divided in order that upon mixing with Al powder for pressing, the sample compacts would be as uniform as possible. Experiments had shown that slow thermal decomposition of the iodate followed by final heating at 800°C would produce pure pentoxide in a suitable very finely divided state. Also, the Pa_2O_5 had to be sufficiently free of impurities so that the amount of Pa could be accurately determined by weighing.

Another product requirement concerned the amount of purified protactinium needed for the final Fast Chopper sample. The sample was to consist of two Pa_2O_5 -Al compacts pressed to about 90% of theoretical density, each measuring $0.070 \times 0.375 \times 1.095$ inches. These two compacts were to be placed in a sealed sample holder for insertion in the neutron beam of the Fast Chopper. Because of its adverse effect on compact strength, no more than about 30 w% oxide could be included in a pressed sample.⁽⁶⁾ Since the sample holder could contain only two such compacts, 800 mg of Pa_2O_5 was about the upper limit of material that could be accommodated in the chopper sample container. This corresponds to 3.7×10^{21} atoms/cm² of Pa-233 in the neutron beam, and provides 83% transmission for thermal neutrons.

The processing objective, then, was to recover about a gram of Pa-233 appropriately purified from thorium, uranium, and fission products and to prepare it in oxide form suitable for weighing, mixing with powdered Al, and pressing into two small compacts. In anticipation of process losses, it was decided that sufficient Th would be irradiated during full-scale runs to produce from 1.5 to 2 grams of Pa-233. The separations work was done in the MTR Hot Alpha Cave, described elsewhere.⁽⁷⁾ Space available in the Cave for chemical processing would be provided in an isolation box about 4 feet square by $3\frac{1}{3}$ feet high in which a dissolver and several other vessels were re-

quired, in addition to supporting services. The handling of very large liquid volumes at very low Pa concentrations was consequently not possible because of space limitations. Solubility, radiation and heating problems were thus increased by use of solutions containing protactinium at concentrations of one gram per liter or greater.

B. LABORATORY STUDIES

Initial chemical processing studies employed Pa-231 as a stand-in for Pa-233. For these studies, a Pa-231 stock solution was prepared from the pentoxide as follows: About 70 mg of Pa_2O_5 was dissolved in 1 ml of 4M HF and diluted to 10 ml with 8M HCl-1M HF. No heating was required for dissolution. This stock solution was analyzed from time to time by aliquoting followed by alpha counting and pulse-height analysis and was found to be homogeneous and stable. Figure 1 shows a Pa-231 alpha spectrum taken from a counting plate prepared by sublimation of Pa_2O_5 in vacuum at $\sim 1500^\circ\text{C}$. Superimposed on this is an alpha spectrum of U-233 taken in the same manner. Since the energies of the major alpha groups differ by about 200 keV, the relative amounts of Pa-231 and U-233 in any sample could be determined by careful alpha pulse-height analysis. This was the analytical scheme used in much of the subsequent laboratory work. Because the Pa-231 had not been purified for several years, it contained a number of alpha-active daughters. The alpha spectrum in Figure 2 shows these daughter activities; at the time the spectrum was taken, Pa-231 represented 85% of the total alpha activity of the stock solution. This percentage of course was not constant; two years later, the Pa-231 contribution to total alpha activity had dropped to $\sim 72\%$.

Some laboratory experiments were carried out in an effort to separate protactinium from thorium and uranium in one pass by use of an HCl-ketone extraction system. Though di-isopropyl ketone has been shown to be an excellent extractant for Pa⁽⁹⁾, its early unavailability led to the use of

hexone (methylisobutyl ketone) as a stand-in. Initial extraction measurements were made under the following aqueous conditions: tracer U-233 and Pa-231, 0.4M Th^{+4} , 0.2M Al^{+3} , 0.1M F^{-} , and about 6M HCl . Such an aqueous phase can easily be obtained by dissolving thorium in 10M HCl with some HF added, then adjusting the resulting dissolver solution with AlCl_3 . Under these conditions, 85% of the Pa-231, 24% of the U-233, and 0.2% of the thorium were extracted in one equal-volume contacting with hexone. To learn whether separation between Pa and U could be improved, the hexone extraction of Pa-231 and U-233 was studied as a function of aqueous HCl concentration. The aqueous solutions were prepared by adding varying amounts of HCl to solutions of constant protactinium, uranium, thorium and aluminum concentrations; thus, hydrogen ion also changed with chloride ion. The initial concentration of Pa-231 was 0.5 mg/ml; that of U-233 was 0.1 mg/ml. Th^{+4} , Al^{+3} and F^{-} were as before.

Aqueous solutions were thoroughly mixed with equal volumes of hexone, centrifuged and separated. Samples of each phase were counted for Pa-231 and U-233 as previously described. The ratios of concentrations in the organic and aqueous phases ($E \frac{O}{a}$) were calculated for Pa-231 and U-233 and are plotted as a function of aqueous chloride concentration in Figure 3. The abrupt change in the effect of HCl on Pa extraction at about 7M Cl^{-} (5M H^{+}) no doubt indicates that competing reactions were occurring between two or more protactinium species, as, for example, a hydrolyzed species at low acidity and a high-chloride anionic complex at high Cl^{-} concentrations. The maximum separation of Pa and U in this system was about 10 for a single extraction. Unhappily, the range of HCl concentration over which this was true was a rather narrow one. To obtain the proper conditions for extraction would require either an exact control of dissolver stoichiometry, or an acid analysis after dissolution followed by adjustment; neither of these alterna-

tives was particularly desirable from a processing standpoint. Since our primary objective was not the elucidation of protactinium chemistry, but rather the development of a workable separative process, this line of study was not pursued further and direct extraction of Pa from dissolver solution was abandoned.

At this point an alternative scheme was examined in which initial separation of Pa from U and Th was accomplished by MnO_2 scavenging from a nitrate solution to be followed by ketone extraction of Pa from a chloride solution to separate the manganese. Two experiments at process concentrations of protactinium were performed with Pa-231 to demonstrate this combination scavenging-extraction process. For each one, synthetic nitrate dissolver solutions were prepared in which MnO_2 was formed in situ by consecutive additions of solutions of KMnO_4 and $\text{Mn}(\text{NO}_3)_2$, followed by digestion at $70^\circ - 80^\circ\text{C}$ for 10-15 minutes. This of course is a well known separative scheme for Pa^(8,9,10) and has been applied to the Thorex process,⁽¹¹⁾ in which thorium and uranium have been shown to be satisfactorily retained in solution under proper conditions of HNO_3 concentration, salt strength, and precipitate washing.

In the scavenging studies described here, MnO_2 was formed in a ratio of 10 mg of MnO_2 for each mg of Pa to be carried. Feed compositions for these experiments are shown in Table 2, and separation results are listed in Table 3. It is felt that inadequate washing contributed to the high uranium carrying in both runs. Cold tests, done to measure the carrying of thorium on MnO_2 , showed that two consecutive precipitations plus careful washings gave clean separations from thorium. After the MnO_2 had been centrifuged out and the supernatant solution decanted and analyzed, the precipitate fractions were dissolved in 8M hydrochloric acid plus 0.2M sodium nitrite and the resultant solutions sampled, adjusted to 7M HCl and extracted with hexone.

TABLE 2

Feed Conditions for MnO₂ Scavenging Experiments

	<u>Run 1</u>	<u>Run 2</u>
Volume (ml)	2.0	1.0
Thorium (<u>M</u>)	0.285	0.56
H ⁺ (<u>M</u>)	2.0	2.0
Cl ⁻ (<u>M</u>)	0.7	1.7
F ⁻ (<u>M</u>)	0.15	0.3
NO ₃ ⁻ (<u>M</u>)	1.3	0.3
Pa-231 (mg/ml)	0.62	1.26
U-233 (mg/ml)	0.21	0.43

TABLE 3

Distribution of Pa-231 and U-233 During
High Concentration Scavenging Experiments

<u>Run No.</u>	<u>Precipitate</u>		<u>Supernate</u>		<u>Material Balance</u>	
	<u>% Pa</u>	<u>% U</u>	<u>% Pa</u>	<u>% U</u>	<u>% Pa</u>	<u>% U</u>
1	83	3.1	2.7	93.6	85.7	96.7
2	90	8.6	0.6	90.4	90.6	99.0

In Run 1, another precipitate formed during extraction and contained over half of the protactinium. This precipitate failed to redissolve except in warm HF. To prevent formation of this material, fluoride was added during dissolution of MnO₂ during Run 2; this complexed the protactinium, preventing subsequent precipitate formation. Before adequate Pa extraction could be achieved, however, it was necessary to add aluminum to complex the fluoride ion. Two moles of aluminum as AlCl₃ per mole of fluoride were added for this purpose. Three equal-volume successive hexone extractions then removed 96% of the Pa present in the solvent extraction feed. Table 4 summarizes these

results. Between successive extractions in Run 2, concentrated hydrochloric acid was added to the aqueous phase in an attempt to maintain the acidity at about 7M HCl. Alpha pulse-height analyses were done on sublimed samples of the feed, precipitate product and extraction product for Run 2. These are shown in Figures 4, 5, and 6, and illustrate the successive purification of the protactinium with respect to uranium contamination.

TABLE 4

Extraction Behavior of Pa-231 and U-233
Using Hexone^(a)

Run No.	<u>1st</u> <u>Extraction</u>		<u>Extraction</u> <u>Precipitate</u>		<u>2nd</u> <u>Extraction</u>		<u>3rd</u> <u>Extraction</u>		<u>Composite</u> <u>Product</u>	
	<u>% Pa</u>	<u>% U</u>	<u>% Pa</u>	<u>% U</u>	<u>% Pa</u>	<u>% U</u>	<u>% Pa</u>	<u>% U</u>	<u>% Pa</u>	<u>% U</u>
1	23	2.3	51	0.8	--	--	--	--	74	3.1
2	30.7	0.7	--	--	25.5	0.5	30.5	0.6	86.7	1.8

- (a) The values given in this table represent the percentages of recovery of the Pa initially present before scavenging, so that the overall recovery in the "Composite Product" reflects both scavenging and extraction losses.

Protactinium was decontaminated from uranium by a factor of 8.6 during the MnO₂ precipitation and by a factor of 4.4 during the subsequent hexone extraction. Two successive, adequately washed precipitations and a ketone extraction would therefore yield an overall decontamination from uranium of about the desired magnitude.

It had been anticipated that use of di-isopropyl ketone (DIPK) in place of hexone would materially aid in Pa recovery, and indeed this proved to be the case. When it became available, DIPK was tested as an extractant for protactinium by treating a portion of the Pa-231 stock solution as follows: a portion of the 8M HCl-1M HF stock containing ~ 6 mg/ml Pa was

diluted with 6.8M CHCl_3 - 0.38M AlCl_3 so as to yield an extraction feed about 1 mg/ml in Pa and 7M HCl , with an Al/F ratio of 2. A single, equal-volume contacting with DIPK extracted 95% of the Pa-231. Thus, under these conditions, DIPK was considerable more effective as a protactinium extractant than was hexone.

C. HOT ALPHA CAVE EXPERIMENTS

As a "hot" test of the combination scavenging-extraction scheme, several full-concentration but very small-scale runs were done in the Hot Alpha Cave, using Pa-233 produced from irradiated thorium. During all small-scale hot tests, the course of Pa-233 separation was followed by holding individual vessels (centrifuge cones, beakers, etc.) close to a fixed radiation monitor. In later large-scale runs, a movable counter head was used to monitor various vessels; certain solutions were also sampled and the Pa-233 content determined by counting. In the first run, 100 mg of irradiated thorium metal were dissolved in about 0.3 ml of 13M HNO_3 - 0.1M HF so that the final solution was about 6M HNO_3 and contained 3 mg/ml Pa-233. At this point, the solution contained about 1 mg of Pa-233 and measured about 200 R/hr at 5 inches. After dilution to 3M HNO_3 with water, 0.4M KMnO_4 was added and the solution warmed. MnO_2 was then formed by addition of 0.43M $\text{Mn}(\text{NO}_3)_2$ solution. The precipitate was digested, centrifuged, and washed with dilute HNO_3 . It carried about 95% of the Pa-233. This precipitate was then dissolved in 4.5M HNO_3 plus sufficient 2M NaNO_2 solution to reduce MnO_2 and then MnO_2 was again precipitated by the addition of new KMnO_4 solution. After separation and washing, the second MnO_2 precipitate, which had carried nearly all of the Pa-233, was dissolved in HCl plus NaNO_2 to give about 0.13 ml of a 7M HCl solution, which monitored ~ 180 R/hr. An immediate but unsuccessful attempt was made to extract the protactinium into DIPK; less than 20% was extracted into 3 volumes of organic in a single contacting. Because of the

difficulty of handling such small volumes of solution with manipulators, this experiment was terminated, and a second run initiated. This second experiment started with one gram of irradiated thorium metal. Dissolution and two MnO_2 precipitates yielded about 90% of the starting Pa-233, which at this stage monitored $\sim 11,000$ R/hr. Five ml of 6.5M HCl extraction feed were prepared and contacted with 15 ml of DIPK. The former poor extraction was confirmed: again only about 20% of the activity extracted, and this time a white precipitate appeared in the aqueous phase as in earlier work with Pa-231. When HF was added to this phase, all Pa-233 activity could be decanted, leaving an inactive white precipitate, probably MnF_2 . A second attempt was made to extract the Pa by adding AlCl_3 to complex F^- ($\text{Al}/\text{F} = 2$) while mixing the aqueous phase with DIPK. Only 10% of the Pa was extracted on this second try.

Drastic measures were now taken. The aqueous phase was treated with hot 10M HF to assure that all Pa existed as an ionic species. After concentration, and an addition of aluminum, a MnO_2 precipitation was done, carrying about 80% of the Pa-233. (The precipitate monitored 6000 R/hr at this point, the supernate about 1000 R/hr.) Following a second such precipitation, the Pa-bearing MnO_2 , monitoring 5000 R/hr, was dissolved in 2 ml of 8N H_2SO_4 - 6N HCl plus a drop of H_2O_2 . This solution was extracted with di-isopropyl carbinol, but about 98% of the activity stayed in the aqueous phase. Apparently the Pa-233 was in a non-extractable form, either highly polymeric or tightly complexed (high concentrations of radiolytic products were present).

It is not clear at just what point during the above procedure the protactinium became so intractable. Both "hot" experiments indicate that the dissolution of Pa-233-bearing MnO_2 yields a form of protactinium which is largely inextractable, though this was clearly not always the case for Pa-231. Radiolytic formation of H_2O_2 , a known complexing agent for Pa, could well have accounted for this extraction difference between the "hot" and

"cold" experiments, since it is not hard to imagine a Pa-peroxide species which, though inextractable, could be easily carried during precipitation.*

In any event, extraction was now abandoned as a method for separating Mn and Pa in favor of the iodate precipitation of Pa, leaving Mn in solution. Cold tests with $\text{Zr}(\text{IO}_3)_4$ and $\text{Th}(\text{IO}_3)_4$ as stand-ins for Pa-iodate indicated that two such precipitations would yield protactinium sufficiently free of manganese. The two consecutive iodate precipitations were to be coupled by a methathesis with strong ammonium hydroxide, followed by sulfuric acid dissolution before preparation of the final product.

This scheme was tried in two more small-scale, but full-concentration, hot runs. For the first time in the development work, small fritted glass filters were used to simulate precipitate techniques planned for the full-scale runs. The second of these experiments in addition made use of a small stainless steel dissolver, fritted glass filters, and glass centrifuge cones interconnected with plastic and stainless steel tubing as a mockup of the final apparatus. Except for an unfortunate spill during the second run, in which over half of the product was lost, these experiments demonstrated that

* Alternatively, it is possible that the nitric-hydrofluoric acid dissolutions of Th-Pa in all of the "hot" runs never had produced a truly ionic (and thus extractable) species of protactinium. The Pa-231 work on the other hand was done using synthetic dissolver solutions wherein the Pa was "spiked" from a true, stable solution or added to the system by stripping from an organic ketone phase, in which case the protactinium had already proved to be extractable. In this sense, then, the Pa-231 work did not truly simulate conditions in the "hot" Pa-233 experiments; unfortunately, "cold" simulation of "hot" dissolution work is very difficult, if indeed it is possible at all.

Subsequent experiments with Pa-233 showed that the white precipitate which formed when MnO_2 carrying Pa was dissolved in HCl or $\text{HNO}_3 + \text{NaNO}_2$ could not be redissolved in HCl or HNO_3 , but could be dissolved in 6M H_2SO_4 . Such sulfate solutions could be filtered through sintered glass without loss of Pa; also, by diluting the sulfate solution to $\sim 1.5\text{M}$ H_2SO_4 , the Pa could again be carried nearly quantitatively on MnO_2 formed in situ. Because sulfate solutions appeared stable, H_2SO_4 was substituted for HNO_3 and HCl in the process. Additional tests were made to see whether process concentrations of Pa-233 could be extracted from 9M $\text{H}_2\text{SO}_4 + 6\text{M}$ HCl with di-isobutylcarbinol.⁽⁹⁾ Such tests showed a poor but nearly reproducible extraction of about 30% per pass.

the chosen process appeared workable.

In each run, 1.3M HNO_3 - 0.1M HF was used as dissolvent for the irradiated Th. Two consecutive MnO_2 precipitations recovered $> 90\%$ of the Pa-233 for each run. In each case, the first MnO_2 was dissolved in $6\text{M H}_2\text{SO}_4$ plus H_2O_2 and diluted to $\sim 1.2\text{M H}_2\text{SO}_4$ before the second MnO_2 was formed. Two consecutive iodate precipitations then followed: the second MnO_2 was dissolved in H_2SO_4 as before, diluted to $1.5\text{M H}_2\text{SO}_4$ and the Pa precipitated by addition of a 10-fold excess of 1M HIO_3 . The slurry was agitated and digested for about 15 minutes and filtered. After a water wash, the protactinium iodate was treated on the filter for 20 minutes with $6\text{N NH}_4\text{OH}$. Following a second water wash, this hydrated Pa-oxide was dissolved in $6\text{M H}_2\text{SO}_4$, diluted and a second iodate precipitated. This was washed with dilute H_2SO_4 - HIO_3 and transferred to a Gooch crucible for drying and firing. (It was during this final transfer that a spill occurred in the second run.) The iodate was dried under a heat lamp until the filter paper was charred, and then ignited to the oxide at 800°C for one hour: this scheme gave a fine easily-handled powder with no "clinker" formation.

The first run produced 10.1 mg of nearly white Pa_2O_5 with a specific activity at ~ 3 -inches from the alpha cave monitor of $\sim 2500 \text{ R/hr/mg}$. This represented $\sim 85\%$ protactinium recovery in the final oxide product. The second run produced 9.7 mg of pure white Pa_2O_5 with a specific activity of about $\sim 3000 \text{ R/hr/mg}$. This represented $\sim 40\%$ of the starting Pa; discounting the spill, these two runs both demonstrated good recovery. The oxide from the second run was mixed with aluminum, pressed into a compact, and used for cross section measurements on the MTR Fast Chopper. The cross-section data indicated that no significant quantities of materials other than Al and Pa_2O_5 were present in the sample.⁽¹²⁾ Gamma-ray spectroscopy indicated no detectable radioactive impurities. Thus what seemed to be an adequate flowsheet had been

developed and hot-tested.

D. EQUIPMENT

Preparation of the Hot Alpha Cave for handling 20,000 Curies of Pa-233 presented problems in two general categories: 1) Modification of the Cave to permit handling the very highly radioactive material involved, and 2) installation of the process equipment itself.

A 1/2-inch thick lead scatter-shield was installed above the working face of the Cave to shield the operators from "skyshine" due to beams of radiation emerging from manipulator holes in the Cave roof. Also, lead rings were placed around the manipulator holes to reduce the "open" area. The Cave front itself provided more than adequate shielding, and as a result of the extra lead scatter-shield, operators received much less than the maximum permissible dose during the Pa-233 separations.

The early small-scale "hot" tests had shown that iodine fission products were evolved in sufficient quantity during processing to seriously (up to 5 R/hr at contact) contaminate the cave box vent pipe and vent caustic scrubber.⁽¹³⁾ To prevent escape of iodine, the cave box was equipped with an activated charcoal trap which proved so effective that thereafter no escape of iodine was noted. The trap consisted of a box about 7 x 7 x 7 inches inside, filled with 14-20 mesh activated charcoal and preceded by an absolute filter. All of the sweep air from the isolation box was drawn through this filter and into the vent scrubber system.

Provision was also required for removal of the final cross-section sample from the highly contaminated environment of the press box. This high level of contamination resulted from working with very highly active and finely divided Pa_2O_5 powders; it proved nearly impossible to load Chopper blocks in the Hot Alpha Cave Mechanical Box without serious surface contamination of the blocks. Consequently, in the first full-level runs, the Pa_2O_5 sample holder was loaded

into a cask from the Alpha Cave Mechanical Box, transferred to another hot cell, washed, and loaded into a clean Chopper block, with no detectable surface contamination resulting. In the last full-level run, a small lucite transfer box was constructed which fit into the transfer port of the Cave. This box was equipped with a simple magnetic manipulator to load the sample into the Chopper block, and thence into the Chopper cask. This scheme held block contamination to a minimum, and made it unnecessary to transfer the sample to another hot cell.

Special sample holders and Chopper blocks were built to adequately contain the highly radioactive Pa-233 samples and still provide easy remote loading and sealing capabilities inside the Hot Cell. Both of these containers were held shut by spring-loaded latches, and were leak tested to 20 lbs/in². The seals were made with special radiation-resistant rubber O-rings which are pliable to $> 10^9$ R/hr as tested in the MTR Gamma Facility.⁽⁶⁾

Equipment for chemical processing was contained in an isolation box located inside the Hot Alpha Cave and operated by Central Research Lab Model 7 manipulators. The box was maintained at a pressure 1/2-inch of water less than the Cave, and was provided with a hot liquid waste line and a hot waste cask for disposal of solid waste. Provision was made for entrance of chemical reagents from a pressure tank outside the Cave capable of attachment to any of a number of polyethylene lines, each going through a shut-off valve prior to entry to the Cave. These lines entered the isolation box through leak-proof bulkhead fittings and were connected to their respective vessels with nylon Swagelock unions.

A diagrammatic sketch of the full-level processing equipment is shown in Figure 7. The dissolver was a flanged four-liter stainless steel vessel with a standard Pyrex reaction kettle top. It was equipped with a reflux condenser and an Ascarite trap on the dissolver vent. There were two filters

and receivers for handling process precipitates. The filters were sintered glass filter funnels with tops sealed onto them to provide total enclosure. The receivers were standard two-liter Pyrex reaction kettles with tops. Glass was chosen for much of the process equipment since it was necessary to observe visually the progress of certain process steps. The final product filtration was made in an open 600 ml stainless steel Millipore filter which had a two-liter suction flask solution receiver. All vessels were interconnected with polyethylene or stainless steel tubing so that solutions and slurries could be transferred from any vessel to another or to waste. Connections were made with stainless steel Swagelock unions, plastic tubing connectors, rubber stoppers, or ground glass joints. All transfers were made either by vacuum or pressure, using a vacuum-pressure pump inside the cave isolation box. All operations prior to the final Pa-iodate filtration were made in a closed system. A heat lamp was provided for precipitate drying.

Adjacent to the chemical box, also inside the Hot Alpha Cave, was a mechanical box of similar dimensions, for sawing open irradiation capsules, firing precipitates, and mixing, weighing and pressing samples. This equipment is described elsewhere.^(6,7)

III FULL-SCALE PROCESSING

Two separate full-scale irradiations of thorium were made. From the earlier small-scale hot work, it was expected that only about 1/2% of the thorium would be converted to Pa-233, or about half of the calculated value. For the first run, 280 grams of thorium metal were placed in a high flux position (K-7-SW) in the ETR and irradiated for one cycle. The thorium was in 16 pieces, each 2 x 5 x 0.15 cm, placed between aluminum heat sinks in four seal-welded aluminum capsules. Use of thin plates minimized neutron self-shielding during irradiation. At the end of the irradiation cycle, the

capsules were cooled for ~ 4 days to permit some fission product decay, and then cut open inside the mechanical box using a remotely-operated power hacksaw. The Th pieces were separated from the capsule parts and the unclad Th was then transferred to the dissolver. The bare metal pieces monitored 2.9×10^5 R/hr at 3 inches using the movable cave monitor.

What had been planned for a thirty-hour chemical processing ran actually took five days of nearly continuous operation. The Pa was separated and purified during the first run by essentially the method described above. Some of the procedures used turned out to be different from those of the small-scale experiments, and nearly all the steps required a much longer time than anticipated.

The 13M HNO_3 - 0.1M HF dissolvent was added incrementally so that the dissolving proceeded at a reasonable rate, with heat being applied at the end to assure complete dissolution. Completion of the reaction was detected by noting the cessation of NO_2 evolution. The dissolver solution was diluted, air-sparged to assure adequate mixing, and sampled to determine the Pa-233 content. This was done by absolute gamma counting. The 2720 ml of dissolver solution contained 1.38 grams of Pa-233, which corresponds to 0.49% conversion of the Th-232 to Pa-233.

An MnO_2 precipitation was carried out as previously described, and the slurry was transferred to the first glass filter. A noticeable amount of MnO_2 redissolved during the filtration, presumably due to radiolytic products reacting with it. When the filtered precipitate was washed with 0.5M HNO_3 , the remaining MnO_2 all dissolved, leaving about 95% of the starting Pa-233 on the filter as hydrated oxide. Whereabouts of the Pa-233 was followed in a semi-qualitative way with the movable-head radiation monitor.*

* When the Pa-233 was concentrated in one place, as on a filter, its presence could be readily detected by darkening the Cave room and observing its glow.

With the Pa-233 on the filter, the vessel monitored 5.9×10^5 R/hr. The Pa slowly redissolved off of the filter using $6M$ H_2SO_4 . Dissolution of the hydrated oxide was very much slower than the dissolution of $MnO_2 + Pa$ during the earlier small-scale tests again suggesting that true Pa dissolution had not been achieved during the earlier work. After the dissolution was complete, the solution was transferred to the dissolver and a second MnO_2 precipitation made by adding $Mn(NO_3)_2$ and $KMnO_4$ solutions. This MnO_2 filtered much more rapidly than the first and was successfully washed without redissolving; however, it carried only about 70% of the Pa, and so the filtrate was saved for possible recovery. When the second MnO_2 was dissolved in 380 ml of $6M$ $H_2SO_4 + H_2O_2$, a cloudy solution resulted, which when filtered produced a clean solution containing the protactinium, and monitoring about 2.3×10^5 R/hr.

When excess $1M$ HIO_3 was added to the solution, the Pa-iodate was at first whitish and milky as expected, but it soon turned black. Iodine vapor from radiolytic decomposition of IO_3^- was observed and some of the blackness was doubtless due to elemental iodine. The darkened precipitate was filtered without excessive Pa loss, and after an H_2O wash was metathesized on the filter by treatment with excess $6M$ NH_4OH . Subsequent addition of $6M$ H_2SO_4 failed to completely re-dissolve the precipitate, no doubt due to the presence of residual iodate, and further treatment with $6M$ $H_2SO_4 + H_2O_2$ was necessary to effect complete dissolution. After the dissolution was complete (about seven hours were required), a second Pa iodate was formed, again yielding an initially whitish precipitate, which rapidly turned black. This precipitate was filtered onto the Millipore filter, washed with water, transferred to a crucible, dried, and ignited at $800^\circ C$. The final oxide product, measuring 2.35×10^5 R/hr, was dark brown due to a considerable amount of Mn_2O_3 . Evidently because of radiolytic destruction of IO_3^- with simultaneous reduction of H^+ concentration, the iodate precipitations had failed to remove all of

the manganese despite the fact that cold tests and the small-scale hot tests had shown previously that a clean separation could be expected. The final product was analyzed (see below) and found to contain 390 mg of Pa_2O_5 and 1860 mg of Mn_2O_3 ! Since Mn has no resonance below 100 eV, and thus does not seriously interfere with lower energy resonance measurements, a small chopper sample containing 130 mg of Pa-233 was made. This was used to study Pa-233 resonances. The remaining Mn-contaminated oxide was reprocessed by dissolving through digestion with HF and H_2SO_4 in a platinum crucible followed by fuming with H_2SO_4 . Part of this solution was analyzed colorimetrically for manganese, by gamma counting for Pa-233 and by alpha pulse-height analysis for U-232. That uranium had been carried through the process was shown by the U-232 counting and by observation of a neutron absorption resonance of U-234 in the chopper sample. However, the cross-section data showed that the oxide contained very little thorium. Pa recovery was only about 30%, considerably below that anticipated.

Two weeks after the first run, the remaining Mn-contaminated Pa solution was added to the Pa-containing solutions left from the first separation and the composite reprocessed to recover a second Pa sample.

The reprocessing consisted first of carrying the Pa on MnO_2 , after which manganese was leached away from the hydrated Pa_2O_5 by treatment with 0.5N HNO_3 plus 0.75% H_2O_2 . This unexpected separation had occurred during the first run as described above, and in this second run was intentionally used as a separative scheme. The leaching, though slow, gave a good separation from manganese without appreciable loss of protactinium. The hydrated Pa_2O_5 left on the filter was then dissolved by long digestion with 6M H_2SO_4 . The solution was sampled and colorimetrically analyzed for Mn and gamma counted for Pa-233; it contained 240 mg Pa-233 and less than 80 mg of Mn. This latter value represents the lower limit of detection for manganese under

these conditions since very large dilutions were required in order to bring a sample out of the Cave for colorimetric Mn determination. (Actually Mn had been removed to undetectable levels as measured by non-observation of the 2000 barn 337 eV resonance of Mn-55.) The Pa-233 solution was then diluted to 1.2M H_2SO_4 and Pa-iodate precipitated. This precipitate was white and remained so. It was filtered, washed, dried and ignited as before. The ignited product was white and weighed 264 mg and appeared to be essentially pure Pa_2O_5 . Two hundred and forty-five mg of it was prepared in a chopper sample. After correcting for decay, this second sample represented another 1/3 of the initial Pa.

In order to increase the effective thickness of the second Pa chopper sample, the Al- Pa_2O_5 compact was cut in half and one of the halves was placed in front of the other in the neutron beam. In this way a Pa sample containing 852 mg Pa-233/cm² was made, providing a reasonably thick sample for thermal cross section measurements.

Two months later, a second, large-scale thorium irradiation was completed, and the third (and largest) Pa-233 chopper sample was prepared, using the same processing equipment as before. However, a number of modifications and replacements were needed before the system was suitable for processing work. Radiation embrittlement had affected polypropylene tubing, nylon fittings, glass tubing, and rubber stoppers. Wherever these components were subjected to any stress, they had to be replaced. With the aid of specially designed wrenches, all the necessary maintenance was accomplished remotely, including replacement of Swagelock tubing fittings, modification of filter entry ports, etc. Despite the high radiation dose to which the fritted glass filters had been subjected, their usefulness was unimpaired, and they continued to function perfectly throughout the second run. Glass vessels had become very dark brown, and strong back-lighting was necessary to see their contents. The

charcoal iodine trap was also replaced.

Sixteen thorium pircs were irradiated as before, and were de-clad and dissolved after less than one day's cooling. An analysis of the dissolver solution indicated that 2.0 grams or over 40,000 Curies of Pa-233 had been formed during irradiation. The thorium was dissolved and two MnO_2 precipitations were done in the usual manner. A sample of the first filtrate showed a Pa-233 content of ~ 200 mg. This was discharged to waste as representing only about 10% of the total Pa. The second filtering was interrupted for about three hours while a leaking transfer line was being repaired. As a result of this delay, most of the second MnO_2 precipitate was re-dissolved through chemical reaction with radiolytic products, and the filtrate, which contained about 700 mg of Pa-233 by analysis, had to be recycled for another precipitation. This time the filtration resulted in adequate Pa recovery, as observed by the movable radiation monitor. Yet a third MnO_2 precipitation was done in like manner in order to assure that the protactinium was adequately decontaminated from thorium and uranium. The final MnO_2 precipitate, which by radiation monitoring contained about 80% of the starting Pa (it measured 2.6×10^5 R/hr), was leached as before with $\text{HNO}_3 - \text{H}_2\text{O}_2$, thus removing the manganese without loss of Pa, which was then subsequently dissolved by prolonged digestion with $6\text{M } \text{H}_2\text{SO}_4$. At this point, the pure Pa-233-sulfate was contained in 195 ml of clear, bubbling solution. Analysis showed that 940 mg of Pa-233 product had been recovered. No manganese was detected. The voluminous, white precipitate resulting from HIO_3 addition was filtered, washed and transferred to a porcelain crucible for drying and ignition. No external heat was required for drying; the nearly 20,000 Curies of glowing precipitate generated sufficient heat not only to dry itself, but to cause decomposition with evolution of iodine. The material was finally ignited at 800°C to assure complete conversion of iodate to oxide. A quantity of 808.4 mg of

pure white Pa-233-oxide was recovered, 787.6 mg of which were compacted and used for checking the thermal and resonance cross sections obtained with the previous samples. The cross section measurements detected no manganese, thorium or initial uranium in this final Pa-233 sample,⁽⁴⁾ and from all indications, the sample contained only aluminum and Pa-233 oxide. The processing flowsheet used to prepare the final Pa-233 sample is shown in Table 5.

A few summary comments are in order regarding the processing of gross amounts of Pa-233. Most of the troubles encountered during the one gram processing runs were due in large measure to the very high radiation levels present. Solutions bubbled much more than those in the small-scale but full-concentration experiments, indicating that the larger solution volumes absorbed a much higher fraction of the Pa-233 radiation than did the small volumes. There was a correspondingly greater tendency in the full-scale work for radiolytic products to dissolve MnO_2 and to reduce iodate, particularly in the presence of manganese. Self-heating of the carrier-free Pa-iodate was sufficient to dry and ignite the precipitates sooner than anticipated, making difficult their subsequent handling. Compacts and sample holders were sufficiently hot to char paper, and had to be stored on metal plates to keep them reasonably cool. Very high levels of contamination were present inside the isolation boxes, making it difficult to remove solution and chopper samples. Prolonged use of processing equipment resulted in material failure due to radiation damage; use of nylon and polypropylene fittings in particular should be avoided in the construction of equipment to operate at levels of radiation comparable to those encountered in this work.

TABLE 5

FLWSHEET FOR SEPARATION AND PURIFICATION
OF ONE GRAM OF PROTACTINIUM-233

Basis: 200 grams Th yield 1 gram Pa-233

1. Th Dissolution 800 cc 13M HNO₃ - 0.1M HF
 1200 cc diluent H₂O after dissolution is complete
2. First MnO₂ 160 cc 0.3M KMnO₄
 170 cc 0.43M Mn(NO₃)₂
 Digest at 70-80°C for 15 minutes
 Filter
 250 cc 0.05M HNO₃ cake wash
3. Precipitate 210 cc 6M H₂SO₄ + 10 cc 30% H₂O₂
Dissolution 840 cc diluent water
 Return solution to dissolver
4. Second MnO₂ 160 cc 0.3M KMnO₄
 Digest at 70-80°C for 15 minutes
 Filter
 250 cc 0.05M HNO₃ cake wash
5. Precipitate 210 cc 6M H₂SO₄ + 10 cc 30% H₂O₂
Dissolution 840 cc diluent water
 Return solution to dissolver
6. Third MnO₂ 160 cc 0.3M KMnO₄
 Digest at 70-80°C for 15 minutes
 Filter
 750 0.05M HNO₃ cake wash
7. MnO₂ Leaching 100 cc 0.5M HNO₃ + 2.5 cc 30% H₂O₂
 Repeat 6 times followed by
 500 cc H₂O for Pa wash
8. Pa Dissolution 50 cc 6M H₂SO₄
 Digest on filter overnight
 50 cc 1.2M H₂SO₄ filter wash
 100 cc diluent water
9. Pa Precipitation 300 cc 1M HIO₃
 Digest for 15 minutes with stirring
 Filter
 100 cc wash/2M H₂SO₄ - 0.1M HIO₃
10. Dry Pa-Iodate Cake
11. Ignite Iodate to Oxide
at 800°C

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Fig. 1 - Alpha Spectra of U-233 and Pa-231
Taken From Sublimed Samples.

Fig. 2 - Alpha Spectra of Sublime Pa-231
Showing Daughter Activity.

Fig. 3 - Extraction of Protactinium and Uranium From Chloride System With Hexone.

Fig. 4 - Alpha Pulse Height Analysis Showing Pa-231 and U-233 - Run 2, Feed.

Fig. 5 - Alpha Pulse Height Analysis Showing Pa-231 and U-233 - Run 2, Precipitation.

Fig. 6 - Alpha Pulse Height Analysis Showing Pa-231 and U-233 - Run 2, Extraction Product.

Fig. 7 - Diagram of Pa-233 Process Equipment