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Radioactivities Produced in the Platinum Group by Bombardment of Uranium with 400 Million Electron Volt Helium Ions

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RADIOACTIVITIES PRODUCED IN THE PLATINUM GROUP BY
BOMBARDMENT OF URANIUM WITH 400 MILLION
ELECTRON VOLT HELIUM IONS

Richard D. White

Special Review of Declassified Reports
Authorized by USDOE JK Bruton
Unclassified TWX P1822062 May 79

REPORT PROPERLY DECLASSIFIED

J N Coen 8/16/77
Authorized Derivative Classifier Date

R D Hunt 8/17/77
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Berkely, California
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University of California
Radiation Laboratory
Berkeley, California
Radioactivities Produced in the Platinum Group by Bombardment of Uranium with 400 Million Electron Volt Helium Ions

Richard D. Wolfe
Radiation Laboratory, University of California
Berkeley, California

ABSTRACT

The purpose of the investigation was to determine the yield of radioactivities of the platinum group metals (ruthenium, rhodium, palladium, osmium, iridium, and platinum) produced when natural uranium is bombarded with 400 Mev helium ions. The reason for interest in the yields of these metals is that the last three lie in a region where the plot of yield vs. mass number may change from a fission-yield curve to a spallation-yield curve. In addition, the first three lie in a region which should indicate whether the center of the curve has a dip (as does the slow-neutron fission-yield curve), or a single maximum indicating symmetrical fission (as in the case of bismuth and thorium).

A scheme of chemistry is presented for the radiochemical isolation of the platinum metals from the other fission products and from each other. The radioisotopes produced are discussed, their decay and energy values presented in the text and in the form of plots, and finally the yield data are presented in tabular and graphical form. The yield data indicate that the fission-yield curve does have a single maximum in the vicinity of mass 120, and that it apparently becomes continuous with a spallation-yield curve at a minimum of about 0.1% the yield of Ba$^{140}$, around mass 180.

To be declassified for use as Thesis.

Introduction

The 184-inch cyclotron of the Radiation Laboratory of the University of California at Berkeley produces helium ions at a maximum energy of 380 to 400 million electron volts, with a beam strength of about one microampere. With this ultra high energy, a great multiplicity of nuclear reactions occurs when a fissionable material such as uranium metal is intercepted by the cyclotron beam. Previous work by O'Connor on uranium fission\(^1\), Goeckermann and Perlman on bismuth fission\(^2\), and Newton on thorium fission\(^3\), indicates it is probable that the whole spectrum of fission products found in the thermal-neutron fission of uranium will appear, although very likely with differing yields. Besides the fission products there should be found a number of nuclei formed by "spallation", in which the impinging helium ion causes the chipping out of many nuclear particles, leaving a new nucleus which may decay through an isobaric chain as do the majority of fission products.

Further, it seems likely that the plot of yield versus isotopic mass number, starting at the heavy end with the target nucleus uranium, may show a continuous trend downward through the decreasing mass numbers of the heavy elements until it becomes continuous with a fission-yield curve in the vicinity of mass 180 at a
minimum in the curve. At present it is impossible to determine from radio-
chemical analysis of the end product whether it was formed by spallation or by
fission; i.e., one cannot state at what point the curve ceases to be a spallation-
yield curve and becomes a fission-yield curve.

In order to determine whether this continuity in the yield curve does in
fact exist it was of interest to examine the radioactivities produced in the
elements osmium, iridium, and platinum by bombardment of uranium with full energy
helium ions, and since the chemistry is similar, to repeat earlier examination of
the other three platinum metals ruthenium, rhodium, and palladium. The first
three have known radioactivities with mass numbers ranging from 185 to 199, near
the supposed minimum point where presumably spallation-yield and fission-yield
curves may be found to join.

These experiments were carried out under the direction of Professor Glenn T.
Seaborg. Grateful acknowledgment is made to Dr. N. E. Ballou, who participated in
and lent direction to the development of much of the scheme of chemistry. Dr. A.
S. Newton likewise has been most helpful with guidance for many of the experi-
ments. I wish to thank Mr. James T. Vale and Dr. Duene C. Sewell and the cyclo-
tron crew for their cheerful cooperation in conducting the bombardments.

This research was supported by the Atomic Energy Commission under contract
No, W-7405-Eng-48, with the Radiation Laboratory, University of California.
Radiochemistry of the Platinum Elements

The chemistry of the six platinum elements $^{44}$Ru, $^{45}$Rh, $^{46}$Pd, and $^{96}$Os, $^{77}$Ir, $^{78}$Pt is notoriously complex. Their separation, purification, and quantitative determination is a classical problem of long standing. The electron configuration of the first three partially fills the 4d orbit, with the possibility of adding 10, 9, and 8 electrons respectively for ruthenium, rhodium, and palladium in order to fill out the 5s and 5p orbits and form the stable noble-gas configuration of 54$^4$Xe. Similarly, osmium, iridium, and platinum partially fill the 5d orbit, and can add 10, 9, and 8 electrons respectively, filling the 6s and 6p orbits to attain the noble gas structure of 86$^6$Rn. Because of this electron structure, all six elements enter into a vast number of coordination complexes, with almost any radical or element, charged or neutral, which can furnish an unpaired electron. Their chemistry is thus characterized first by extensive complex formation in which all six behave very similarly and are therefore difficult to separate; and secondly by difficulty in getting the elements into solution, and once they are in solution, further difficulty in getting them quantitatively out of solution.

A number of satisfactory schemes are presented in the literature for quantitative and qualitative determination of these elements. However, these schemes are generally inapplicable, without extensive modification, to fission product procedures. In the first place, the classic methods aim at separating the platinum metals from ores or naturally occurring sources, where contaminating substances are relatively few, and are fairly well known as to type. Secondly, these schemes aim at quantitative determination of the platinum elements, entailing precise, time consuming, cumbersome, and involved chemical procedures.
The fission product procedure for these elements has a different aim. Although 100% recovery of the six elements sought is not required provided their chemical yield can be ascertained, decontamination factors from other radioactive elements must often be as large as $10^6$ and sometimes larger, since activities in osmium, iridium, and platinum are very low compared to activities of contaminants. In addition, separation must be made not from a few known elements occurring with these metals in natural ores, but from the majority of the elements in the periodic table. Finally, the separations performed must be relatively specific and rapid, in order to permit observation of the shorter-lived periods.

With the foregoing requirements in mind, the literature was surveyed for reactions which might be combined to achieve the separations desired. Fission-product procedures for ruthenium and palladium are extant. That developed for rhodium was insufficient, in that no separation from iridium had been provided for. No fission-product procedures existed for osmium, iridium, and platinum, although the standard qualitative method of distilling osmium tetroxide seemed adaptable to the requirements. Thus the separation of iridium, rhodium, and platinum comprised the chief problems.

It seemed likely that the pyridine separation for rhodium developed by Ballou$^5$ might be adaptable also to iridium, since the two metals behave almost identically in their chemical reactions. Further reference to the old literature$^6$ established the existence of a number of pyridine complexes of iridium. After many weeks of trial and error, the proper conditions were discovered for simultaneous formation of pyridine complexes of rhodium and iridium, and for their extraction into excess pyridine. The method was finally tested using active iridium as a tracer, to prove the results. The method is particularly elegant since it separates in one step from the bulk of uranium in the solution, and from all of the non-platinum group fission products except silver. Subsequent
separation of rhodium from iridium was achieved by adapting the method of Gilchrist and Wichers, precipitating rhodium metal from the boiling sulfate solution by addition of titanous chloride, leaving the iridium in solution.

Based on the use of ethyl acetate to extract platinum, a number of experiments were performed to determine optimum conditions for platinum extraction. Clean extraction of platinum reduced by stannous chloride was attained above four normal acid concentration. Butyl acetate was found preferable to ethyl acetate because of lower water solubility.

The target for each bombardment consisted of a small strip of uranium metal, of natural isotopic composition, 0.04 inches thick and about two inches long. It was mounted on a copper holder, which was inserted on a probe into the vacuum chamber of the cyclotron such that the beam struck the leading edge of the target perpendicular to the metal strip. After bombardment and removal of the target from the copper backing plate, the unbombarde parts were cut off in order not to encumber the chemistry with too much inactive material. It was found desirable to clean the target in nitric acid and keep it in an evacuated dessicator until just prior to bombardment, to prevent formation of the difficultly-soluble oxides of uranium.

After each bombardment the target was allowed to "cool" before it was treated chemically, for from four to eight hours, to permit the very short-lived initial fission products to decay into more stable forms.

Following is a more detailed resume of the scheme of chemistry finally arrived at.

1. **Osmium.** The central portion of the target is cut out and boiled with concentrated HCl to dissolve it and to expel germanium. Iodide and iodate are added to expel iodine. Carriers, in known amount, of the six platinum elements, and of gold are added. A 20% aliquot is withdrawn for later determination of
barium, ruthenium, and rhodium.

The solution is placed in a special all-glass distilling flask having a thistle tube entry and an air entry. Concentrated $\text{HNO}_3$ is added through the thistle tube, and $\text{OsO}_4$ is distilled into $6\text{ N NaOH}$ in an ice bath. Subsequent purification involves precipitation of the sulfides of osmium, followed by redistillation of the sulfides from perchloric acid containing a little $\text{H}_3\text{PO}_4$ to hold back molybdenum. The $6\text{ N NaOH}$ receiving solution is cooled in ice, and carefully neutralized with $\text{HCl}$ to slight excess of acid. Powdered magnesium is added, and after the osmium has been reduced to lower valence state, the solution is boiled for 10-15 minutes, with addition of magnesium and dilute $\text{HCl}$ as needed, until the metallic osmium is completely precipitated. The precipitate is weighed to determine chemical yield. The osmium activity is sufficiently low that the entire sample can be mounted without exceeding normal counting rates.

2. Platinum. The residue from the first distillation of osmium is placed in a beaker, 10 ml of 70% $\text{HClO}_4$ added and the solution boiled to fumes of $\text{HClO}_4$ to expel ruthenium. The solution is diluted, ruthenium carrier, $\text{I}^-$ and $\text{IO}_3^-$ added, and the fuming repeated. One ml of dilute $\text{HCl}$ is added, the solution is diluted to $4\text{ N}$ and extracted twice with butyl acetate to decontaminate from mercury and from gold, the latter being present by independent formation and as the 3.3 day daughter of 31 minute $\text{Pt}^{199}$. The solution is then diluted to approximately 0.5 $\text{N}$ in $\text{H}^+$, dimethylglyoxime added and the palladium precipitate filtered off. Sufficient concentrated $\text{HCl}$ is added to the supernatant to make normality about five. $\text{SnCl}_2$ is added dropwise until the cherry-red of $\text{Pt}^{++)}$ is apparent. Platinum is extracted with three portions of butyl acetate.

The butyl acetate extract of platinum is subsequently purified by two cycles of alternately washing the platinum out of the dimethylglyoxime with $6\text{ N NaOH}$, acidifying, and re-extracting into butyl acetate. Platinum is precipitated as
the sulfide from alkaline solution gradually acidified to 6 N. The sulfide precipitate is dissolved in aqua regia, made alkaline, and two \( \text{La(OH)}_3 \) scavenging precipitations made. Two scavenging precipitations are made from buffered acetate solution with 8-hydroxy quinoline, which serves to remove molybdenum and palladium plus 36 other elements. Platinum is finally isolated as the metal by precipitation from acid solution with powdered magnesium. The precipitate is weighed to determine chemical yield. As with osmium, activity is sufficiently low to permit mounting the entire sample.

3. Palladium. The dimethylglyoxime precipitate from the platinum procedure is dissolved in \( \text{HNO}_3 \) and palladium re-precipitated with dimethylglyoxime. Subsequent purification involves several hydroxide scavengings with lanthanum and zirconium carrier, and several precipitations of silver as \( \text{AgCl} \), following essentially the standard fission product procedure\(^{10} \). Finally the solution is diluted to known volume, transferred to a plastic Lusteroid container (to prevent adsorption of silver daughter activity on glass walls) and wrapped in Parafilm to prevent evaporation. The solution is allowed to stand for 12 to 16 hours until the 3.2 hour \( \text{Ag}^{112} \) daughter of 21 hour \( \text{Pd}^{112} \) has grown to equilibrium. Thereafter at intervals of about a day known aliquots are "milked" of the \( \text{Ag}^{112} \) daughter in order to determine decay of \( \text{Pd}^{112} \).

4. Iridium. The aqueous layer from the platinum extraction must next be fumed with \( \text{HClO}_4 \) to prepare for the pyridine extraction of rhodium and iridium. Since the solution contains organic material (dimethylglyoxime and butyl acetate), care must be taken to avoid an explosion. Following the method of Noyes and Bray\(^11 \) for destroying organic material with \( \text{HClO}_4 \), \( \text{HNO}_3 \) is added to the solution, it is evaporated on a steam bath for one-half hour, then heated gently to fumes of \( \text{HClO}_4 \). An additional 10 ml of \( \text{HClO}_4 \) is added, and the solution is fumed strongly
for several minutes to destroy the chloride complex of rhodium and iridium.
The solution is cooled, diluted to 4 N, and 20 ml of pyridine added. The solu-
tion is then boiled for five minutes, placed in a separatory funnel, and 19 N
NaOH added to separate the free-base pyridine layer (the high concentration of
NaOH is merely to keep the volume low). The dark blue pyridine layer is washed
three times with dilute HCl, the layers being separated each time with 6 N NaOH.
To the pyridine layer is added a few drops of 6 N NaOH, and the pyridine evaporated
off. H₂S is passed into the boiling alkaline solution for several minutes, while
the solution is acidified dropwise with HCl. The precipitated sulfides of iridium
and rhodium are dissolved in a little aqua regia, and fumed strongly with con-
centrated H₂SO₄ to convert completely to the sulfate complex. The solution is
diluted with water twenty to one, then boiled, and rhodium metal precipitated by
adding TiCl₃ dropwise until very slight excess is present. The supernatant
(containing iridium) is diluted to 0.5 N in H⁺, cooled in ice, and titanium is
precipitated with cupferron and filtered off. Palladium and platinum carriers are
added, and palladium is precipitated with dimethylglyoxime and filtered off.
The supernatant solution is then heated to boiling and neutralized to the brom
cresol purple end-point with NaHCO₃ solution, and bromate added to oxidize the
iridium up to plus four. Hydrated IrO₂ is precipitated leaving platinum in solu-
tion. The precipitate is dissolved in a little concentrated HCl, and rhodium
carrier is added. The solution is fumed with H₂SO₄, and the rhodium precipitation
with TiCl₃ is repeated (although the TiCl₃ precipitation of rhodium leaves only
about 0.1% of the rhodium in solution; nevertheless, rhodium activity may be 10⁴
times as high as that of iridium; hence a single separation may still leave
several times as much rhodium activity in solution as iridium activity). Finally,
metallic iridium is precipitated from the HCl solution of the IrO₂ by addition of
powdered magnesium. The iridium is weighed to determine chemical yield, and
mounted. As with osmium and platinum, the entire sample may be mounted without exceeding optimum counting rate.

5. Ruthenium, Rhodium, Barium. These three elements are determined from the 20% aliquot of the sample, which was taken from the original solution prior to the osmium distillation. The only rhodium formed is 36.5 hour Rh$^{105}$, daughter of 4.5 hour Ru$^{105}$. Thus we have two choices in determining the yield of the 105 chain: we may either isolate ruthenium early in order to observe the decay of the 4.5 hour period and ignore the rhodium; or we may allow the sample to "cool" until all the 4.5 hour Ru$^{105}$ has decayed into Rh$^{105}$, and determine yield by isolating the Rh$^{105}$. In practice, both methods were used, with agreement between results, and with no indication of independent formation of Rh$^{105}$. Ruthenium must be isolated in any case, in order to observe 42 day Ru$^{103}$ and 1.0 year Ru$^{106}$.

6. Barium. Barium is determined as a reference for the yield of other elements. The procedure followed is the standard method$^{12}$ of BaCl$_2$·H$_2$O precipitation from concentrated HCl-ether reagent. In order to get a sample of convenient counting rate, a small aliquot ($\sim 10^{-7}$) of the original solution is evaporated and counted to determine level of activity. Since barium activity is found to comprise about 10-15% of total, an aliquot of proper size can be taken to give a barium sample of initial counting rate about 3000 counts per minute.

7. Ruthenium. To the balance of the 20% aliquot, left after removal of the barium sample, is added HNO$_3$, ruthenium carrier, I$^-$ and IO$_3^-$. The solution is boiled to expel osmium and iodine. The solution is placed in a special all-glass distilling flask like that used for the osmium distillation, distilled and purified according to standard procedure$^{12}$. The final solution is diluted to known volume, and 100$\lambda$ evaporated and counted to determine level of activity. An aliquot of proper size to yield an initial count of $\sim 5000$ c/m is then evaporated and mounted. The bulk of the solution is then treated with magnesium to
precipitate ruthenium metal which is weighed for chemical yield, and mounted for later use in making absorption curves.

8. Rhodium. If ruthenium has been isolated sufficiently soon after bombardment to observe the 4.5 hour period, rhodium may be ignored, unless it is desired to have a check on the ruthenium yield. If the 4.5 hour Ru\textsuperscript{105} is not observed, then the yield of the 105 chain is determined from rhodium. Rhodium is isolated from the residue from the ruthenium distillation by a similar but abbreviated method to that used with the bulk of the sample in the iridium procedure. The residue from the ruthenium distillation is fumed strongly with the HClO\textsubscript{4}, diluted and extracted with pyridine. The sulfides are precipitated, and dissolved in aqua regia. Gold, platinum, and palladium carriers are added. Gold is extracted with butyl acetate. Palladium is precipitated with dimethylglyoxime and filtered off. The solution is fumed with H\textsubscript{2}SO\textsubscript{4}, diluted, neutralized to the brom cresol purple end point, and BrO\textsubscript{3}\textsuperscript{-} added to precipitate the hydrated oxides of rhodium and iridium, leaving platinum in solution. The oxides are dissolved in concentrated HCl and diluted to known volume. A small aliquot (∼100 \textmu L) is withdrawn, evaporated and counted to determine level of activity. The activity is almost all due to rhodium, activity of iridium being about 10\textsuperscript{-3} that of rhodium. An aliquot of proper size to give ∼5000 c/m is withdrawn and a known amount of rhodium carrier added to it. It is fumed with H\textsubscript{2}SO\textsubscript{4}, diluted twenty to one, and metallic rhodium precipitated from the boiling solution with TiCl\textsubscript{3}. Subsequently the bulk of the rhodium can be precipitated in the same manner to determine overall chemical yield.
Chapter II

Radioactivities Found in the Platinum Metals

The platinum group activities which should be produced in fission are shown in Fig. 1. As indicated, all are neutron-heavy negative beta-particle emitters. No "light" K-capturing isotopes were expected or found. Following is a presentation of the technique and equipment employed, and a discussion of each isotope observed.

1. General. All samples were counted on standard Geiger-Müller counters having a sealing circuit of 64, using a tube filled to ten centimeters total pressure with argon and containing 5% ethanol as a quenching agent. The counter windows are thin mica sheets with absorption of about three mg/cm². The tubes are mounted on a standard five-shelf frame. Coincidence correction for these tubes averages 1.2% per thousand counts per minute.

To eliminate geometry corrections and allow direct comparison of results, all samples were counted on the third shelf (geometry about 4,7%). To eliminate back-scattering corrections, all samples were mounted identically on one-inch watch glasses resting in a 7/8 inch hole in a standard 2 1/2" x 3 1/2" card, covered with a 2.8 mg/cm² cellophane sheet whose edges were held down by scotch tape. Total absorption of cellophane, air, and counter window averaged 10.3 mg/cm². No correction was made for self-scattering or self-absorption.

Absorption curves were taken where activity permitted, both to determine energy of radiation and to permit correction to zero absorber. When activity was too low to permit accurate absorption curves to be taken, curves published in the literature were used if available. As a last recourse, curves were constructed using a Feather analyzer by comparison to the RaE curve, based on published energy values for the radiation, in order to arrive at an approximate factor with which
Platinum Group Activities Which Should Appear in High-energy Uranium Fission

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Legend:
- isotope
- kind of radiation and energy in Mev.
- Cross-hatched squares indicate stable isotopes.

K-capture
- Long K + $\beta^-$
- Short 43Rh_{105} 36.5 h. -0.65 $\gamma$ 0.65
- 45Rh_{106} 30 s. -4.5 No $\gamma$
- 44Ru_{105} 4.5 h. -1.4 $\gamma$ 0.76
- 44Ru_{106} 1.0 $\gamma$, very soft No $\gamma$

Long-lived isotopes with K-capture and $\beta^-$ decay.

47Ag_{112} 3.2 h. -0.2 $\gamma$ 0.86
46Pd_{109} 13 h. -1.1 No $\gamma$
46Pd_{112} 21 h. -0.2 No $\gamma$

Cross-hatched squares indicate stable isotopes.
to extrapolate to zero absorber.

Wherever possible in making absorption curves, two or three samples were prepared, the second and third having activities roughly 10 times and 100 times the activity of the initial sample. Thus, starting with the first sample having, say, 2000 c/m with no added absorber, as the activity is reduced by adding absorber until it is below the level for good statistical accuracy on short counts (200-500 c/m), the second and third samples are successively counted in place of the first, and their observed activities normalized to that of the first. Thus as in Fig. 3 where the \( \gamma \) "tail" is plotted on the aluminum absorption curve at ca. 40 to 45 counts per minute, this count is actually derived from an observed count of 220 to 250 counts per minute on a more active sample.

Chemical yield was determined in each case by adding a known amount of standardized carrier at the start (ca. 20 mg for each element), and by determining the weight of each element recovered. Osmium, iridium, platinum, ruthenium, and rhodium were determined as the elements by precipitation with magnesium. Palladium was determined as \( \text{Pd}(C_4H_2N_2O_2)_2 \), the palladium compound of dimethylglyoxime. Barium is determined as \( \text{BaCl}_2 \cdot \text{H}_2 \text{O} \).

For each isotope, the decay curve was extrapolated back to zero time, and this \( A_0 \) corrected to 100% chemical yield, total sample, and zero absorber. Setting this corrected \( A_0 = \lambda N_0 \), the number of atoms at zero time was calculated and compared to the number of atoms of \( \text{Ba}^{140} \) at that time to obtain a relative yield. Zero time was taken as the midpoint of the bombardment for bombardments up to one hour, since the shortest half-life recovered was 4.5 hour \( \text{Ru}^{105} \).

For longer bombardments, a median time was computed by the method of D. W. Osborne.\(^{15} \)
2. **Ruthenium Isotopes.** The ruthenium decay curve is shown in Fig. 2. It is complex, consisting of four components, which are fortunately sufficiently different in length of half-lives to permit resolution of the curve.

   a. The longest-lived component is 1.0 year Ru$^{106}$, whose radiation is so soft that it has never been observed. The radiation actually counted is the hard $\beta^-$ from the 30 second Rh$^{106}$ daughter which is in equilibrium with parent Ru$^{106}$. Since time did not permit waiting for the 42 day Ru$^{103}$ to decay into the 1.0 year Ru$^{106}$, the aluminum absorption curve (Figs. 3 and 4) was resolved and the activity of 1.0 year Ru$^{106}$ (again, actually the 30 second Rh$^{106}$ daughter) determined at the time the absorption curve was taken. An ideal one-year line was drawn through this point, and successive subtractions made to arrive at the other three components of the curve.

   b. The next shorter activity is 42 day Ru$^{103}$, which has a very soft $\beta^-$ of $\sim 0.2$ Mev which must be resolved from the aluminum absorption curve (Fig. 4), and a gamma ray of 0.58 Mev as shown in Fig. 3.

   c. The next activity up the scale is the 36.5 hour Rh$^{105}$ daughter of 4.5 hour Ru$^{105}$ (see below).

   d. Finally there is found 4.5 hour Ru$^{105}$, which has a $\beta^-$ of 1.36 Mev and a gamma of 0.72 Mev, as shown in Fig. 5.

3. **Rhodium Isotopes.** The two rhodium isotopes found are both ruthenium daughters, and appear in the ruthenium decay curve as mentioned above. The 36.5 hour Rh$^{105}$ daughter of 4.5 hour Ru$^{105}$ has a $\beta^-$ of 0.66 Mev and a gamma of 0.33 Mev which is 90% converted (literature values$^{14}$; no absorption curve taken). The half-life of 30 second Rh$^{106}$ was not determined, since the activity of this isotope is observed from the equilibrium mixture with its parent 1.0 year Ru$^{106}$, and with the latter's half-life. The energy of the $\beta^-$ of the 30 second Rh$^{106}$ was measured in Fig. 3 as 4.2 Mev.
Figure 2.

Ruthenium Decay Curve
Resolved Into Its Four Components

1.0 year Ru$_{106}$
42 day Ru$_{104}$
36.5 hour Ru$_{105}$
4.5 hour Ru$_{105}$

Legend
- data
- result of 1st subtraction
- result of 2nd subtraction
- result of 3rd subtraction

Activities taken from aluminum absorption curve,

Days, May 1948
4. Palladium Isotopes. The palladium decay curve is complex, being composed of 13 hour Pd$^{109}$ having a $\beta^-$ of 1.1 Mev, 21 hour Pd$^{112}$ having a very soft $\beta^-$ of 0.2 Mev, and its daughter 3.2 hour Ag$^{112}$ having a hard $\beta^-$ of 3.6 Mev. The curve is difficult to resolve, both because the half-lives of the two palladium isotopes are so similar, and also because the weak radiation of Pd$^{112}$ counts with much lower efficiency than either of the others. Resolution of the decay curve was therefore not attempted, but the decay of 21 hour Pd$^{112}$ observed by milking its daughter Ag$^{112}$ at one-day intervals after equilibrium had been established. This resulted in a series of decay curves for 3.2 hour Ag$^{112}$, each of which was extrapolated to time of separation to get a point on the decay curve of 21 hour Pd$^{112}$. Fig. 6 shows a typical decay curve for Pd$^{112}$, with the Ag$^{112}$ decay curves from which it was constructed.

5. Osmium, Iridium, and Platinum. The yield of activity of all three of these elements was very low in comparison with that of the first three platinum elements, and also in comparison with the majority of the other fission products formed in bombardment. This fact necessitated, for iridium and platinum, rigorous and repeated decontamination procedures, as explained in Chapter I, which resulted in low chemical yield as well. The net result of these two factors meant low counting rates with attendant statistical inaccuracies.

After several short bombardments had demonstrated the yields to be expected, two confirmatory bombardments were planned: a short one for ruthenium, rhodium, and palladium, and a long bombardment of greater than five hours for osmium, iridium, and platinum in order to increase activities produced and approach closer to saturation values. In the longer bombardment it was intended to look only for these three elements produced in low yield.
Complex Aluminum and Lead Absorption Curves
Activity includes 42 day Ra105, 1-6 year Ra106, and 20 second Rh106.
Expansion of the Upper Part of Figure 3,
Absorption Curve in Aluminum,
Resolved to Show the Beta-ray of 42 day Ru$^{105}$.
At that time, however, the cyclotron became unavailable for alpha particle bombardments, and remained unavailable for nearly a month (April 5 to May 3), when a two and one-half hour bombardment was finally secured. Since time was growing short, and prospects for a second bombardment appeared remote, all six of the platinum elements were taken out, with essentially the same results as previously secured. These results are reported below.

6. Osmium Isotopes. Two osmium activities were recognized, that of 32 hour Os$^{191}$, and 17 day Os$^{193}$. Both were in very low yield, and no attempt to take absorption curves was made. Typical decay is shown in Fig. 7.

7. Iridium Isotopes. Only one of the expected two iridium isotopes was found, that of 60 day Ir$^{192}$. Decay is shown in Fig. 8. As in the case of osmium, activity was too low to permit absorption curves to be taken. What happened to the 19 hour Ir$^{194}$ activity is unexplained except in one bombardment, in which iridium was separated last and after most of the 19 hour activity would have decayed.

Finding the longer-lived isotope Ir$^{192}$ is of considerable interest since it is shielded by stable isotopes on both sides (Os$^{192}$ and Pt$^{192}$). It therefore must be formed independently, cannot be formed by fission-chain decay. Spallation is indicated as a probable means of formation, and since its yield (of Chapter III) is comparable to the yields of osmium and platinum these likewise may probably be produced by spallation reactions.

7. Platinum Isotopes. The only platinum isotope observed was 18 hour Pt$^{197}$. Activity was low. No absorption curve was taken. The only other platinum isotope probably present would be 31 minute Pt$^{199}$ having too short a half-life for observation using the described means of separation. Its activity might however have been computed by taking out its 3.3 day Au$^{199}$ daughter, and
extrapolating its activity back to zero time, then finding the activity of Pt\textsuperscript{199} from the relationship $A_2/A_1 = \lambda_2/(\lambda_1 - \lambda_2) = T_1/(T_2 - T_1)$ where subscript 1 refers to parent and 2 to daughter. Platinum decay is shown in Fig. 9.

8. Barium Isotopes. In each bombardment, the barium fraction was allowed to stand for several days before barium was isolated. As a result, all the 85 minute Ba\textsuperscript{139} had decayed, and the only activity observed was that of 12.8 day Ba\textsuperscript{140} and its 40 hour La\textsuperscript{140} daughter, which was allowed to grow to equilibrium. Ba activity at zero time was found by two methods as a check: extrapolation of 12.8 day Ba\textsuperscript{140} from time of separation, and by calculation from the extrapolated value of the equilibrium (gross) decay of the Ba\textsuperscript{140} with its 40 hour La\textsuperscript{140} daughter. A typical growth and decay curve is shown in Fig. 10.
Chapter III

Yields of Activities Produced

As stated above, the initial number of atoms of each isotope formed was calculated, and this number compared with the number of atoms of $\text{Ba}^{140}$ formed. Taking the yield of $\text{Ba}^{140}$ as 100%, the relative yields of the other isotopes found are shown in Table I. Results from several bombardments are averaged. No attempt was made to determine accurately the absolute fission yield of $\text{Ba}^{140}$, although previous work\(^1\) indicates that it is of the order of three to four percent in high-energy fission of uranium, as compared to ca. six percent in slow-neutron fission of $\text{U}^{235}$. Estimating the beam strength of the cyclotron as 0.5 microamperes on the average, an approximate cross section for fission for $\text{Ba}^{140}$ is

$$\sigma_f = 2.5 \times 10^{-28} \text{ cm}^2 = 2.5 \times 10^{-4} \text{ barns}.$$  

The yield data are plotted in Fig. 11. Superposed on the plot for comparison, are the slow-neutron fission yield curve\(^16\), and the fission-yield curve obtained by O'Connor for the high-energy fission of uranium with helium ions mixed with deuterons. Also shown are the yields of the heavier elements found by O'Connor in the high-energy deuteron bombardment of uranium. The data here presented give no evidence for a dip in the middle of the fission-yield curve. On the contrary, it appears more likely that the curve has a single smooth maximum in the vicinity of mass 120, i.e., a maximum yield for the symmetrical splitting of the $\text{U}^{238}$ nucleus. This is somewhat analogous to the case of thorium and bismuth fission.

In conclusion, it can be seen from the yield curve that the data collected do so far support the initial hypothesis that what may be assumed to be spallation yield does fall off with decreasing mass number, until at a minimum of around 0.1% at about mass number 180 it again rises in a fission-yield curve probably similar in shape to that for thorium or bismuth.
Figure 9.
Decay Curve of 18 hour $p_{197}$

Time in Days; May 1948
Figure 10.

Growth and Decay of 12.8 day Ba$^{140}$ and Its 40 hour La$^{140}$ Daughter.

Equilibrium mixture extrapolated to midpoint of bombardment.

This extrapolated total $A_0 = A_{Ba} + A_{La}$.

and $A_{La}/A_{Ba} = T_{La}/(T_{La}+T_{Ba}) = 12.8/(12.8+40/24)$

$A_{La} = 1.15 A_{Ba}$, Total $A_0 = 2.15 A_{Ba}$, or $A_{Ba} = 0.66$ (Total $A_0$), which figure should agree with that found below.

Activity at time of separation extrapolated on ideal 12.8 day line to activity = $A_0$ at midpoint of bombardment. This value should agree with the value found above by extrapolation of the equilibrium mixture.

Time in Days; April-May 1948
<table>
<thead>
<tr>
<th>Isotope</th>
<th>Percentage Yield Relative to $\text{Ba}^{140} = 100%$</th>
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<tbody>
<tr>
<td>$\text{Ru}^{103}$</td>
<td>136.6</td>
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<tr>
<td>$\text{Ru}^{105}$</td>
<td>94.4</td>
</tr>
<tr>
<td>$\text{Ru}^{106}$</td>
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<tr>
<td>$\text{Pd}^{112}$</td>
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<tr>
<td>$\text{Pt}^{197}$</td>
<td>0.46</td>
</tr>
</tbody>
</table>
References


15. Osborne, D. W., Calculation of Cross Sections in Bombardments (Report, Argonne National Laboratory, 17 April 1946).
