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FRACTIONATION OF FISSION PRODUCTS AND HEAVY ELEMENTS

BY VOLATILIZATION METHODS

N. D. Erway and O. C. Simpson

August 27, 1946

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FRACTIONATION OF FISSION PRODUCTS AND HEAVY ELEMENTS

BY VOLATILIZATION METHODS

N. D. Erway, O. C. Simpson

1. ABSTRACT

An attempt was made to separate the long lived fission product oxides by volatilization methods. A fractionation crucible having a number of collector plates above the oven section was used. This crucible was heated in vacuo in such a manner that while the oven section was at 2000°C, the top plate was at 1000°C. The collected activities were analyzed both by absorption measurements on the collector plates and by chemical separation of the activities. The results of these fractionations give a fair idea of the relative volatilities of the longer lived fission products and also of the degree of separation of the plutonium oxide or oxides from the fission products.

The separation of several of the heavy elements was also attempted using the above method with the exception that the degree of separation was determined by pulse analysis of the alpha activities. The separation of 96^{242} from 95^{241} was quite effective. One plate in such a fractionation contained 39% of the total 96^{242} activities with only 2% of the more volatile 95^{241} as impurity.

Data obtained by a fractionation of the above type using a charge of 94^{239} metal containing about 0.16% of 95^{241} by activity gave evidence that the 95 isotope could be concentrated considerably on the cooler plates. A second attempt was made at the separation of these elements using a crucible which had a spiral baffle system. This baffle system was to act as a fractionating column and thus give a more complete separation of the sample components than would be achieved with a simple crucible. In this experiment the volatilizing activities were collected on a single plate above the crucible orifice. The plates were changed at intervals and the deposits then analyzed. In this work it was found that under certain optimum conditions of oxidation of the sample the concentration of 95^{241} in the collected activities could be increased by a factor as great as 150 over that in the original sample. These optimum conditions were, however, very difficult to maintain.

2. Introduction

In view of the current interest in the high temperature uranium oxide pile it was considered important to determine the relative volatilities of

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the longer lived fission products. In early work on this type of pile it was suggested that the fission products might be removed from the pile material by volatilization. Thus it was important to know what activities would diffuse from the uranium oxide under normal operating conditions and at what portion of the pile or coolant circulatory system these fission product activities would be concentrated.

Considerable work has been done in the past⁽¹⁾ on the extent of diffusion of fission products from various pile materials (U_3O_8 , solid uranium metal, and uranium metal above its melting point) but there is not much information as to the extent of diffusion from uranium dioxide (UO_2) or of the relative volatilities of the fission product oxides.

Another use of fission product volatility data is in the study of the possibility of separating plutonium from the pile material and accumulated fission products by volatilization. An earlier investigation⁽²⁾ showed that though the separation of plutonium from uranium carbide and fission products by volatilization was not complete there was considerable concentration of the plutonium in the collected material.

The authors do not know of any previous attempt to separate the elements (atomic numbers) 95 from 94, and 96 from 95 by volatilization methods. This work is important not only in that it gives information as to the relative volatilities of these elements but also as to the feasibility of such a separation on a larger scale.

3. Method and Theory

The method used in carrying out a thermal fractionation of the various fission products is outlined below.

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A crucible (Fig. 1) was used in which there were a number of thin tantalum disks which served as condensation plates above the base or oven section. These plates were separated by tantalum spacers and each plate had a small orifice off-center. In assembling the crucible these plates were arranged so that successive orifices were on the opposite sides of the crucible, thus leaving no straight-line path to the top of the crucible.

The crucible was heated by induction in high vacuum in such a manner that while the bottom or oven was at 2000°C , the top was at 1000°C with a linear temperature gradient between these points.

In the case of fission product separation the charge, consisting of a pellet of UO_2 which had been irradiated in the pile at Clinton, was put in the oven section and the crucible was heated in high vacuum for a time sufficiently long to permit countable amounts of fission products to volatilize. These products diffuse upward toward the cool end of the crucible and condense preferentially on the metal plates, the more volatile components on the upper plates, the less volatile nearer the oven.

The plates thus serve both as collector plates and as baffle plates. If the period of heating is sufficiently long this preferential condensation on the various plates could give a reasonably good separation of the products present in the sample. The number of fission products present also influences the completeness of separation, especially if they vary but little in their respective volatilities. This condition, that there be a limited number of fission products in the sample charge, was met by allowing several months to elapse after the samples were taken from the pile so that most of the short-lived products would be out of the sample.

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FIG. 1.. PLATE TYPE FRACTIONATION CRUCIBLE

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After the heating had been discontinued the procedure was to break the vacuum, remove the crucible and disassemble it, thus making the individual plates available for counting analyses.

In fission product separations this analysis was made by comparison of the Geiger-Müller activities as a function of absorber thickness with standard absorption curves for known fission product activities. Further identification and checking was carried out by measuring the half-lives of the activities on certain plates. In several instances where the absorption curves were too complex for positive identification the activities were separated chemically for further identification.

The distribution of alpha activity was also determined to indicate whether the plutonium in the sample was separated from the fission products.

In heavy element separation by this method the counting analysis was somewhat different. In the two separations attempted (96^{242} from 95^{241} , and 95^{241} from 94^{239}) all of the components exhibited alpha activity. Here the method was to determine the total alpha activity on each plate and then to ascertain the percentage of this activity which was due to each of the isotopes present by running pulse analyses on the plates using the method and apparatus described by Ghiorso et al. (3)

A second type of crucible (Fig. 2) was used which had a spiral baffle system instead of the baffle collector plates. This baffle system (a screw having 4 square threads per inch and having a major diameter such that it would just slide into the crucible body) served as a fractionating column, thus giving a more complete separation of the components of a sample than would be possible with a simple crucible. With this type crucible the vapor effusing from the orifice was collected on a quartz plate (1 mm thick) which was

736-7

FIG 2

SPIRAL BAFFLE TYPE FRACTIONATION CRUCIBLE

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supported on tantalum wires over the orifice. The plate was far enough above the crucible top to be sufficiently cold so as to condense the effusing vapors, yet low enough so that practically 100% geometry was achieved with respect to the issuing vapors.

In operation this crucible was heated by induction in high vacuum so that there was a linear gradient between the top and bottom; the gradient was such that, for example, when the top was 1025°C, the bottom was 1225°C. After each exposure the quartz target was removed and was analysed as described above.

The data obtained from the above described experiments give useful qualitative information as to the relative volatilities of the different fission products and heavy isotopes investigated. However, the methods employed do not give sufficient information to permit quantitative calculations of relative volatilities and vapor pressures. For example, in order to calculate the relative volatilities of two alpha active components of a sample at a stated temperature, it is necessary to know not only in what percentage each component is present on the plate, but also the percentage composition of the material in the crucible at the time of the exposure. If one determines the original composition of the sample by pulse analysis, or other means and knows how much of each component has been removed prior to the exposure in question, then one can calculate the average composition of the material in the crucible. But with a crucible of, for instance, the spiral baffle type, it is necessary to know the composition of the material at the orifice, which will be considerably different than the composition in the base. As there is no way of determining this composition the treatment of the data must be of a qualitative nature.

A quantitative determination of the relative volatilities from the data obtained with the plate type crucible is also not justified because of the lack of certain fundamental information.

A vapor pressure experiment of the type described in CN-3223⁽⁴⁾ is now in progress using a mixture of 94^{239} and 95^{241} isotopes. The results of this experiment will be reported soon.

4. Fractionation

4.1 Description of the Apparatus.

(a) The multi-plate fractionation crucible. When a fractionation type crucible of the design shown in Fig. 1 was first considered, it was realized that some method must be developed whereby the plates or disks would be easily removed for counting analysis. Also the leakage between the spacers and the plates should be negligible; i.e. the total area of leakage around each plate should be small as compared to the area of the orifice in the plate.

From previous work in determining the vapor pressure of plutonium and its compounds by effusion methods⁽⁴⁾ it was found that the metal tantalum was very satisfactory for use as a crucible material for several reasons: (1) it has a high melting point, (2) it is easily machined, (3) tantalum does not alloy with plutonium metal nor react chemically with compounds of plutonium, and (4) a very small amount of plutonium metal suffices to give saturation pressures in a tantalum effusion crucible because the metal seems to creep on tantalum thus giving an increased evaporating surface.

However, previous work also has shown that a tantalum-to-tantalum interface shows a very great tendency to cohere at high vacuum and high temperature (though far below the melting point of tantalum) thus making the separation of the parts after the experiment difficult if not impossible. For this reason it was deemed necessary either (1) to separate the tantalum disk from the tantalum spacer by a thin layer of some refractory substance or

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(2) to use different metals for the disk and for the spacers. As there has been some evidence that a tungsten-tantalum interface might prove satisfactory it was decided to make a trial crucible having only two plates, one of two-mil sheet tungsten and one of two-mil sheet tantalum. The base of the crucible was tantalum as were the two spacers. After heating this crucible in vacuo at approximately 1900°C for three hours it was removed and an attempt was made to separate the disks from the spacers. The tungsten sheet was very brittle around the edges and was stuck to the tantalum spacer very firmly. The tantalum disk, as had been expected, was also welded to the spacer and could not be pried loose.

A second experimental crucible was then prepared in which the tantalum spacers and tantalum disks were separated by graphite washers $1/32''$ in thickness. After heating in vacuo at 1900°C for two hours this crucible was examined. The graphite was securely bonded to the tantalum and where the tantalum had touched the graphite it was extremely brittle; so much so in fact that the center of the plate cracked away from the outer edge.

A third crucible was prepared in which thin layers of refractory oxides were used to separate the tantalum disks from the tantalum spacers. A three-plate crucible was used and the three refractory oxides used were beryllia, thorium, and zirconia. The oxide "washers" were formed by spreading a small amount of the oxide paste (thorium in saturated thorium nitrate solution, the others in water) on the spacers and pressing the disk onto the spacer. The crucible was then placed in an oven for partial drying at 110°C for twelve hours, after which it was put into the vacuum system and outgassed at 1900°C for five hours.

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An examination of the crucible showed that the beryllia reacted to quite an extent with the tantalum, the zirconia showed little reaction but was fused to the spacer and disk so that removal was effected only by considerable prying. The thorium reacted very slightly, and the disk came off quite easily.

It was decided that the crucible with tantalum disks and thorium washers would be most satisfactory of those tried. If the presence of the radioactive thorium should be undesirable in a given experiment, zirconia could be used.

(b) The vacuum system. The vacuum system consisted of a Cenco Megavac pump connected by means of a large stopcock (10 mm bore) to a two-liter "ballast" flask, thence through a trap to a two-stage mercury-vapor pump. A second and larger liquid nitrogen cooled mercury vapor trap between the mercury-vapor pump and the experimental section of the apparatus in which the crucible is supported served to freeze out the mercury leaving the pumps in the direction of the apparatus. The pressure in the forevacuum was measured by means of a "Kipp" type McLeod gage attached between the main stop-cock and the "ballast" flask, while the residual gas pressure in the high vacuum region was measured with a VG-1A Distillation Products, Inc. Ion Gage sealed through large tubing to the system at a point between the large liquid nitrogen trap and the cell in which the crucible was to be heated.

The electrical circuit used in conjunction with the VG-1A Ion Gage was a Model 3A Ion Gage Control Circuit obtained from the Instrument Section of the Metallurgical Laboratory, University of Chicago.

The experimental section of the vacuum system consisted of a quartz cell (Fig. 3) constructed from 30 mm quartz tube having a crucible-supporting structure built into the lower end of the tube. This structure was surmounted

736-12

Fig 3

EXPERIMENTAL SECTION OF VACUUM SYSTEM
SHOWING CRUCIBLE AND INDUCTION COIL

736-13

by a quartz table of semi-kinematical design on which the crucible was supported. The table had a conical cavity, a V-groove and a flat similar in most respects to that described by Phipps, Seifert and Simpson.⁽⁴⁾ This type of table gave a very stable and reproducible position to the crucible, which was very necessary in view of the extreme height ($3\frac{1}{4}$ ") of the crucible and the small diameter of the cell. There was only 2.5 mm clearance between the tube walls and the crucible. As shown in Fig. 3 the quartz cell was sealed through a quartz-to-Pyrex graded seal to a pyrex tube which had an optical Pyrex window on its upper end.

(c) The heating arrangements. The crucible was heated by induction with a "Thermonic Generator" Model 520, 10 kw, Series No. 394, made by the Induction Heating Company of New York. The coil for use in the heating by induction was made from $3/16$ " copper tubing and consisted of 13 turns. Preliminary attempts were made to establish a temperature gradient in the crucible such that the bottom of the crucible would run at approximately 2050°C while the top would run at approximately 1000°C by lowering the induction coil, thereby leaving the top of the crucible above the coil. This proved impractical, for with this arrangement the top eight spacers remained below a glowing temperature. Consequently it was decided to raise the coil again so that the bottom of the crucible was just inside the coil and then to extend or stretch the upper seven or eight turns in such a manner that the distance between each turn was progressively greater. A trial run with this type coil showed that while the bottom ran at a maximum temperature of approximately 2050°C , the top of the crucible was at 1105°C and the spacers appeared to be progressively cooler as one observed them from bottom to top.

Because of the small clearance between the crucible and the quartz cell walls and also because of the high temperature at which the crucible was to

736-14

be heated it was necessary to have the quartz tube water cooled. This was done by immersing the quartz tube in a larger glass tube filled with distilled water. This water was rapidly circulated through a water cooled heat-interchanger by means of a small centrifugal pump. It was essential that distilled water be used in this cooling system for if tap water was used the maximum power output of the induction heater was lowered.

A calibrated Leeds and Northrup Optical Pyrometer was used to determine the temperature of the top of the crucible, sighting through the optical window at the top of the cell.

4.2 Procedure in Fractionation Run I.

(a) Fractionation. A fractionation crucible of the design described above and having thoria washers was assembled. It had 17 spacers and 17 tantalum plates. After preliminary drying of the refractory oxide in an oven at 110°C , the crucible was placed in the vacuum system and was slowly heated to a glowing temperature. It was then outgassed for two hours at the highest temperature obtainable (2000°C at bottom). The coil was then raised to a high position and the outgassing was continued. This was repeated until the whole crucible had been outgassed at 2000°C . The vacuum was then broken with helium which had been purified by means of a dip-trap in liquid nitrogen.

The crucible was removed and a sample pellet was placed in the base section by means of the tapered plug shown in Fig. 1. The 1.4415 gm pellet was depleted uranium oxide (UO_3 heated in vacuo to UO_2) which had been irradiated at Clinton for 50,000 K.W. days, 107 days prior to this experiment. The plug was tapped in solidly, the crucible was replaced in the cell and was slowly heated to a maximum temperature (2045°C on bottom - 1061°C on top)

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and was held at this temperature for two hours. The vacuum was again broken with helium and the crucible was removed and disassembled.

(b) Analysis of plate deposits. Each plate was then counted for total alpha and total beta-gamma activity and an attempt was made to identify the fission products which condensed on the various plates. This was done by making aluminum absorption measurements on each plate and comparing the resulting curves with those of Metcalf et al.⁽⁵⁾ A study was also made of the distribution of the beta-gamma activity on the tantalum plates. This distribution was determined by the following procedure. The side of the plate #14, for example, which had been towards the oven end of the crucible was covered with Scotch Tape to hold the deposit intact. The plate was then counted with the taped side down in the chamber and gave a net count of 1005 per minute. The untaped side was then polished with carborundum paper to remove any active deposit and the plate was recounted with the taped side down and gave a net count of 1015 per minute. This result indicated that the active material was deposited only on the side of the plate which was towards the oven. The slight increase in c/m was probably due to thinning the target during the polishing operation.

This distribution is to be expected if one considers that each plate, due to its extreme thinness, will be at very nearly the same temperature throughout. In each small chamber formed by two tantalum plates and one spacer, the upper plate will be at the lowest temperature. Molecules condensing in this chamber will condense at the coldest spot, which is the bottom side of the upper plate. Therefore, the deposit on each plate will be found on its bottom side.

To determine whether a correction for beta activity from the thorium decay products was necessary, an absorption curve was run for a small amount (comparable

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to the amount which would cling to the plate) of thorium which was used in assembling the crucible. The correction was found not to be appreciable and was not applied in plotting the absorption curves.

Due to its alpha emission the small amount of thorium from the refractory washers which adhered to the edges of the plates made the alpha activity analyses very difficult. From this point of view the use of zirconia as a washer material would have been more practical. However, by cutting away the outer edge of some of the plates and thereby removing the thorium it was possible to show that there was considerable alpha activity other than that due to the thorium. Plate #15, for example, showed approximately 700 c/m when treated in this manner.

(c) Results. Table 1 gives the data obtained in this fractionation. The temperatures given are for the most part approximations but are probably correct to $\pm 50^{\circ}\text{C}$. The method of obtaining temperatures was to take the top temperature through the optical window with the optical pyrometer and then the bottom temperature was taken with a pyrometer through the water cooling system and the quartz cell walls. A linear gradient was then assumed and the intermediate temperatures were calculated. The top temperature, when corrected for window absorption and the emissivity of the tantalum, was probably accurate to within five degrees. The bottom temperature had a greater uncertainty and only rough corrections for absorption by the cooling jacket walls, non-optical cell walls and the water itself were made. Evidence to be presented later will show that the error in these temperature approximations was not large.

The total beta activities on each plate are shown graphically in Fig. 4. The beta absorption curves were taken in the standard manner described by

736-17.

TABLE 1
THE FRACTIONATION OF FISSION PRODUCTS
RUN I; PLATE TYPE CRUCIBLE

Plate number	Net beta c/m	Temperature of plate, °C	Identity from absorption curve
1	969	1060	
2	1489	1120	Sr(55d + 30y)
3		1180	
4	5697	1250	"
5	6671	1310	"
6		1370	
7	6496	1440	Sr + Ce
8		1500	
9	3131	1560	"
10	4290	1630	Ce(28d + 300d)
11		1690	
12		1750	
13	79358	1810	"
14		1880	
15	45435	1940	"
16		2000	
17	245416	2060	Ce + ?

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Metcalf et al⁽⁵⁾ and were plotted on three cycle semi-log paper having twelve divisions per inch of abscissa. Thus by superposition of the standard over the unknown absorption curve it was possible to identify the activity on plates which were nearly pure in one activity.

By this method it was found that plates #2-5 (in all cases the plates are numbered from top to bottom, the highest numbered plate having been next to the oven) were nearly pure strontium ($^{89}_{38}\text{Sr}$ + $^{90}_{38}\text{Sr}$). The active material on plates #10-15 was identified as a mixture of 28 day cerium ($^{141}_{58}\text{Ce}$) and 300 $\pm$ 30 day cerium ($^{144}_{58}\text{Ce}$). For example, Fig. 5 shows the standard absorption curve for cerium from Metcalf et al⁽⁵⁾ with the circled points being the absorption data obtained from plate #15. The agreement of these points with those of the standard curve would appear to make the identification quite definite. The identification of strontium by comparison with the corresponding standard absorption curve was equally as good.

The plates between #5 and #10 were assumed to be mixtures of 55 day strontium and the cerium isotopes. The absorption curve for plate #17 was similar to those of #10-16 but there was evidence that some other isotope or isotopes were present.

The identifications listed above were only tentative and further data (half-lives, range, etc.,) would have been required for confirmation. However, in view of the fact that a new run would have to be made before the distribution of the alpha activity could be determined it was decided to see whether or not a better separation of the beta activities could be obtained. A longer heating period with a smaller sample was proposed for the next run which was to be carried out in a crucible having zirconia washers instead of ones made of thoria.

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FIG. 4. BETA DISTRIBUTION ON PLATES IN FRACTIONATION OF FISSION PRODUCTS RUN 1. PLATE #17 WAS NEAREST OVEN SECTION.

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4.3 Procedure in Fractionation Run II.

(a) Fractionation. The crucible was assembled and outgassed at 2000°C for four hours in high vacuum. It was then loaded with a 0.03 gm charge of pulverized UO_2 which had been irradiated with the sample of Run I and the crucible was heated in vacuo at the highest temperature attainable (2094°C oven temperature, 1082°C top temperature) for 200 minutes.

The vacuum was broken with purified helium and the crucible was removed and taken apart. Only one plate, that nearest to the oven, gave any difficulty in this operation and it was removed with a little prying.

(b) Analysis of plate deposits. The plates were individually counted for total alpha and total beta activity and again absorption curves were made for each plate. These data and the half-life data for several of the plates are included in Table 2 along with the probable identity of the predominant fission product on each plate in cases where the curves obtained are sufficiently similar to those of Metcalf et al⁽⁵⁾ to establish this identity.

A further check on the theory that all or at least nearly all of the deposit on a given plate would be on the side which had been towards the oven during the heating was obtained by counting the reverse side of the plate which had the greatest alpha activity. This plate, number 11, had 932 alpha c/m on the side which had been toward the oven, but only 8 c/m on the reverse side, showing quite conclusively that the active material deposits predominately on the side towards the oven.

(c) Results. The graph of Fig. 6 shows the distribution of the alpha and beta activity on the plates of Run II. As can be seen from the sharpness of the beta peaks the separation was much better than that achieved in Run I.

736-22

TABLE 2
THE FRACTIONATION OF FISSION PRODUCTS
RUN II; PLATE TYPE CRUCIBLE

Plate number	Net beta c/m	Net alpha c/m	Temperature of plate, °C	Identity from absorption curves and chemical separation
1	114	10	1080	
2	133	6	1150	
3	161	9	1210	?
4	647	7	1270	Sr(55d + 30y)
5	2305	6	1330	"
6	10795	7	1400	"
7	2659	43	1460	Sr + Ce
8	1162	155	1520	Ce(28d + 300d)
9	2918	396	1590	"
10	5198	567	1650	"
11	5722	932	1710	"
12	3429	650	1780	"
13	2228	293	1840	"
14	2360	306	1900	"
15	4044	337	1960	Ce + Y(57d)
16	19500	309	2030	Ce + Zr(65d) + Y
17	16595	69	2090	Zr + Y + Ce + Ru

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It was first assumed that one of the two peaks in the alpha distribution curve was due to plutonium oxide activity, while the other was due to uranium oxide activity. However, an alpha pulse analysis on plate #11 showed that the plate had both uranium and plutonium alpha activity in approximately equal amounts. The alpha activity on plate #16 was found in a similar manner to be 70% uranium alphas and 30% plutonium alphas.

Such a distribution of activities would result if the curve in Fig. 6 were the summation of one plutonium peak with a maximum at plate #11 and two uranium peaks with maxima at about plates #11 and #16. Therefore the uranium must have been present as two different oxides, having different volatilities, whereas the plutonium oxide and the uranium oxide on plate #11 must have vapor pressures of the same order of magnitude at the temperature of this plate. This is supported by the data on the vapor pressure at equilibrium on tantalum for PuO_2 ⁽⁴⁾ and by earlier data on the vapor pressure of UO_2 .⁽⁶⁾

A study of the absorption curves made possible the identification of several fission products and served in general to confirm the tentative identifications of Run I.

The absorption curve for plate #6 (this plate had the highest Geiger-Müller activity of any in its peak) was nearly identical with the curve given by Metcalf et al⁽⁵⁾ as a standard for a 55 day strontium and 30 year strontium mixture. The standard curve is shown in Fig. 7 with the points being those obtained from plate #6. Half-lives taken with zero and 130 mg/cm² of aluminum gave values of 55.21 and 55.03 days respectively.

The absorption curves for plates #8 to #14 in the second peak were very similar to the standard curve for a 28 day cerium plus 300 $\pm$ 30 day cerium mixture. A half-life determination on plate #11 with 258 mg/cm² of aluminum

736-24

FIG. 6. ALPHA AND BETA DISTRIBUTION ON PLATES IN FRACTIONATION OF FISSION PRODUCTS RUN II. PLATE #17 WAS NEAREST THE OVEN SECTION.

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absorber gave a value of 320 days. Since both 28 and 300 day cerium have beta ranges less than 258 mg Al/cm² this half-life is approximately that of the 17 minute ⁵⁹Pr⁽¹⁴⁴⁾ daughter of 300 day cerium decaying with the half-life of its parent.

A half-life determination on target #11 at zero absorber gave a value of 129 days. Assuming that this plate has no activities other than that due to 28d Ce and 300d Ce one can calculate the ratio of 28d Ce to 300d Ce on plate #11 which will give the above half-life. Such a calculation gave a ratio of 28d Ce to 300d Ce of 0.53 by activity as compared to the ratio of 0.5 for the mixture from which the standard curve was determined. One would expect this 28d Ce to 300d Ce ratio to be identical for all plates containing any of the cerium isotopes at any given time, for this type of experiment will give negligible separation of the isotopes of a given element.

The absorption curve for plate #13 is shown with the standard curve for the cerium mixture in Fig. 8. Also shown in Fig. 8 are the absorption curves for plates #15, 16 and 17. The curve for plate #15 although similar to the standard curve shows evidence of some other activity. In the curve of plate #16 this difference is quite marked and in that of plate #17 the curve is quite different than the standard curve. A study of the progressive change in the character of the curves from #13 to #17 leads one to suspect that the material on these plates is a mixture of the cerium isotopes with some isotope or isotopes having a fairly short beta range and a fairly high-energy gamma component. The presence of the cerium activities in two different peaks on the beta distribution plot (Fig. 6) is probably due to the presence of two oxides of cerium having different volatilities.

Further evidence as to the nature of the impurity was obtained by half-life measurements. With zero absorber plate #16 had a half-life of 72 days.

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A comparison of this value with that for plate #11 (supposed to be a mixture of 28d Ce and 300d Ce) which was 129 days would indicate that the half-life of the unknown activity or activities is less than that for the cerium mixture by a considerable amount.

The character of the curve for plate #17 is quite similar to that for 65 day Zr and all half-life data taken at various times and with different amounts of absorber could be explained as to order of magnitude and direction by assuming that the material on plates #16 and 17 was a mixture of this isotope, its daughter 35 day Cb and the cerium mixture of plate #11.

Quantitative calculations, however, were complicated by several factors and were less satisfactory. For example it was not known to what extent the 65 day Zr was separated from its daughter 35 day Cb in the fractionation process and hence the ratio of these isotopes at any one time was indefinite. Moreover there may have been small amounts of other activities on some of the plates which though not in amounts great enough for identification may have affected the half-life data enough to cause the discrepancies in the attempted calculations.

A chemical separation of the activities on plate #17 showed the presence of cerium, zirconium, yttrium and ruthenium in roughly the following percentages 138 days after the fractionation: Ce 11%, Zr 52%, Y 29% and Ru 8%. The activity on plate #16 is probably a mixture of the same elements with a higher percentage of yttrium than #17.

The fact that a larger number of fission product activities were not found is due to several reasons. The length of time (approximately 150 days) between the irradiation of the sample and the final absorption analysis limits the readily detectable fission products considerably. Moreover the volatility

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of several of the possible products is great enough so that they would not be collected on even the coolest plate while others undoubtedly have volatilities too low to be deposited in detectable quantities at the temperatures used.

5. Separation of Heavy Elements

5.1 Separation of 96^{242} from 95^{241} .

(a) Fractionation. A sample of 95-96 tracer was obtained which had a total alpha activity of about 82,000 c/m and a beta activity due to some impurity of about 11,000 c/m. By pulse analysis it was found that the sample consisted of 88.2% of 96^{242} by activity, with the other alpha activity being due predominantly to 95^{241} . A small amount of 94^{239} was also present but only to the extent of 0.88% of the total alpha activity. The sample also contained approximately 0.2 mg of lanthanum as hydroxide which was used as a carrier in the precipitation of the active components of the sample.

This sample was placed in a crucible of the type described above (Fig. 1, zirconia washers, 17 spacers) which had been thoroughly outgassed. The top of the crucible was at 1080°C during the heating period which lasted for $4\frac{1}{2}$ hours.

(b) Analysis of plate deposits and results. The plates were counted for total alpha activity and a pulse analysis was run on certain of the plates to determine the percentage of 96^{242} present. This data is given in Table 3 along with other pertinent information. Fig. 9 is a graph showing the percentage distribution of the alpha activity on the plates: i.e. the percentage of the total amount on all the plates on any given plate. Also plotted is the percentage of 95 isotope on each of these plates. From this graph it can be seen that the more volatile 95^{241} is concentrated on the upper plates while the 96^{242} is largely on the plates nearest the oven. Although the separation

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of the two isotopes was not complete it was quite effective. For example, plate #16 had 39% of the total 96^{242} activity with an impurity of $2\frac{1}{2}\%$ of 95^{241} by activity. A second fractionation of this material would give nearly complete separation.

A count was made on the reverse side of the plate having the greatest alpha activity (plate #15, 20,129 c/m) and again it was found that the active material was deposited almost entirely on the side of the plate which was towards the oven. In this case the reverse side showed only 79 alpha c/m.

The plates were also analyzed for total beta activity in order to determine whether or not the beta active impurity was separated from the 96^{242} . As can be seen from the data of Table 3, and the graph of Fig. 9, this was not the case. An aluminum absorption curve was run on plate #15 in an attempt to identify the impurity. Comparison of the curve with those of Metcalf et al⁽⁵⁾ showed quite conclusively that this impurity was 65 day zirconium. This was confirmed by determination of the half-life—a value of 65.0 days being obtained with 75.8 mg/cm² of aluminum absorber—and by a chemical separation of the zirconium activity.

5.2 Separation of 95^{241} and 94^{239} using a Plate-Type Crucible.

(a) Fractionation. The fractionation crucible, vacuum system and other apparatus used in the separation of 94^{239} from 95^{241} were identical in every respect to the equipment used in Run II of fission product separation described above.

The charge consisted of a pellet of 4.9 NG metal (plutonium which had been irradiated at Hanford long enough to produce approximately 7% of 94^{240} and corresponding percentages of 94^{241}) which had been separated and reduced to metal 100 days previous to this experiment. The pellet was nearly spherical so its

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TABLE 3
THE SEPARATION OF 96^{242} FROM 95^{241}

PLATE TYPE CRUCIBLE

Plate number	Net alpha c/m	96^{242}	Temperature of Plate, °C.	Net beta c/m 2.47mg/cm^2 Al
1	1.0	Pure 95^{241}	1080	19.6
2	0.9		1140	19.8
3	1.8		1200	23.2
4	1.6		1270	25.4
5	1.6		1330	26.9
6	3.1		1400	29.9
7	29.2		1460	32.2
8	23.0		1520	27.3
9	31.3		1580	24.9
10	62.4	0	1650	29.1
11	148	3	1710	26.6
12	229	21.8	1770	15.3
13	941	51.5	1840	18.3
14	4166	80.6	1900	138
15	20129	90.9	1960	905
16	16971	97.5	2020	1161
17	1082	~100	2090	131

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FIG. 9. GRAPH SHOWING THE PERCENTAGE DISTRIBUTION OF THE ALPHA ACTIVITY ON THE PLATES IN THE SEPARATION OF 96^{242} AND 95^{241} . ALSO PLOTTED IS THE PERCENTAGE OF THE 95 ISOTOPE ON EACH PLATE AND THE DISTRIBUTION OF THE BETA ACTIVITY DUE TO ZIRCONIUM IMPURITY. PLATE #17 WAS NEAREST THE OVEN SECTION.

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weight was approximated by measuring its average diameter with a filar micro-meter and calculating the weight from its volume, assuming a density of 18 g/cm^3 . It was found to weigh approximately 0.148 mg.

According to the available data of Ralph James to be published later, 0.75 mg of 49 NG metal gave approximately 24,200 alpha c/m of 95^{241} after growing in for 27.7 days. Calculations based on these data show that in the 0.148 mg sample which was used in this experiment, there should have been approximately 1.72×10^4 c/m of 95^{241} after growing in for 100 days.

The pellet was removed from its evacuated capillary, was covered with a very small amount of Apiezon N grease and was placed in a thoroughly out-gassed crucible. The purpose of the grease was to keep the sample from oxidizing during the evacuation of the system and the early stages of heating.

The crucible was heated very slowly to permit the Apiezon N grease to volatilize rather than carbonize as it might be expected to do under rapid heating to a high temperature. The vacuum was held to better than 1×10^{-5} mm Hg during this preliminary heating period and was always better than 5×10^{-5} at the high temperature. The crucible was heated over a period of 350 minutes to the temperature at which the fractionation was to be carried out (the top of the crucible was held at 910°C) and the heating was continued for two hours at this temperature.

(b) Analysis of plate deposits and results. The vacuum was then broken with dry helium, the crucible was taken apart, and the plates counted for total alpha activity present on each plate. The plates were then analysed by means of the alpha pulse analyser to determine the percentage of 94^{239} and 95^{241} present on each plate. Table 4 shows the data obtained by these measurements. These data, though not complete show quite definitely that there is a separation

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TABLE 4
THE SEPARATION OF 95^{241} FROM 94^{239}

PLATE TYPE CRUCIBLE

Plate number	Net alpha c/m	% 95^{241} by c/m	Net c/m 95^{241}	Temperature of plate, °C
1	0			910
2	0			970
3	4.0	~100		1030
4	10.1			1090
5	21.0			1140
6	89.1	53	48	1200
7	3077	37	1123	1260
8	8940			1320
9	19640			1380
10	69453			1440
11	77810			1500
12	393706			1560
13	996443			1610
14	927387			1670
15	1137994	0.22	2503	1730
16	951560			1790
17	404165	~0		1840

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of the 94 and 95 isotopes, the more volatile 95^{241} being concentrated on the cooler plates at the top of the crucible while the 94^{239} is concentrated on the plates near the oven. If further pulse analyses had been run on the other plates one would undoubtedly have found the total of 95^{241} counts to be nearly equal to the amount of 95^{241} calculated to be present in the original sample.

Assuming a specific activity of 71,000 c/m per microgram for plutonium one would expect to find a total of approximately 1.05×10^7 alpha c/m on the plates and remaining in the oven. A summation of the data of Table 4 shows a total of 5.3×10^6 c/m detected on the plates, or 50.4% of the total alpha activity calculated to be present. Further heating would have brought the total alpha activity on the plates to a value nearer the total amount calculated to be present but would have done so at the expense of decreasing the concentration of 95^{241} on the plates for the concentration of the more volatile 95^{241} in the oven was probably very low by the time that one half of its total material had been volatilized.

Although the above fractionation is far from complete a second such experiment using the material which was deposited on the upper 10 plates or so of this experiment would give very nearly complete separation of the 95^{241} from the 94^{239} .

5.3 Separation of 95^{241} from 94^{239} using a Spiral Baffle Crucible.

(a) Fractionation. A crucible having a spiral baffle system (Fig. 2) was constructed and was outgassed thoroughly at 2000°C . The induction coil was adjusted so that the top of the crucible ran at 1025°C while the bottom was about 1225°C with a linear gradient between these two points.

The sample was a second pellet of the 49 N.G. metal similar to that used in the separation of 95^{241} from 94^{239} with the plate-type crucible. This sample

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weighed approximately 0.216 mg (calculated from average diameter and density of 18 g/cm³) and since 185 days had elapsed since it was purified, one would expect to find a total of approximately 46,548 alpha c/m in the sample due to 95^{241} .

Before placing the sample in the oven it was covered with a thin film of Apiezon N. A small piece of freshly cut barium metal (slightly larger than the sample) was also covered with an Apiezon N film and was placed in the plug cavity along with the sample. The purpose of the barium metal was to keep the sample in the reduced state, which was desirable because the vapor pressure of the metal is greater than that of the oxide. Much longer exposures would be necessary if the metal were permitted to oxidize.

The length of exposure and the temperature at which the crucible was held were changed after each exposure to obtain conditions under which countable amounts of the components were deposited on the target. The crucible was heated in high vacuum, the vacuum was broken with helium and the quartz target removed while dried helium flowed through the apparatus. Then a new target was placed in position and the system was again evacuated.

Each target, after exposure, was counted for total alpha activity, and the percentage by activity of each isotope on the plate was determined by pulse analysis. From this information it was possible to decide what changes in length of exposure or temperature were necessary to give countable deposits and to some extent what conditions favored the separation of the two isotopes. Thus after exposures 6 through 8 (data given in Table 5) it was evident that while the amount of 95^{241} being deposited per unit time was increasing to some extent the amount of 94^{239} depositing in the same time was increasing very

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TABLE 5
THE SEPARATION OF 95^{241} FROM 94^{239}
SPIRAL BAFFLE TYPE CRUCIBLE

Target	Net alpha c/m	% 95^{241} by c/m	Net 95^{241} c/m	Exposure time min.	Temperature of exp. °C.	Vacuum mm Hg
1	0			120	980	9×10^{-6}
2	0			120	1140	1×10^{-5}
3	0.28	Not anal- yzed		120	1190	5×10^{-6}
4	3.28	Not anal- yzed		120	1250	8×10^{-6}
5	184.7	75	139	120	1310	1×10^{-5}
6	9454	47	4415	380	1310	8×10^{-6}
7	33029	20	6751	380	1310	1×10^{-5}
8	109472	1.9	2069	60	1380	8×10^{-5}
9(a)	47059	6.8	3191	60	1390	1×10^{-5}
10(b)	53968	3.0	1630	120	1390	2.5×10^{-3}
11(b)	41252	2.2	889	120	1370	2.4×10^{-3}
12(c)	40244	2.3	930	120	1360	1×10^{-4}
13	26551	2.1	743	120	1360	1×10^{-4}
14	27925	2.0	545	120	1360	1×10^{-4}

(a) H_2O put into crucible and heated

(b) Air leak used

(c) Air leak closed

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greatly. This would seem to indicate that the sample was either partially oxidized originally or was oxidized in the initial stages of heating in spite of the presence of the barium. The resulting 94^{239} oxide was being reduced to the metal or a lower more volatile oxide either by the barium or by the high-vacuum high-temperature conditions and hence its vapor pressure was increasing rapidly. The 95^{241} was either not oxidized initially or it was being less rapidly reduced. On this assumption it was thought that possibly the ratio of 95^{241} to 94^{239} condensing on the plate could be increased by oxidizing the 94^{239} in the crucible. This was accomplished to some extent by putting a few drops of water into the crucible and heating at less than 100°C in an atmosphere of helium for two hours. As was expected, the rate of effusion of the 95 isotope was not lowered appreciably, while the 94^{239} must have been oxidized somewhat for its rate of effusion was diminished by 50%, even though the temperature of this exposure was slightly higher.

After exposure #9, a method by which the sample could be kept in a slightly oxidizing atmosphere at all times was attempted. A capillary air leak was sealed to the system between the liquid N trap and the experimental cell. This air leak was such that the pressure at the leak was approximately 2×10^{-3} mm Hg. This portion of the experiment did not proceed exactly as was expected for the rate of effusion of both the 95 and 94 isotopes decreased steadily and the percentage of 95^{241} on the targets fell to a value too low for practical purposes. Closing the leak and heating the crucible at the highest temperature and vacuum attainable failed to reduce the sample or increase the 95^{241} percentage. At this point the experiment was discontinued.

(b) Results. The data obtained for the fourteen exposures are given in Table 5. The amount of activity of 95^{241} isotope calculated to be present

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in the 216 mg sample was approximately 4.65×10^4 alpha counts per minute. The total alpha c/m of the sample was approximately 1.53×10^7 c/m assuming a specific activity of 71,000 c/m per microgram of 94^{239} . Thus in the original sample the 95 isotope was present to the extent of 0.30% by activity.

The total number of 95^{241} alpha c/m removed during the run was 2.13×10^4 or 45.8% of the total present in the sample. The total number of alpha c/m (due to 95^{241} and 94^{239}) on the plates was 3.63×10^5 c/m and the percentage by activity of 95^{241} on the plates was thus 5.87%. This indicates a factor of approximately 19 in the concentration of the 95 isotope over that in the original sample.

If the conditions existing in the crucible during exposure 6 could have been maintained throughout the experiment, this factor of separation would have been considerably higher. On the plate of exposure #6, 9.5% of the total amount of 95 isotope calculated to be present in the original sample was present to the extent of 46.7% of the total alpha c/m on the plate. This would indicate a factor of concentration of the 95 isotope over that present in the original sample of 154.

Of course even under such more nearly ideal conditions the decrease in concentration of 95 isotope in the crucible will cause the percentage of 95 on the plates to fall steadily.

A second fractionation of this nature using the material of plates 6 and 8 would give a few thousand counts of very nearly pure 95^{241} .

It is very difficult to correlate the change in the data (Table 5) from exposure to exposure with certain facts which are known about the behavior of plutonium metal and oxides unless certain assumptions are made. For instance, it is fairly well established that once plutonium metal has been oxidized it

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cannot be reduced to the metal state by high-vacuum high-temperature treatment alone.⁽⁴⁾ Yet, the increase in rate of effusion of the 94 isotope in exposures #6, 7, and 8 would seem to indicate that the plutonium had existed as the oxide and was being reduced. And since the sample was supposed to have been held in the reduced state by the barium metal, the presence of plutonium oxides is difficult to account for in any case.

The following explanation, though not necessarily the complete or correct one, seems to fit fairly well the inconsistencies presented by the data. In the first place the barium metal, in spite of the precautions taken, was undoubtedly oxidized to some extent. The barium metal which was not oxidized was probably removed early in the heating and certainly before exposures 6-8, for the crucible was held above the boiling point (1146°C) of barium for eight hours before exposure #6 was started.

The fact that deposits of barium were not observed on the quartz plates may well be due to the high temperatures of these quartz plates during the exposures. No ready method of estimating this temperature was possible, but when the top of the crucible was at $1100-1200^{\circ}\text{C}$, the plate may well have been $800-900^{\circ}\text{C}$. Thus the metallic barium would volatilize from the plates and condense on the tube walls in quantities too small to be evident.

However, the barium oxide remaining in the crucible could, according calculations based on available vapor pressure data⁽⁷⁾, have been present in the crucible through the exposure in question. This was confirmed in that visible deposits of barium oxide were on plates 4-7.

If then the decomposition pressure of this residual barium oxide were greater than that of one of the plutonium oxides one might expect that the

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plutonium metal would at least be partially oxidized and the excess barium metal be removed from the crucible by volatilization. Or as is quite conceivable, the tantalum crucible, being as massive as it was, may have^{been} insufficiently well outgassed so that the residual oxygen pressure would be high enough to oxidize the metal at the high temperatures used. In any case if an oxide layer were formed on the plutonium metal sample the sample may not have melted at its true melting point ($627 \pm 5^{\circ}\text{C}$)⁽⁸⁾ but may have behaved in the manner described by Z. Jasaitis.⁽⁹⁾ In this earlier work it was noted that plutonium metal samples were sometimes heated in vacuo on tantalum to temperatures as high as 1350°C before melting. This anomalous effect was attributed to an oxide layer formation on the metal which had sufficient tenacity to hold the metal together at temperatures far above the true melting point. That explanation was substantiated by having an X-ray analysis made on the outer portion of the sample. This crust was reported to be an oxide of plutonium, probably PuO .

This effect has since been repeatedly observed^(10,11) so it seems reasonable that the same situation may have occurred in this experiment. If this was the case the pellet may have retained its shape through exposure #7 with only a very small amount of the oxide volatilizing. Due to the partial pressure of oxygen from the barium oxide the metal that succeeded in effusing through the oxide crust would have been oxidized almost completely. During exposure #7 the barium oxide was depleted and hence ceased to oxidize the volatilizing metal. The temperature was then raised considerably for exposure #8 and the vapor pressure of the metal became sufficient to break the oxide crust and spread over the crucible bottom, giving a much larger and cleaner evaporating surface of metal. This would cause the crucible to become more nearly saturated with the vapor and would give rise to an immediate and large increase in the amount of plutonium being deposited on the target per unit time.

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The fact that the rate of effusion of the 95 metal did not change appreciably during exposures 7 and 8 and only doubled at the melting of the sample pellet can be explained by assuming that the 95 metal is less easily oxidized and was being volatilized as metal throughout the experiment. Moreover, the percentage of 95 isotope in the crucible was being lowered rapidly so one would not expect a very great increase in rate of effusion with the melting of the sample.

The steady decrease in the amounts of both 94 and 95 isotope deposited after exposure #10 can be explained both by the re-oxidation of the metal by the water added and the effects of the air leak, and by assuming that the active material in the crucible was being depleted. This latter supposition is warranted, for during the addition of the water before exposure #9 it was suspected that the crucible had a slight leak around the loading plug. This suspicion was verified by measuring the activity on the supporting structure after the crucible was removed. Although this leak was very small as compared to the orifice area, the temperature of the lower end of the crucible was two hundred degrees or more above that of the orifice so one would expect that some of the charge and especially the more volatile 95 metal may have effused out in appreciable amounts.

The decrease in rate of effusion after the oxidation with water and the air leak did not, however, bring the activity on the plates down to as low as it had been before the spreading out of the sample. It is quite possible, however, that during these earlier exposures the crucible was unsaturated because of the large ratio of orifice area to sample area and the fact that much of the sample may have been buried in barium oxide. The larger area for evaporation after the metal was spread out and oxidized, combined with

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the fact that the temperature of the crucible was higher would then account for this discrepancy.

There may be other methods of explaining most of the above inconsistencies but without further investigation it seems futile to make any definite assertion as to their causes. The authors hope to have much better data on the relative volatilities of plutonium and "americium" (95) in the very near future.

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