due to a preferential absorption of the negative mesons by the lead, and the present results are in accord with this hypothesis.

If the assumption that the soft component is symmetrical be accepted as true, then the asymmetry of the hard component arriving at the apparatus should be given by calculation from the "no lead" measurements, and in view of the evidence that an absorber influences the results we are of the opinion that this procedure gives a more reliable value than that obtained directly. It is to be noted that this would give values falling on a curve slightly above the broken curve in the figure, and appreciably below the values predicted by Johnson's theory (the solid curve) for a lower energy limit of $2\times10^8$ ev.

From the data at present available it appears that Johnson's theory gives the form of the asymmetry *vs.* zenith angle curve but the values are slightly large. Further experiments are being conducted, and it may be possible to suggest some modification of the theory when more results are available.

ACKNOWLEDGMENTS

The authors have pleasure in expressing their sincere thanks to Professor A. L. McAulay for his ready advice and for stimulating discussions during the course of the work. We are very grateful to the Electrolytic Zinc Company, Ridson, Tasmania, for a grant for apparatus and to the C.S.I.R. for a grant which enabled one of us (D.W.P.B.) to take part in this research. It is a pleasure to acknowledge the encouragement received from Dr. T. H. Johnson when we first proposed to conduct these experiments. We would also like to thank Mr. R. L. Propsting for his enthusiastic help in the construction of the apparatus.

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# A New Radioactive Series—The Protactinium Series*

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A new series of radioactive isotopes has been produced by deuteron and helium ion bombardments of thorium. The identity and the properties of the five members of this series, which is a branch of the uranium-radium series and joins it at RaC′, are shown to be the following:

$$\mathrm{Pa}^{230}\xrightarrow[17d]{\beta^-}\mathrm{U}^{230}\xrightarrow[20.8d]{\alpha(5.86\ \mathrm{Mev})}\mathrm{Th}^{226}\xrightarrow[30.9m]{\alpha(6.3\ \mathrm{Mev})}$$

$$\mathrm{Ra}^{222}\xrightarrow[38s]{\alpha(6.5\ \mathrm{Mev})}\mathrm{Em}^{218}\xrightarrow[0.019s]{\alpha(7.1\ \mathrm{Mev})}\mathrm{RaC}'\xrightarrow[1.5\times10^{-4}s]{\alpha}$$

$$\mathrm{RaD}\xrightarrow[22y]{\beta^-}\mathrm{RaE}\xrightarrow[5d]{\beta^-}\mathrm{RaF}\xrightarrow[140d]{\alpha}\mathrm{RaG\ (stable)}$$

A simple electronic device was developed to measure the short half-life of $\mathrm{Em}^{218}$.

## 1. INTRODUCTION

THREE radioactive series, the thorium series, the uranium-radium series, and the actinium series, are known to occur in nature. These series are often called the $4n$, the $4n+2$, and the $4n+3$ series, respectively, after the general formula for the mass numbers of the isotopes in each of the three series. For a long time a fourth series, the $4n+1$, was predicted and sought for, but it was not until the advent of artificial transmutation that such a series was

* This report is based on work done in 1946 under the auspices of the Manhattan District at the Metallurgical Laboratory, the University of Chicago.

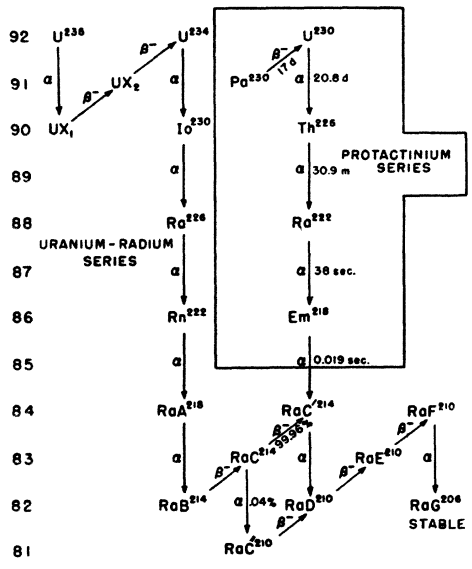


FIG. 1. Relation of the protactinium series to the uranium-radium series.

found.[1,2] This artificially produced decay chain is called the neptunium series after $Np^{237}$, the long-lived parent of the series.

From time to time individual, synthetically-made isotopes have been added to these main series as extensions or as one-member branches. The existence of a branch of the uranium-radium series consisting not of one but of a whole series of isotopes is reported in this paper. It is proposed to call this new series of synthetically-made isotopes, the protactinium series, after $Pa^{230}$, its first member.

In Fig. 1 the relation of the protactinium series to the uranium-radium series is presented.

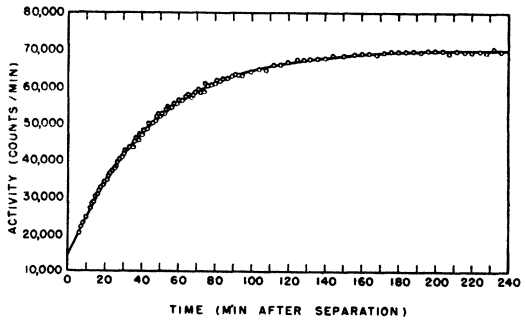


FIG. 2. Growth of alpha-activity in a sample of $U^{230}$.

[1] F. Hagemann, L. I. Katzin, M. H. Studier, A. Ghiorso and G. T. Seaborg, Phys. Rev. **72**, 252 (1947).

[2] A. C. English, T. E. Cranshaw, P. Demers, I. A. Harvey, E. P. Hincks, J. V. Jelley and A. N. May, Phys. Rev. **72**, 253 (1947).

## 2. THE PRODUCTION OF THE PROTACTINIUM SERIES

The protactinium series was originally produced by a $d,4n$ reaction in a 100-microampere-hour bombardment of thorium metal with 19 Mev deuterons and by an $\alpha,p5n$ reaction (with probably some contribution from an $\alpha,6n$ reaction) in a 30-microampere-hour bombardment of thorium metal with 38 Mev helium ions. The bombardments were carried out in the 60-inch cyclotron at the University of California by Dr. J. G. Hamilton, T. Putnam and the cyclotron crew. Their assistance is gratefully acknowledged.

The type of nuclear reaction in which more than two particles are ejected had been encountered in only one or two instances at the time that these experiments were begun. The $\alpha,p5n$ and $\alpha,6n$ reactions were probably observed for the first time in this work.

## 3. THE DISCOVERY OF THE PROTACTINIUM SERIES

The nuclear reactions which produced the protactinium series in the bombardments of thorium metal were accompanied by numerous other reactions such as $d,n$; $d,p$; $\alpha,n$; $\alpha,p$, etc. In addition, some deuteron and helium ion induced fission of the $Th^{232}$ occurred. The resulting mixture of radioactive isotopes complicated the determination of the decay scheme of the new series.

One of the first clues, leading to the discovery of the new series was the decay of alpha-activity in the pure uranium fraction of the thorium-helium ion bombardment which indicated the presence of an isotope of uranium with a half-life of approximately three weeks. The uranium isotopes which could conceivably be present were those of atomic mass 235 or less; $U^{235}$, $U^{234}$ and $U^{233}$ are all so long-lived ($10^5$ years or more) that they would have shown no change in counting rate. A sample of the 70-year[3] $U^{232}$ should show an increase of about ten percent per month from the growth of alpha-emitting daughters. Hence it was necessary to conclude that a new uranium isotope of mass 231 or less was present.

[3] R. A. James, A. E. Florin, H. H. Hopkins and A. Ghiorso, Plutonium Project Record **14B**, No. 22.8; Report CC-3860.

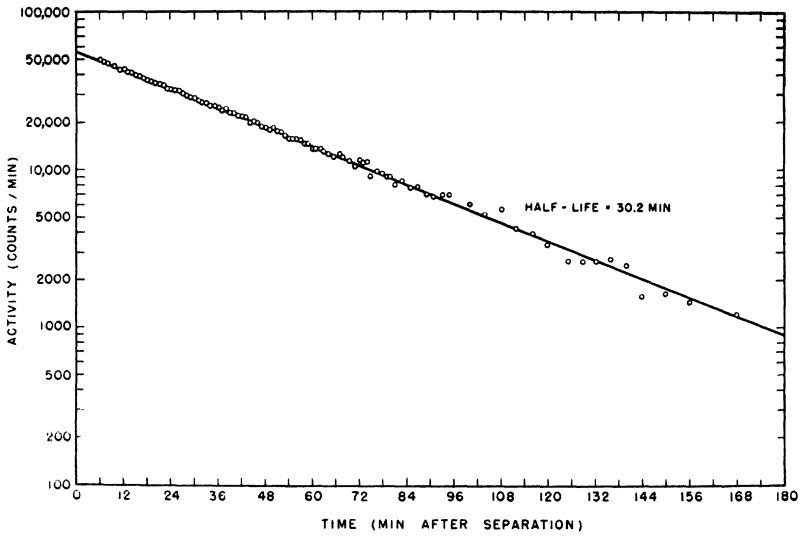


FIG. 3. Complement of the growth curve of a sample of $U^{230}$.

An examination of the alpha-particle energies of the activity in the uranium fraction a few hours after separation revealed the presence of five equally-abundant activities with energies greater than that of the $U^{232}$ alpha-particle. The growth of the daughters of the uranium isotopes of mass 232 or higher would not be appreciable within a few hours. This is further evidence for a uranium isotope of low mass followed by four daughters with relatively short half-lives.

The rate of growth of daughters was determined by observing the increase of alpha-activity in a uranium sample very soon after purification. By extracting the uranium into ether and evaporating the ether directly on a platinum plate, it was possible to get the sample to an alpha-counter within six minutes after purification. There was an initial rapid growth of alpha-activity which had essentially reached its maximum after four hours, at a level of activity which was five times the activity present at the time of purification. (See Fig. 2.) This is further evidence for the presence of four alpha-emitting daughters of the new uranium isotope. A decay curve which is complementary to the exponential growth of Fig. 2 may be obtained by plotting on semi-logarithmic graph paper the difference between the equilibrium counting rate and the observed counting rate as a function of time. A curve obtained in this manner gave a straight line with a half-life period of one-half hour. (See Fig. 3.) The fact that four alpha-emitters grew in with this half-life indicates that the first daughter of the uranium, a thorium isotope, has the half-hour period and that the others have considerably shorter half-lives. After equilibrium had been reached, the total activity decayed with a half-life of three weeks.

When it was found that the uranium fraction from a deuteron bombardment of thorium also contained five distinct alpha-activities and decayed with the same half-life as the uranium fraction from the helium ion bombardment of thorium, the mass number 231 was eliminated from consideration. The fact that a uranium isotope could not be formed directly by a deuteron bombardment of thorium, but could only result from the beta-decay of a protactinium isotope, together with the fact that $Pa^{231}$ is a long-lived alpha-emitter, justifies this elimination. Therefore the isotope was assumed to be $U^{230}$.

The next logical step, the search for a uranium daughter in the protactinium fraction, was carried out by isolating a protactinium fraction free from uranium and subsequently extracting a uranium fraction from this after a growth period of several days. The five new alpha-activities and a decay of total activity were observed in the uranium fraction so isolated. This experiment was repeated with the same results on the protactinium fraction of the thorium-helium ion bombardment. This repetition was important since the uranium isotope

in the helium ion bombardment could have been formed directly by an $\alpha,6n$ reaction or indirectly by an $\alpha,p5n$ reaction producing $Pa^{230}$, followed by a beta decay of $Pa^{230}$. The evidence indicates that an $\alpha,p5n$ reaction does occur. It does not eliminate the possibility of some $U^{230}$ being formed directly by an $\alpha,6n$ reaction. It was calculated that more $U^{230}$ was isolated in the first uranium separation from the thorium-helium ion target than could be accounted for by $Pa^{230}$ beta-decay on the basis of the later growth of $U^{230}$ into the protactinium fraction. This was interpreted as evidence for the occurrence of an $\alpha,6n$ reaction. Four alpha-disintegrations starting with $U^{230}$ would produce $_{84}Po^{214}$ which is RaC′ of the uranium series. (See Fig. 1.) The energy of the most energetic of the five unknown alpha-emitters is indeed identical within experimental error with the 7.68 Mev energy of the RaC′ alpha-particle. (See Section 4.8.) If the original protactinium isotope were $Pa^{229}$ or $Pa^{228}$, the resulting polonium isotopes would be $Po^{213}$ or ThC′ with energies (8.28 Mev and 8.776 Mev respectively) much greater than that of RaC′. Furthermore, on theoretical grounds, the $d,5n$, the $d,6n$, the $\alpha,p6n$ and the $\alpha,p7n$ reactions required to produce $Pa^{229}$ and $Pa^{228}$ are probably not energetically possible with 19 Mev deuterons and 38 Mev helium ions.

## 4. EXPERIMENTAL PROCEDURE

4.1 *Milling and Chemical Treatment.*—The cyclotron targets in these bombardments consisted of thorium metal disks, 8 cm in diameter and 5 mm thick. The 25 by 65 mm rectangular area actually struck by the beam was located by inspection and by radiographs. The top layer of metal was removed from this area by clamping the disk in a special covered mount and milling off 0.030 inch. The millings, which amounted to 15 g of thorium metal, were dissolved and put through an extensive chemical separation scheme to isolate pure protactinium and uranium fractions free from fission products, decay products and other radioactive and inert impurities.

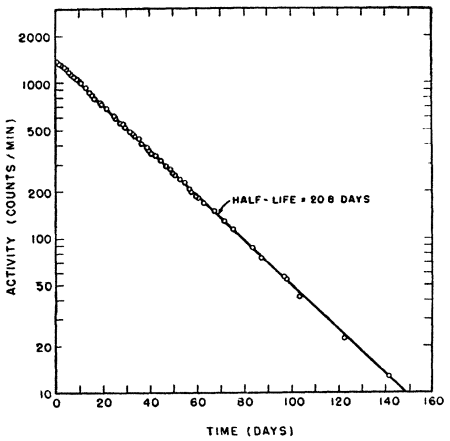


FIG. 4. Determination of the half-life of $U^{230}$ from the decay of alpha-activity of a sample of $U^{230}$ in equilibrium with its daughters.

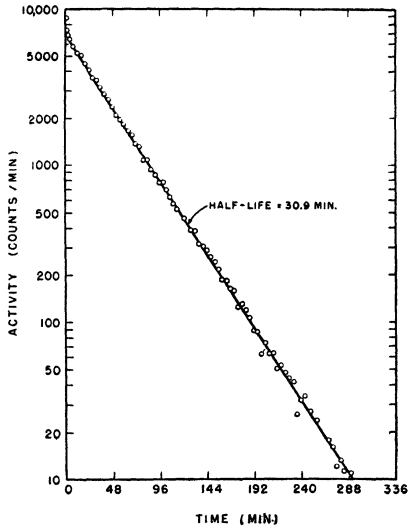


FIG. 5. Determination of the half-life of $Th^{226}$ by the decay of alpha-activity from the recoil atoms of $U^{230}$.

Samples of these fractions were used in the studies described below.

4.2 *Determination of the half-life of* $U^{230}$.—Samples of $U^{230}$ were prepared by extracting the uranium which had grown into an originally pure protactinium fraction. Several protactinium isotopes besides $Pa^{230}$ were present. $Pa^{233}$, $Pa^{232}$ and $Pa^{231}$ were formed in the deuteron bombardment by $d,n$, $d,2n$ and $d,3n$ reactions, respectively. (In the helium ion bombardment these isotopes were formed by $\alpha,p2n$, $\alpha,p3n$ and $\alpha,p4n$ reactions respectively. The $\alpha,p4n$ reaction was not specifically observed.) $Pa^{231}$ offered no complications because it is a long-lived alpha-emitter. The $U^{233}$ daughter of $Pa^{233}$ is so long-lived that the presence of $Pa^{233}$ does not interfere with the preparation of a $U^{230}$ sample. However, it does complicate the characterization of the beta-

radiation of $Pa^{230}$ since the half-life of $Pa^{233}$ (27.4 days)[4] is close to that of $Pa^{230}$ (17 days). This necessitated the indirect determination of this half-life as described in Section 4.5. The presence of the 1.33 day[5] $Pa^{232}$, which decays to a 70-year $U^{232}$ daughter, made impossible the isolation of a uranium sample suitable for the half-life determination of $U^{230}$ as long as it was present. This difficulty was eliminated by letting the protactinium activity decay 16 days. By this time the $Pa^{232}$ had decayed to 0.1 percent of its original value while the $Pa^{230}$ had decayed by only about half. At the end of this period the uranium was completely separated and discarded. Pure $U^{230}$ was permitted to grow in for a period of a week and then isolated from the protactinium. Samples of this uranium were mounted on platinum counting disks for following changes in alpha activity.

It was early discovered that samples of $U^{230}$ would leave a residual activity in the counter upon removal. This was first thought to be due to an emanation, but experiments to be described below (Section 4.4) demonstrated that it was due to recoiling nuclei from the energetic disintegrations of $U^{230}$ and its alpha-emitting daughters. This behavior was later found to be very useful (see Sections 4.3 and 4.6), but in counting samples for the decay of $U^{230}$ it was necessary to consider the danger of erratic results due to loss of recoil atoms. This difficulty was resolved in two ways: (1) by permitting the sample to come to equilibrium in the counter before making a count, and (2) by covering the sample with a light film of zapon to prevent the recoil atoms from excaping.

A $U^{230}$ decay curve is shown in Fig. 4. A "least squares" treatment of the data yielded 20.8 days for the half-life.

4.3 *Identification of* $Th^{226}$ *and the determination of its half-life.*—It was postulated above from the characteristic growth of alpha-activity of a sample of $U^{230}$ (Figs. 2 and 3) that the first daughter of $U^{230}$, $Th^{226}$, has a half-life of approximately one half-hour. This was shown to be correct by a chemical isolation of the thorium.

The thorium was isolated by coprecipitation with zirconium iodate which is known to be a specific carrier for thorium. Zirconium nitrate was added to a dilute nitric acid solution of $U^{230}$ in equilibrium with its daughters, and zirconium iodate was precipitated by the addition of iodic acid. The precipitate was dissolved with sulfur dioxide, reprecipitated with iodic acid, and mounted on a counting disk. The rate of decay of alpha-activity was determined, and a half-life of 31 minutes was found.

With the $Th^{226}$ positively identified, a more accurate value of the half-life was determined by following the decay of recoil activity left in an alpha-counter after removal of an active sample of $U^{230}$ (100,000 alpha-counts per minute) which had been in the counter for an hour. An initial activity of over 8000 counts per minute decayed to the background of the counter. (Any contribution by "aggregate" recoil to the recoil activity must be negligible since no long-lived

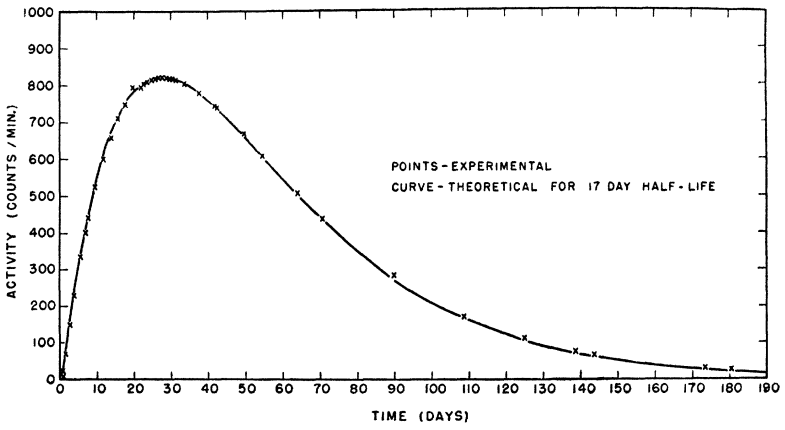


FIG. 6. Determination of the half-life of $Pa^{230}$ from the growth and decay of daughter alpha-activity.

[4] A. V. Grosse, E. T. Booth and J. R. Dunning, Phys. Rev. **59**, 322 (1941).
[5] A. H. Jaffey and E. K. Hyde, Plutonium Project Record **17B**, No. 9.20; Report ANL-4102.

component due to $U^{230}$ was found in the recoil atom decay curves.) (See Fig. 5.) The small amount of short-lived activity indicated by the beginning of the curve was later shown to be due to $Ra^{222}$. The half-life obtained for $Th^{226}$ is 30.9 minutes. This is slightly greater than the value obtained from analysis of the growth curve (Fig. 3) because in the latter case the approximation was made that the $U^{230}$ activity was constant during the time of growth.

4.4 *The emanation isotope.*—It was mentioned above that the 30.9 minute contamination of the alpha-counter by samples of $U^{230}$ was thought at first to be due to the emanation isotope, $_{86}Em^{218}$. However, this was shown not to be the case even before the 30.9 minute activity was identified with $Th^{226}$ by the following evidence: (1) Replacing the air in the contaminated chamber did not remove the contamination. (2) Placing in the counter an active $U^{230}$ sample covered with two baffle plates, which permitted the escape of an active gas by an indirect route but offered no direct path for the escape of a recoil atom, did not result in any measurable contamination of the chamber.

These experiments demonstrated that the contamination was due to recoiling nuclei. They do not disprove the existence of an emanation but merely indicate that if it exists it must be short-lived. To set an upper limit on the half-life of the emanation a series of experiments was performed by P. Fineman, B. Weissbourd and T. P. Kohman. They arranged to flush the gas emerging from a boiling solution of $U^{230}$ directly into a special alpha-counting chamber. This was done without detecting any counts above background. From their work they set an upper limit of one second for the half-life of $Em^{218}$. This half-life was later determined to be 0.019 sec. by an electronic coincidence method. (See Section 4.7.)

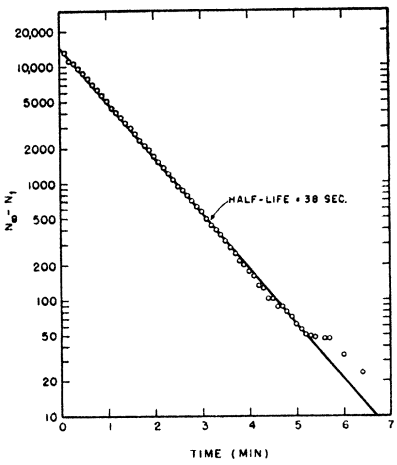


FIG. 7. Determination of the half-life of $Ra^{222}$ by the decay of recoil activity from a $Th^{226}$ sample. $N_\infty$ = total counts observed on complete decay. $N_t$ = total counts observed to any given time.

This short half-life excludes the possibility of chemical identification of $Em^{218}$ and the evidence for the existence of the isotope is indirect, resting on the over-all evidence for the series and particularly on the identification of its RaC′ daughter by energy measurements.

4.5 *Determination of the half-life of* $Pa^{230}$.—The direct determination of the half-life of $Pa^{230}$ using the protactinium fractions of the thorium bombardments was rendered difficult because of the presence of $Pa^{233}$. Hence indirect methods were used.

A preliminary value was obtained by extracting and measuring the $U^{230}$ produced in equal periods of time from a sample of protactinium. In this manner a crude half-life value of about two weeks was obtained.

A more accurate value was obtained by mounting a sample of $Pa^{230}$ on a platinum counting disk and following the rate of change of the alpha-activity of the sample. Such a growth and decay curve is shown in Fig. 6. By substituting the half-life of $U^{230}$ determined above into the standard growth equation,

$$N_2 = N_{01}(\lambda_1/\lambda_2 - \lambda_1)(e^{-\lambda_1 t} - e^{-\lambda_2 t}),$$

where $N_2$ = number of $U^{230}$ atoms, $N_{01}$ = number of $Pa^{230}$ atoms initially present, $t$ = time, $\lambda_1$ = disintegration constant of $Pa^{230}$, and $\lambda_2$ = disintegration constant of $U^{230}$, theoretical curves for various half-lives of $Pa^{230}$ can be calculated and compared with the experimental curves. In this way using three separate samples a value of $17.0 \pm 0.5$ days was determined for the half-life of $Pa^{230}$.

4.6 *Identification of* $Ra^{222}$ *and the determination of its half-life.*—From the failure to observe any alpha-activity in a radium fraction, isolated rapidly by chemical precipitation of a barium chloride carrier from an active solution of $U^{230}$

in equilibrium with its daughters, an upper limit of one minute was set on the half-life of the radium isotope. Although attempts to identify the $Ra^{222}$ chemically were unsuccessful, an alpha-emitting $Th^{226}$ is proof of its existence. It will be recalled that the decay of recoil activity from a $U^{230}$ sample (Fig. 5) indicated the presence of a short lived component with a half-life of the order of half a minute. Since both $Em^{218}$ and RaC′ have half-lives much shorter than a second, this activity must be due to $Ra^{222}$. It was also noted that a sample of $Th^{226}$ leaves a recoil activity in an alpha-counting chamber after the sample is removed, and the decay of this activity was used to determine the radium half-life.

To get a maximum amount of recoil from a $Th^{226}$ sample, thorium was isolated with a minimum of carrier from a solution containing 100,000 counts per minute of $U^{230}$ and mounted on a platinum disk. By inserting this disk in a standard alpha-counting chamber for a few minutes it was possible, because of the recoil phenomenon, to introduce several thousand counts per minute of $Ra^{222}$ into the chamber. The $Th^{226}$ was then removed and the recoil activity was measured at intervals of 0.1 minute until decay was complete.

It was necessary to measure the activity at such short intervals of time because of the short half-life of $Ra^{222}$, and as a result the statistical fluctuations were rather severe. An integral decay curve was plotted to minimize the scatter of points. The error, which would result from the use of the usual differential curve because the duration of each measurement was an appreciable fraction of a half-life, was also eliminated. By subtracting $N_t$, the total number of counts up to a given time, from $N_\infty$, the total number of counts observed on complete decay, and plotting the difference, $N_\infty - N_t$, on a logarithmic scale as a linear function of time, a straight line was obtained, the slope of which determined the half-life. Such a curve is shown in Fig. 7. The half-life of $Ra^{222}$ based on four such determinations is 38.0 seconds.

4.7 *The measurement of the half-life of* $Em^{218}$.— A simple electronic circuit which can be used to measure the time intervals between the emission of successive alpha-particles was devised to measure the half-life of the emanation isotope.

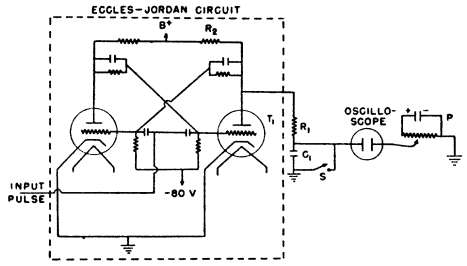


FIG. 8. Diagram of electronic circuit used to measure short time intervals between successive pulses.

In the decay chain

$$A \xrightarrow{\alpha} B \xrightarrow{\alpha} C$$

let $A$ be the (long-lived) parent of $B$, the half-life of which is to be measured.The instant that the atom $A$ emits an alpha-particle the atom $B$ is formed, and the time elapsing until $B$ emits an alpha particle is the lifetime of $B$. An average of a number of measured time intervals between the successive disintegrations of $A$ and $B$ gives the mean life of $B$ which is equal to $1/\lambda$ where $\lambda$ is the decay constant of $B$. The half-life is then given by $0.693/\lambda$.

A schematic diagram of the circuit which was used for the measurement of the short time intervals between pulses is shown in Fig. 8. A resistor, $R_1$, in series with a condenser, $C_1$, was connected between the plate and cathode of one of the tubes, $T_1$, in the first trigger pair in the scaling circuit of a standard alpha-counter. The horizontal deflecting plates of an oscilloscope were connected across the condenser, $C_1$, through a potentiometer, $P$, which was included in the circuit to control the horizontal position of the fluorescent spot on the oscilloscope screen. The scaling pair was of the Eccles-Jordan type[6] with 6C5 triode tubes. Such a trigger pair ex-exhibits two stable states in which one tube is conducting while the other is non-conducting. A pulse fed to the grids of the pair will cause the tubes to reverse from the one stable state to the other in a few microseconds. Because of the potential drop across the plate resistor, the plate voltage of a tube in the conducting state is much lower than that in the non-conducting state. Whenever a pulse is fed to the grids of the scaling

[6] W. H. Eccles and F. W. Jordan, Radio Rev. 1, 143 (1919).

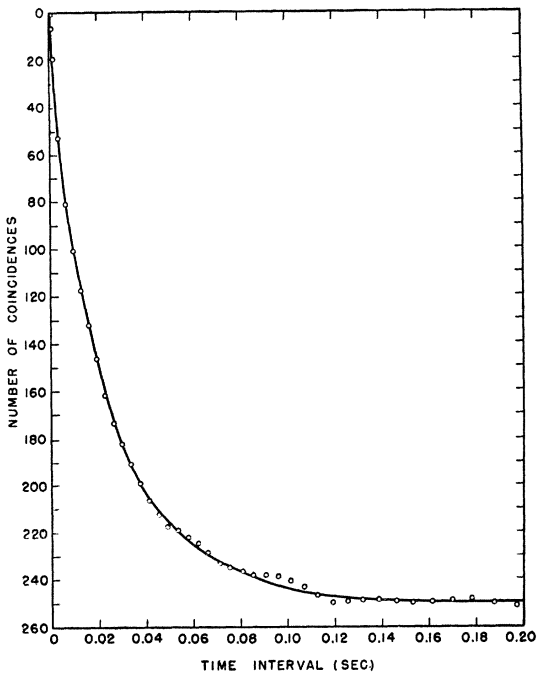


FIG. 9. Integrated frequency distribution of $Ra^{222}$-$Em^{218}$ coincidences representing the exponential decay of $Em^{218}$.

pair, there is a sudden change in the plate voltage of tube, $T_1$, which is reflected in a change in voltage across condenser $C_1$.

The circuit was operated as follows: The potentiometer, $P$, was adjusted until the fluorescent spot was at the extreme right of the oscilloscope screen when tube, $T_1$, was in the conducting state. A pulse caused by alpha-disintegration in the ionization chamber of the counter would cause tube, $T_1$, to change to the non-conducting state with a consequent sudden rise in plate voltage. Condenser, $C_1$, would charge through $R_1$ and $R_2$ at a rate dependent on the time constant, $(R_1+R_2)C_1$. As the voltage across the condenser increased, the spot moved across the screen. If another alpha-disintegration occurred during the period of the sweep, the stable states of the trigger pair would again reverse and the spot would return to its original position as the condenser discharged. The distance that the spot traveled across the screen was a measure of the time interval between the two successive alpha-disintegrations. Such "coincidences" were observed visually with the aid of a calibrated screen. A sine wave from an oscillator was put in the vertical deflecting plates of the scope to give a moving band which was more easily observed than a moving spot. Resistor, $R_1$, and condenser, $C_1$, were so chosen that the sweep was about 0.2 sec. If a single alpha-disintegration occurred, the spot would move across the screen and off the other side. It could be brought back to its original position by shorting out condenser, $C_1$, with the switch, $S$.

A time calibration of the sweep is dependent on the time constant of the circuit, the voltage difference between the conducting and non-conducting states of tube, $T_1$, and the sensitivity of the oscilloscope. The time constant, $(R_1+R_2)C_1$ was 0.1658 second, the voltage difference was 235 volts, and the sensitivity of the oscilloscope was 43 volts per inch. The equation for the instantaneous potential difference between the horizontal deflection plates of the oscilloscope was

$$v=235(1-e^{-t/0.1658}).$$

From the above equation and the sensitivity of the oscilloscope a calibration curve was constructed relating time with sweep distance.

It is necessary to consider the possibility of chance coincidences between alpha-particles arising from the background of the sample and counter. The relative number of such random coincidences can be minimized by using low-counting rate samples and low-background counters. Although it may not be feasible to eliminate the background effect completely, corrections can be made for random coincidences. Since the time distribution of particles emitted from a long-lived radioactive substance is purely random, the probability that $n$ particles arrive in time, $t$, is given by the Poisson equation,

$$W_n=(x^n e^{-x}/n!),$$

where $x$ is the average number of particles which arrive in time, $t$. In the experimental apparatus the emission of an alpha-particle starts the sweep on the oscilloscope. Since for a purely random distribution the probability of a future event is not affected by the previous occurrence of an event, the chance that another particle arrives during the period of the sweep is given by the Poisson equation. Thus it is only necessary to determine the number of random alpha-

disintegrations to correct for random coincidences.

In order to obtain a high ratio of true coincidences to random events a high geometry factor is essential. A geometry factor approaching 100 percent was obtained by mounting the samples on a very thin film of zapon supported on a 0.002-in. wire ring about one cm in diameter. The samples were supported in a vertical position between the parallel plate electrodes of the ionization chamber of the counter.

The half-life of $Em^{218}$ was obtained by measuring the time interval up to 0.2 sec. between successive alpha-particles from a sample of $U^{230}$ in equilibrium with its daughters. The half-life of RaC′ is so short and the resolving time of the counter which was used was so long that in most cases the arrival of alpha-particles from $Em^{218}$ and RaC′ were recorded as a single event. When the pulse due to the RaC′ alpha-particle was resolved, it resulted in "triple" coincidences; thus if the $Ra^{222}$ alpha-particle starts the sweep, the $Em^{218}$ alpha-particle reverses it, and the RaC′ alpha-particle reverses it again. Only about three percent of the total observed coincidences were such triple coincidences, and they were neglected. It can be seen from the decay chain that half of the alpha-particles will be observed as true $Ra^{222}$-$Em^{218}$ coincidences. The half-life of $Ra^{222}$ is so long by comparison with the sweep time that relatively few $Th^{226}$-

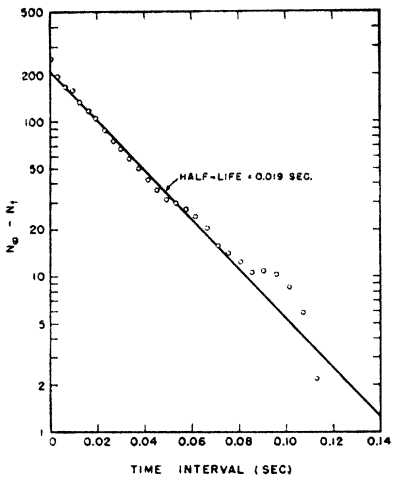


FIG. 10. Decay curve of $Em^{218}$. $N_\infty$ = total number of observed coincidences. $N_t$ = number of coincidences observed within the time interval, $t$.

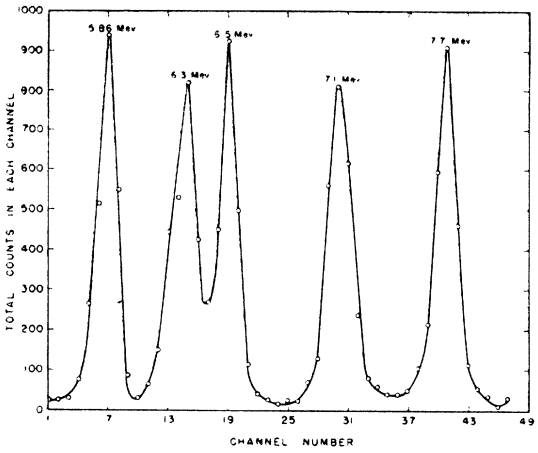


FIG. 11. Alpha-spectrum of the protactinium series as obtained with a multi-channel, differential pulse analyzer.

$Ra^{222}$ coincidences occur. Those that do occur will result in triple coincidences. The total random counting rate is that due to $U^{230}$ and $Th^{226}$, or half the total counting rate. (The background of the counter was negligible.) Since two alpha-particles are involved in each coincidence, the total number of random events will be twice the number of coincidences. The average total disintegration rate of the sample was 12.3 per minute. The probable number of random coincidences was calculated for the observed time intervals and subtracted from the recorded number of coincidences. The correction varies almost linearly with time over the range represented by the period of the sweep. About 4 percent of the total coincidences observed within 0.2 sec. were estimated to be due to random events.

Figure 9 shows the frequency distribution of the observed time intervals after correction for random events. The total number of observed coincidence periods equal to or less than a given time interval is plotted against the time interval. The integral curve so obtained is exponential within the errors of the experiment and represents the decay of $Em^{218}$. The mean interval is 0.027 sec. corresponding to a half-life of 0.019 sec.

The same value was obtained by the following graphical analysis. Since the maximum interval observed is greater than ten half-lives of $Em^{218}$, essentially all of the coincidences which occurred were observed. By subtracting $N_t$, the number of coincidences observed within a given time in-

terval, from $N_\infty$, the total number of observed coincidences, and plotting this number, $N_\infty - N_t$, against the time intervals on semi-logarithmic paper a straight line was obtained as shown in Fig. 10. The slope of the line is a measure of the half-life of $Em^{218}$.

It is difficult to evaluate the reliability of the above data because of the uncertainty of the human error involved in the observations. However, it is believed that the half-life is not in error by more than 0.002 sec.

4.8 *Determination of the alpha-energies of the members of the protactinium series.*—The energies of the alpha-particles were determined with a multi-channel, differential pulse analyzer.[7] The instrument which was used has 48 electronic channels which register all the alpha-pulses within a selected range of energies. Each channel records only the pulses falling within a narrow energy band; the 48 channels cover contiguous energy bands of equal width increasing in energy from channel No. 1 to channel No. 48. By plotting the total number of alpha-counts per channel against channel number, a peak will be observed for each alpha group. The position and size of the peak measure respectively the energy and abundance of the alpha group.

Figure 11 shows the entire series of peaks in the protactinium series. These correspond to $U^{230}$, $Th^{226}$, $Ra^{222}$, $Em^{218}$ and RaC′. To determine the energies it was necessary to calibrate the instrument with several isotopes whose known energies are approximately the same as those of the protactinium series. Among these standards was a preparation containing RaC′ which served to establish the identity of the most energetic alpha in the protactinium series. With the aid of these standards the unknown energies were estimated to be $5.8_6$ Mev, 6.3 Mev, 6.5 Mev, 7.1 Mev and 7.7 Mev.

The question of the assignment of each energy to the proper isotope remained. The RaC′ assignment was the only certain one in the beginning. The assignment of the energy of the $U^{230}$ alpha-particle was made very definite by means of two experiments:

(1) An active sample of pure $Pa^{230}$ was permitted to decay to $U^{230}$ for several days. Then a rapid extraction was performed to remove uranium. A sample of the $U^{230}$ was quickly inserted in the pulse analyzer instrument and the alpha-spectrum was measured before the daughter isotopes had time to grow into their equilibrium value. The alpha-spectrum was redetermined after several hours when the daughter isotopes were in equilibrium. The $5.8_6$ Mev peak, which was prominent in the first curve, was assigned to $U^{230}$ and its relation to the other peaks was established by the second curve.

(2) In a second experiment a two-inch aluminum disk was fastened to the upper electrode of a standard alpha-counting ionization chamber and used to collect recoil atoms from a very active sample of $U^{230}$ placed on the lower electrode. This disk was quickly removed and subjected to pulse analysis. All peaks except the $5.8_6$ Mev peak were present, which confirms the assignment of this peak to $U^{230}$

*Note added in proof:* On the basis of a re-analysis of pulse-analyzer data on the protactinium series isotopes, A. H. Joffey reports the following revised figures for the first four alpha-energies: $5.85 \pm 0.01$ Mev, $6.30 \pm 0.02_5$ Mev, $6.51 \pm 0.03$ Mev, and $7.12 \pm 0.02_5$ Mev.

This leaves three energies to be assigned. A rigorous assignment is made very difficult because of the short half-lives and because of the rapidity with which these isotopes come into equilibrium with each other. Experimental methods are known by which such a problem may be solved, but up to the present the necessary instruments have not been available to perform such experiments. Hence the remaining energies have been tentatively assigned to the isotopes in the inverse order of their half-lives. Thus the 6.3 Mev alpha-particle is assigned to the 30.9 minute $Th^{226}$, the 6.5 Mev alpha-particle to the 38 second $Ra^{222}$, and the 7.1 Mev alpha-particle to the 0.019 second $Em^{218}$.

## ACKNOWLEDGMENTS

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[7] A. H. Jaffey, Plutonium Project Record **14A**, Chap. XVI; A. Ghiorso, A. H. Jaffey, H. P. Robinson and B. Weissbourd, Plutonium Project Record, **14B**, No. 16.8.