The 4n+1 Radioactive Series: The Decay Products of U^{233}*

FRENCH HAGEMANN, L. I. KATZIN, M. H. STUDIER, G. T. SEABORG,† AND A. GHIORSO†
Argonne National Laboratory, Chicago, Illinois
(Received March 13, 1950)

The path of the artificially produced $4n+1$ radioactive decay series between U^{233} and Bi^{209} and the properties of the individual members of the chain are developed.

I. INTRODUCTION

THE three families of radioactive heavy isotopes found in nature are differentiated by their mass characteristics. Thus, the family represented by thorium (Th^{232}) and its decay products has masses corresponding to the formula $4n+0$; the family represented by U^{238} and its decay products corresponds to $4n+2$; and the family represented by U^{235} and its decay products corresponds to $4n+3$, where n in all cases represents an integral multiplier. The family corresponding to the formula $4n+1$ has not been found in nature, except for the stable end-member of the chain, bismuth (Bi^{209}).

Previous experiments[1–7] in artificial production of nuclei have resulted in the formation of the isotopes Pb^{209}, Th^{233}, Pa^{233}, U^{233}, and Np^{237}, whose masses correspond to the $4n+1$ formula. From the isotope Np^{237}, which decays by emission of an alpha-particle, beta-active Pa^{233}, and from it alpha-active U^{233}, are formed by successive decays. We may therefore consider Np^{237} to represent the long-lived parent of a $4n+1$ radioactive decay family, and in analogy with natural series this may be called the neptunium series.[8]

The purpose of this paper is to trace the path of the decay chain between the isotopes U^{233} and Bi^{209}, and to give the properties which have been found for the isotopes which are members of the chain. The findings may be summarized as in Fig. 1 and Table I. Work in another laboratory, where parallel studies have been made of the U^{233} decay chain,[9] has resulted in essentially similar findings.

Various predictions have been made in the literature with regard to the properties of the $4n+1$ series. Russell[10] suggested the following decay scheme:

$$U^{237}\xrightarrow{\alpha}Th^{233}\xrightarrow{\beta^-}Pa^{233}\xrightarrow{\alpha}Ac^{229}\xrightarrow{\beta^-}Th^{229}\xrightarrow{\alpha}Ra^{225}\xrightarrow{\alpha}$$
$$Em^{221}\xrightarrow{\alpha}Po^{217}\xrightarrow{\alpha}Bi^{213}\begin{cases}\xrightarrow{\beta^-}Po^{213}\xrightarrow{\alpha}\\ \xrightarrow{\alpha}Tl^{209}\xrightarrow{\beta^-}\end{cases}Pb^{209}\xrightarrow{\beta^-}Bi^{209}.$$

On the basis of analogy with the three known series Ra^{225} was predicted to be alpha-active, decaying to alpha-active Em^{221}. Later Widdowson and Russell[11] revised this prediction to suggest branching decay of Ra^{225} and beta-decay of the emanation isotope. The chain would thus proceed through isotopes of elements 87 and 85 to predominantly beta-active Bi^{213} as shown below, rather than through the even-numbered elements as in the natural series.

$$Em^{221}\begin{cases}\xleftarrow{\alpha}\\ \xrightarrow{\beta^-}\end{cases}\begin{matrix}Ra^{225}\\ \\ \\ Ac^{225}\end{matrix}\begin{matrix}\\ \xrightarrow{\beta^-}\\ \xleftarrow{\alpha}\\ \\ \end{matrix}87^{221}\xrightarrow{\alpha}85^{217}\xrightarrow{\alpha}Bi^{213}\xrightarrow{\beta^-}\text{etc.}$$

Sometime later Turner[12] predicted that U^{237} would be a beta- rather than an alpha-emitter and supported Meitner, Strassman, and Hahn's[3] assignment of a 25-day Pa activity to Pa^{233}. The decay scheme proposed by Turner was:

$$U^{237}\xrightarrow{\beta^-}Np^{237}\xrightarrow{\alpha}Pa^{233}\xrightarrow{\beta^-}U^{233}\xrightarrow{\alpha}Th^{229}\xrightarrow{\alpha}Ra^{225}\xrightarrow{\beta^-}$$
$$Ac^{225}\xrightarrow{\alpha}87^{221}\xrightarrow{\alpha}85^{217}\xrightarrow{\alpha}Bi^{213}\begin{cases}\xrightarrow{\beta^-}Po^{213}\xrightarrow{\alpha}\\ \xrightarrow{\alpha}Tl^{209}\xrightarrow{\beta^-}\end{cases}Pb^{209}\xrightarrow{\beta^-}Bi^{209}.$$

Turner's scheme leads through elements 85 and 87, as did the prediction of Widdowson and Russell, but an emanation would not be formed in the main line.

---

* Preliminary report, Phys. Rev. **72**, 252 (1947). Except where otherwise indicated, these studies were made during 1944–46, at the Metallurgical Laboratory, University of Chicago (now the Argonne National Laboratory), under the auspices of the Manhattan Engineer District.

† Present address: Radiation Laboratory and Department of Chemistry, University of California, Berkeley, California.

[1] R. L. Thornton and J. M. Cork, Phys. Rev. **51**, 383 (1937); R. S. Krishnan and E. H. Nahum, Proc. Camb. Phil. Soc. **36**, 490 (1940); K. Fajans and A. F. Voight, Phys. Rev. **60**, 619 (1941); W. Maurer and W. R. Ramm, Zeits. f. Physik, **119**, 602 (1942).

[2] Fermi, Amaldi, D'Agostino, Rasetti, and Segrè, Proc. Roy. Soc. **A146**, 483 (1934).

[3] Meitner, Strassmann, and Hahn, Zeits. f. Physik **109**, 538 (1938).

[4] Grosse, Booth, and Dunning, Phys. Rev. **59**, 321 (1941).

[5] Seaborg, Gofman, and Kennedy, Phys. Rev. **59**, 321 (1941).

[6] Seaborg, Gofman, and Stoughton, Phys. Rev. **71**, 378 (1947).

[7] A. C. Wahl and G. T. Seaborg, Phys. Rev. **73**, 940–941 (1948).

[8] G. T. Seaborg, *The Transuranium Elements: Research Papers* (McGraw-Hill Book Company, Inc., New York, 1949), Paper No. 22.3, National Nuclear Energy Series, Plutonium Project Record, Vol. 14B.

[9] English, Cranshaw, Demers, Harvey, Hincks, Jelley, and May, Phys. Rev. **72**, 253 (1947).

[10] A. S. Russell, Phil. Mag. **46**, 642–656 (1923).

[11] W. P. Widdowson and A. S. Russell, Phil. Mag. **48**, 293–306 (1924).

[12] L. A. Turner, Phys. Rev. **57**, 950 (1940).

TABLE I. The $4n+1$ radioactive series.

Isotope	Type of radiation	Half-life[a]	Energy of radiation (Mev)
Np^{237}	α	2.20×10^6 yr.[b]	4.77
Pa^{233}	β^-	27.4 days[c]	0.23[f]
U^{233}	α	$1.62(\pm0.01)\times10^5$ yr.[d]	4.80[g]
Th^{229}	α	$(7.34\pm0.16)\times10^3$ yr.	4.85; (~70%) 4.94; (~20%) 5.02; (~10%)
Ra^{225}	β^-	14.8±0.2 days	*ca.* 0.2
Ac^{225}	α	10.0±0.1 days	5.80
Fr^{221}	α	4.8±0.1 min.	6.30
At^{217}	α	0.018±0.002 sec.	7.00
Bi^{213}	β^-(98%), α(2.0%)	47±1 min.	*ca.* 1.2(β^-); 6.0(α)
Po^{213}	α	Very short	8.34
Tl^{209}	β^-	2.20±0.07 min.	1.8
Pb^{209}	β^-	3.3 hr.[e]	0.7[e]
Bi^{209}	Stable		

[a] Limits of error for Ra^{225}, Ac^{225}, Fr^{221}, and At^{217} are estimates rather than true probable errors.
[b] L. B. Magnusson and T. LaChapelle, J. Am. Chem. Soc. **70**, 3534–8 (1948).
[c] See reference 4.
[d] E. K. Hyde, Paper No. 19.15, National Nuclear Energy Series, Plutonium Project Record, Vol. 14B; AECD Doc. No. 2457.
[e] See reference 1.
[f] E. Haggstrom, Phys. Rev. **62**, 144–150 (1942).
[g] A. H. Jaffey (private communication).

Feather[13] believed Turner's predictions should be correct, and in actual fact the only deviation from our experimental findings is his prediction that the major decay of Bi^{213} would be by alpha-emission, whereas this mode accounts for only two percent of the disintegrations.

Ponisovsky,[14] from independent studies of the regularities in the stable isotopes, predicted essentially the same series, with the difference that the Th^{229} was predicted to be beta-active, leading to an alpha-active Pa^{229} which would give the Ac^{225} without the appearance of a radium isotope in the chain.

With respect to possible alpha-branching of the Ra^{225}, our experiments on separating this isotope from its daughter Ac^{225} have enabled us to set 0.5 percent as an upper limit on the amount of such branching. No emanation has been detected. No evidence for beta-decay of the Th^{229} has been found. The properties of the theoretical product, Pa^{229}, are known from material produced by cyclotron bombardment.[15]

II. EXPERIMENTAL

U^{233}

The U^{233} used in these experiments was prepared by neutron irradiation of thorium in the uranium-graphite pile. Following decay of most of the intermediate Pa^{233} and Th^{233}, the uranium was separated by extraction with ether.[16] A series of extractions and precipitations of sodium uranyl acetate, ammonium diuranate, and uranyl peroxide served to give complete chemical and radiochemical purification of the uranium. The nuclear properties of this isotope are described elsewhere.[17]

The U^{233} prepared in this way contained four to five percent of U^{238} since the thorium compounds irradiated normally contained a part or so per million of natural uranium impurity. Account was taken of the UX activity introduced from this isotopic impurity in experiments involving thorium isotopes. Lower members of the uranium decay chain were present in amounts which were insignificant because of the long half-life of U^{234} and its low concentration.

An important impurity was found to be U^{232},[18] which decays to radiothorium of 1.9-yr. half-life, followed by ThX(Ra^{224}) and its short-lived daughters. The RdTh half-life is so short relative to that of the Th^{229} that even with only one disintegration of U^{232} in 5700 disintegrations of U^{233} (the concentration actually found in one sample of the material by a mica absorption method), a significant fraction of the daughter radioactivities extracted after a decay period of a few weeks or months is due to this impurity. The ratio of Th^{229} activity to RdTh activity was found to vary from about 2 to 3.5 for different samples of U^{233}. The origin of the U^{232} is probably a small Pa^{231} impurity in the thorium irradiated, since Pa^{231} has a high neutron capture cross section.[19] The capture product, Pa^{232}, decays to U^{232} by

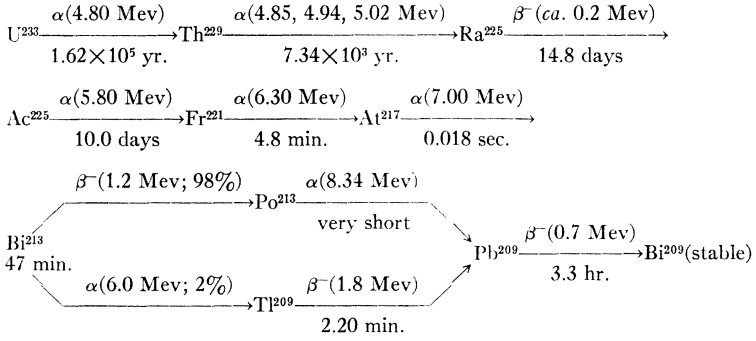

FIG. 1. Decay scheme of the neptunium series.

[13] N. Feather, British Atomic Energy Projects, Report Br-339 (1943).
[14] L. Ponisovsky, Nature **152**, 187–8 (1943).
[15] Hyde, Studier, Hopkins, and Ghiorso, Paper No. 19.17, National Nuclear Energy Series, Plutonium Project Record, Vol. 14B.
[16] Hagemann, Katzin and Studier, Paper No. 3.15, National Nuclear Energy Series, Plutonium Project Record, Vol. 17B.
[17] M. H. Studier, Paper No. 1.3, National Nuclear Energy Series, Plutonium Project Record, Vol. 17B; AECD Doc. No. 2444.
[18] J. W. Gofman and G. T. Seaborg, Paper No. 19.14, National Nuclear Energy Series, Plutonium Project Record, Vol. 14B.
[19] A. H. Jaffey and Q. Van Winkle, Paper No. 9.16, National Nuclear Energy Series, Plutonium Project Record, Vol. 17B.

beta-emission with a half-life of 1.4 days.[18] An alternative possibility is that there is a sufficient flux of fast neutrons to give a sufficient $(n, 2n)$ reaction with the thorium to yield the Pa^{231}. A second-order absorption would then give rise to the U^{232}.

III. ISOTOPIC ASSIGNMENTS

Assignments of the periods and radiations to definite isotopes were made on the basis of radiochemical separations together with range analyses of the alpha-particles by means of a pulse analyzer.[20]

The isolated thorium activity showed the anticipated alpha-activity. After RdTh daughters had grown to equilibrium, alpha-activity continued to grow over a period of time at a rate corresponding to a half-life of the order of three weeks. The number of alpha-activities growing from the Th^{229} was shown to be four, both from range analysis by the pulse analyzer, and from the increase in alpha-activity. Radiochemical isolation of a combined radium-actinium fraction (e.g., on barium sulfate) gives an alpha-active preparation into which two alpha-activities grow with a 5-min. half-life, followed by growth with a $\frac{3}{4}$-hr. half-life. Separation into radium and actinium fractions shows the radium isotope to possess a soft beta-particle of about 0.2-Mev energy, while the actinium fraction repeats the alpha-activity growth pattern noted above. Four alpha-activities (the actinium isotope and its daughters) grow into the radium fraction on a time relation dependent on the 14.8-day radium period and the 10.0-day actinium period. The fact that two alpha-activities (and no beta-) grow in with a 5-min. period indicates that the half-life of At^{217} is considerably shorter than the 5-min. period of Fr^{221}. This is borne out by the period determined from coincidence measurements (0.018 sec.). A bismuth fraction free of lead isotopes shows an alpha-activity and a beta-activity decaying with the same half-life, leaving a beta-activity ascribable to daughter Pb^{209}. The short half-life of the alpha-emitter and the long range of its particle indicate it to be due to Po^{213}, produced by beta-decay of Bi^{213}. Evidence from range analysis of the Bi^{213} activity indicates some branching by alpha-decay.

Th^{229}

This isotope, together with the radiothorium impurity mentioned above, was isolated from the purified U^{233} solution after a suitable period of time by co-precipitation with zirconium iodate.

One- to two-tenths of a milligram of zirconium nitrate and sufficient iodic acid solution to give a final iodate concentration of $0.05M$ were added per milliliter of solution of U^{233} in $0.1N$ nitric acid. The precipitated zirconium iodate was washed by centrifugation, dissolved in sulfur dioxide-water, heated to remove liberated iodine, diluted, and the zirconium reprecipitated with iodic acid. Four to five of these cycles served to decontaminate completely from uranium, radium, and most of the actinium. Bismuth and lead activities were removed by one or two lead sulfide by-product precipitates.

In order to prepare samples sufficiently free from carrier for alpha-particle range measurements in the pulse analyzer, and to remove any traces of actinium activity, the following procedure was used. The thorium isotopes were co-precipitated from the final zirconium iodate solution with lanthanum fluoride carrier (0.1–0.2 mg/ml), the fluoride precipitate was

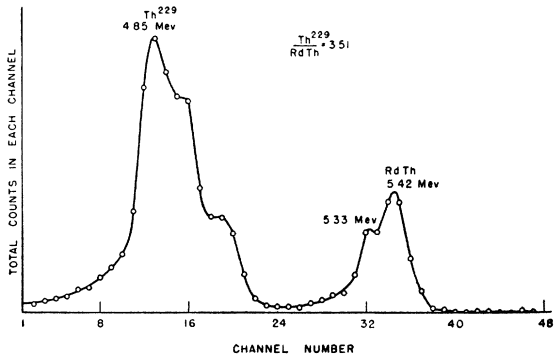

FIG. 2. Pulse analyzer curve of thorium alpha-activity showing the three alpha-peaks of Th^{229} together with the peaks due to RdTh impurity.

metathesized to hydroxide with concentrated potassium hydroxide and the hydroxide dissolved in $0.05M$ nitric acid. Separation of the thorium activity from the lanthanum carrier was accomplished by extraction with a solution[21] of $0.15M$ thenoyl trifluoracetone (TTA) in benzene. The change from zirconium to lanthanum carrier was necessary since the former extracts into TTA solution under the conditions used, whereas the latter does not. In some cases, TTA extraction of the thorium isotopes was made directly from the uranium solution. The TTA benzene solution was in either case evaporated directly on the counting plate or re-extracted with $8N$ nitric acid and the latter solution evaporated. Ignition of the resulting plates left an essentially weightless film of the thorium activity.

Since the Th^{229} half-life is several thousand years, indirect methods of determining this property were necessary. A known amount of freshly purified U^{233} was allowed to decay for a measured time. UX_1 tracer was then added and the thorium activity isolated as above. The UX_1 tracer provided a means of determining the yield of thorium through the separation procedure without introducing any foreign alpha-activity. The ratio of Th^{229} to radiothorium was determined by alpha-pulse analysis. Applying corrections for yield and for radiothorium impurity the amount of Th^{229} activity which had grown into the U^{233} could be calculated, and

[20] Ghiorso, Jaffey, Robinson, and Weissbourd, Paper No. 16.8, National Nuclear Energy Series, Plutonium Project Record, Vol. 14B.

[21] J. C. Reid and M. Calvin, AECD Doc. No. 1405.

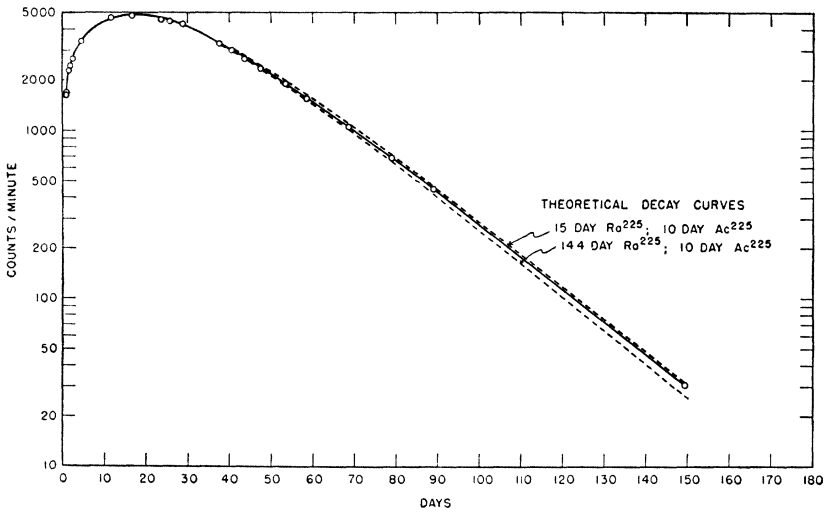

FIG. 3. Growth and decay of alpha-activity from Ra^{225} preparation. Theoretical decay curves calculated on the basis of 10-day Ac^{225} and 15-day and 14.8-day Ra^{225} are shown by the broken lines.

from this the Th^{229} half-life. The mean value obtained from eight such determinations was 7340±160 yr.

The alpha-radiations of Th^{229} have been found to be complex. The main group of alpha-particles, which constitutes approximately 10 percent of the total, has an energy of 4.85±0.01 Mev. Two small longer-range groups of particles have been resolved from the main group, differing from it by 90 and 170 kev, respectively. The less energetic of these two small groups contains roughly about twice the number of alpha-particles in the other. Figure 2 is a typical curve obtained with the pulse analyzer of the thorium activity which shows the two longer-range groups of the Th^{229} alpha-particles together with the RdTh impurity mentioned above. The energy of the Th^{229} particles was determined by placing a standard containing some Po^{210} (5.30 Mev) and ionium (4.66 Mev)[22] in the chamber together with the sample. The Th^{229} energy was found by interpolation from the resulting calibration curve using the Po^{210}, Io, and RdTh peaks[23] as reference points of known energy.

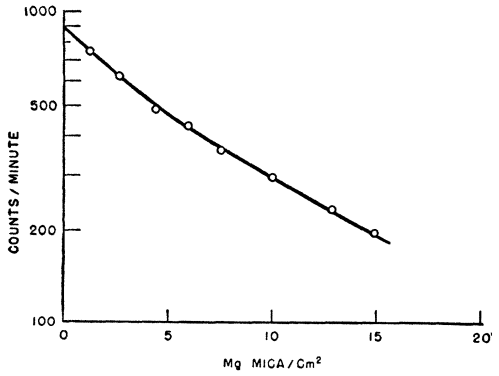

FIG. 4. Absorption of Ra^{225} beta-particles in mica.

[22] Clark, Spencer-Palmer, and Woodward, British Atomic Energy Projects, Report Br-522.
[23] Clark, Spencer-Palmer, and Woodward, British Atomic Energy Projects, Report Br-584.

Ra^{225}

The Ra^{225} (and ThX from the U^{232} impurity) was isolated from the U^{233} solution from which the thorium activity had been separated. Both barium sulfate and barium chloride were used as carriers for the radium activity; the latter was found to be preferable with regard to decontamination from actinium. With barium chloride the procedure was to add 1 mg of barium as nitrate to the U^{233} solution, reduce the volume to a few tenths of a milliliter and add seven times the volume of hydrochloric acid-ether solution (six parts conc. HCl to one part ethyl ether). After cooling with ice the precipitated barium chloride was centrifuged and reprecipitated in the same manner. Two or three precipitations were sufficient to remove the U^{233}; however, appreciable actinium activity still remained at this point. This was removed either by precipitating lanthanum fluoride from the solution or by adding lanthanum nitrate, evaporating to dryness and extracting the lanthanum and actinium nitrates with absolute ethyl alcohol. Lead sulfide was precipitated from the solution to remove bismuth and lead activity and the barium finally precipitated and mounted for counting as the sulfate.

Because of the low energy of its beta-particles, the half-life of the Ra^{225} was determined indirectly by following the growth and decay of the alpha-activity in the fraction isolated as above. Figure 3 shows an experimental curve and theoretical curves calculated on the basis of a 10.0-day Ac^{225} daughter (see below) assuming different values for the Ra^{225} half-life and using the observed maximum count. The growth of the alpha-activity does not follow precisely the theoretical values due to the presence of the ThX impurity. However, by the time the maximum value has been passed the short-lived ThX has largely decayed away. The experimental points best fit a theoretical curve for a 14.8-day radium half-life. Since the Ra^{225} and Ac^{225} half-lives are similar,

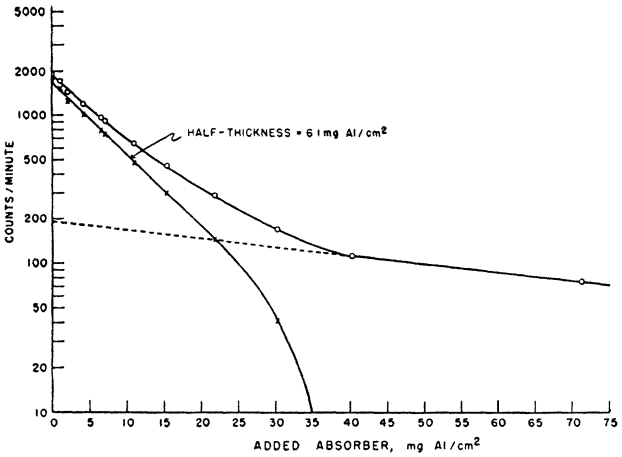

FIG. 5. Absorption of Ra^{225} beta-particles in aluminum measured with a mica-window Geiger tube (3 mg mica/cm²).

transient equilibrium is approached very slowly and direct determination of the Ra^{225} half-life from the decay curve with reasonable accuracy would be possible only after some hundreds of days of decay.

Beta-ray absorption curves were taken on samples of Ra^{225} repurified after having been allowed to stand for some weeks in order to eliminate the effects of the ThX daughters. The original Ra fraction was isolated from the U^{233} solution upon barium chloride carrier and reprecipitated to remove uranium and thorium activities. After about 30 days the Ra^{225} was purified from daughter activities by the above procedure, mounted on platinum, and absorption curves taken. Figure 4 is the curve of the absorption in mica obtained in a windowless G-M counter 5 hr. after separation. One-half hour after separation this sample showed 30 alpha-counts/min. Other curves taken with a low absorption counter using a thin (*ca.* 0.1 mg) zapon window and aluminum absorbers were similar. The amount of Ra^{225} activity which should have been present initially was calculated from the growth of alpha-activity in the sample to be about 650 counts/min., making approximate corrections for counting geometry. This agrees well with the value for zero absorber extrapolated from the curve after correcting for backscattering and self-absorption. Figure 5 shows the curve taken on another sample, 8 hr. after separation, with an ordinary thin window (3.3 mg/cm² of mica) Geiger counter using Al absorbers. The absorption half-thickness of the Ra^{225} beta-particle from these curves is about 6 mg/cm² and the range about 35 mg Al/cm², corresponding to an energy of *ca.* 0.2 Mev. No reliable information is available about gamma-rays.

Ac^{225}

Ac^{225} was separated by means of lanthanum fluoride carrier from the U^{233} solution from which Th^{229} had been removed. The fluoride precipitate was metathesized by means of potassium hydroxide and then dissolved in nitric acid. After two or three such cycles barium nitrate was added to "holdback" radium, the solution was evaporated to dryness and the lanthanum and actinium extracted from the residue with absolute ethyl alcohol.[24] The alcohol was evaporated, more barium nitrate added, and the extraction repeated. After precipitating lead sulfide to remove any bismuth or lead activity remaining, the lanthanum and actinium were precipitated as fluoride and mounted. A typical growth and decay curve for the Ac^{225} fraction is shown in Fig. 6 from which the half-life of 10.0 days was obtained by least-squares analysis.

Fr^{221}

Analysis of the growth curves of the alpha-activity in the Ac^{225} sample showed that three alpha-activities grew in, two with a half-life of about 5 min. followed by a third with a half-life of about 45 min. The 5-min. growth appeared to start with the flaming of the plate containing the activity, and after equilibrium had been reached, the 5-min. growth reappeared if the sample

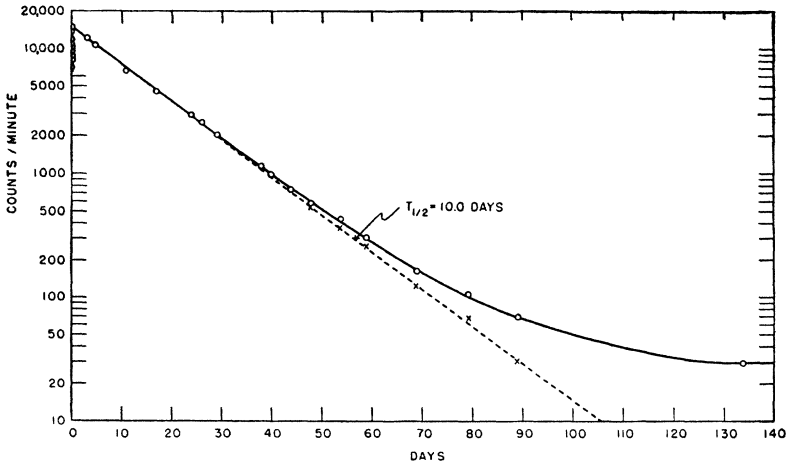

FIG. 6. Growth and decay of alpha-activity from Ac^{225}.

[24] M. Haissinsky, Comptes Rendus **196**, 1778–9 (1933).

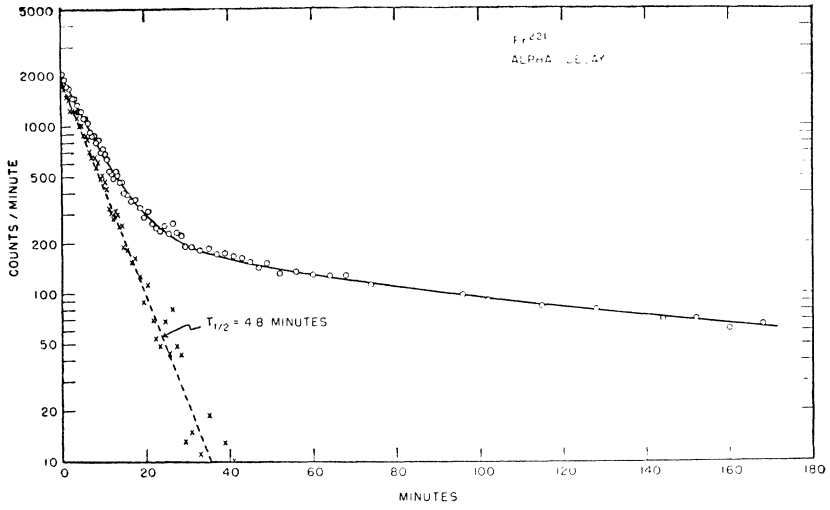

FIG. 7. Decay of alpha-activity from Fr^{221}. The long-lived background is probably unseparated Ac^{225}.

was reflamed. Experiments with Cs^{134} tracer indicated that this activity could be volatilized by flaming under similar conditions. These facts indicated that the daughter of Ac^{225}, Fr^{221}, was an alpha-emitter of about 5-min. half-life and its daughter was also an alpha-emitter of much shorter life.

The half-life of the Fr^{221} was determined more accurately by precipitating lanthanum hydroxide from a small volume of a solution of Ac^{225} and its daughters with ammonia, mounting the supernatant solution and following the decay of the alpha-activity obtained. Figure 7 shows the observed decay curve together with the curve resulting from subtraction of the non-decaying background (probably Ac^{225}). The half-life obtained from the latter curve gave a value of 4.8 min.

At^{217}

The half-life of At^{217} was determined by measuring the time interval between successive alpha-particle emissions by Fr^{221} and At^{217}. A sample of Ac^{225} was mounted upon a very thin zapon film supported on a 2-mil wire ring of about 1-cm diameter. The ring was placed in a vertical position on the bottom electrode of the chamber of a standard alpha-counter.[25] A resistor and condenser in series were connected between the

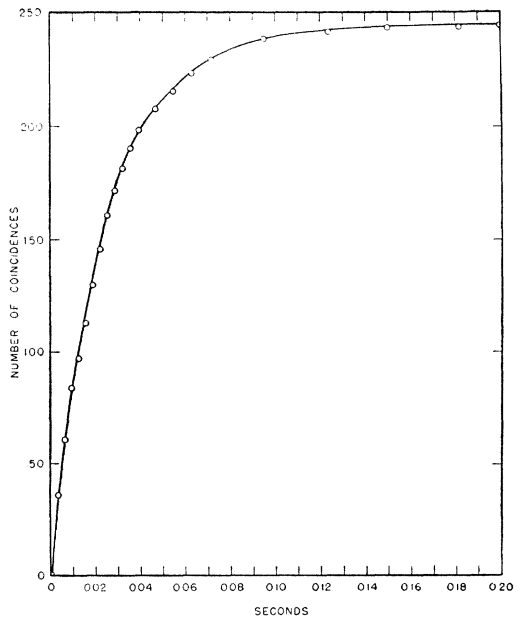

FIG. 8. Integral curve obtained by plotting the total number of coincidences observed between Fr^{221} and At^{217} alpha-particles in fixed time intervals against the time intervals. Corrections for chance coincidences have been applied.

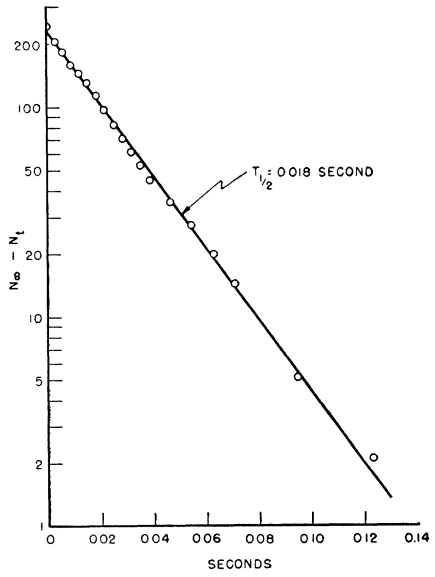

FIG. 9. Plot of the difference between asymptotic value of the total number of observed coincidences, N_∞ (from Fig. 8), and the number observed within a fixed interval, N_t, against the time interval. The slope of the line gives directly the half-life of At^{217}.

[25] A. H. Jaffey, Chapter 16, "Radiochemical assay by alpha and fission measurements," National Nuclear Energy Series, Plutonium Project Record, Vol. 14A.

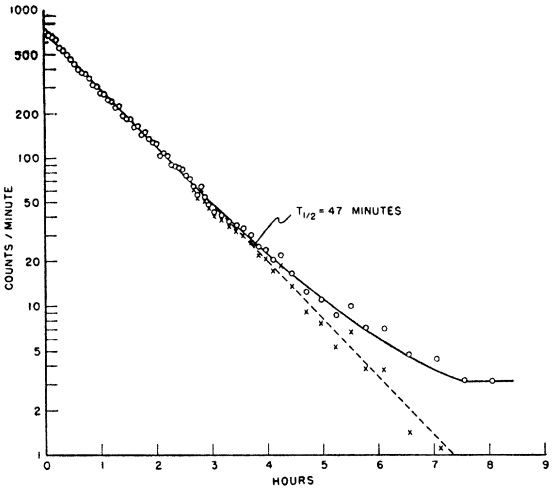

FIG. 10. Decay of alpha-activity from Bi^{213} and daughters.

plate and cathode of one of the tubes of the first Eccles-Jordan trigger pair[26] in the scaling circuit of the counter. The voltage changes across the condenser were followed by an oscilloscope, the plates of which were connected directly across the condenser. Thus a single alpha-particle would cause the electron beam of the oscilloscope to sweep across the screen at a rate dependent upon the time constant of the circuit. If two successive alpha-particles were emitted within this time interval the beam would be returned to its original position and the distance through which it swept would be a measure of the interval between the pulses. The time constant of the circuit was such that 0.2 sec. was required for the beam to sweep across the screen. The distances swept for successive emissions were observed visually. A detailed description of the circuit and method may be found in another paper from this laboratory.[27]

The purpose of the special mounting of the sample was to increase the geometry factor to approximately 100 percent in order to obtain as high a ratio of coincident to single pulses as possible. A low counting rate (9 counts/min.) was used in order to reduce corrections for coincidences due to random emission of single particles. This correction varied linearly in the range of interest, from nil at zero time to about three percent at 0.2 sec.

Figure 8 shows the integral curve obtained by plotting the total number of coincidences within a given time interval against the time interval. The mean interval was 0.026 sec. from which was obtained the value 0.018 sec. for the half-life of At^{217}. The same half-life was obtained by graphical analysis (Fig. 9). Essentially all the coincidences were observed within the maximum time interval. By plotting the difference between the total number of coincidences and the number within a given time interval against the interval on semi-logarithmic paper a straight line was obtained the slope of which determined the half-life of At^{217}.

Bi^{213}

The Bi^{213} was isolated from a solution of Ac^{225} by precipitation with bismuth sulfide carrier. Four or five cycles consisting of solution of the sulfide in hot concentrated hydrochloric acid, dilution with water and reprecipitation of bismuth with hydrogen sulfide served to remove all except lead activity, which was separated upon lead sulfate precipitates.

The Bi^{213} half-life was determined by following the

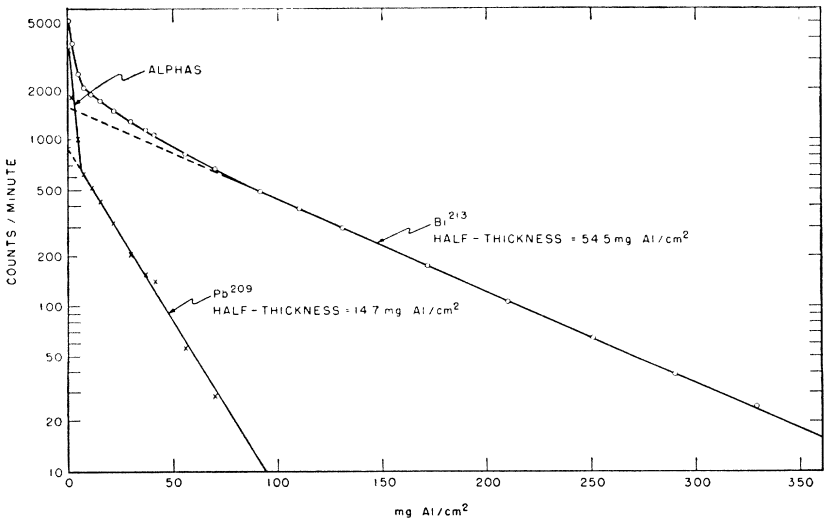

FIG. 11. Absorption curve in aluminum of beta-particles from Ac^{225} and daughters showing the two components due to Pb^{209} and Bi^{213}.

[26] W. H. Eccles and F. W. Jordan, Radio Rev. **1**, 143 (1919).
[27] M. H. Studier and E. K. Hyde, Phys. Rev. **74**, 591–600 (1948).

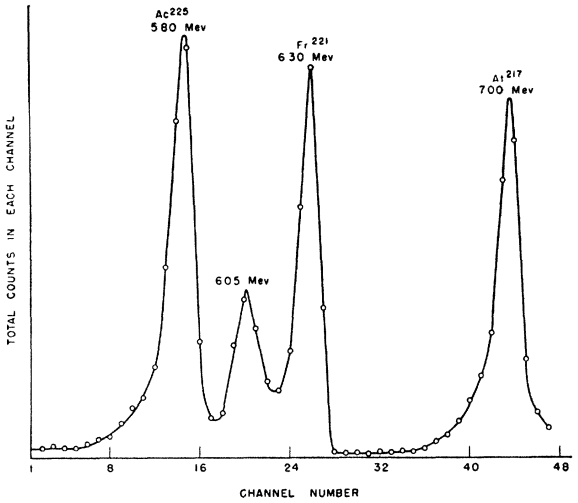

FIG. 12. Typical pulse analyzer curve of the alpha-particles from Ac^{225} and daughters.

very short-lived alpha-emitting daughter Po^{213} rather than the beta-particles of Bi^{213} itself since the growth of beta-particles of 3.3 hr. Pb^{209} is a complicating factor in the latter case. Figure 10 shows a sample decay curve of the Bi^{213} fraction which, after subtracting the non-decaying counter background, gives a value of 47 min. for the Bi^{213} half-life. This same isotope has been identified as a decay product of Pa^{229} by other workers in this laboratory.[15]

Absorption curves in aluminum of the beta-particles from Bi^{213} and Pb^{209} were taken with samples of Ac^{225} in equilibrium with its daughters. Figure 11 shows an experimental curve which has been resolved into the two components; the beta-particles of Bi^{213} have a half-thickness of 54.7 mg Al corresponding very roughly to an energy of 1.2 Mev and those of Pb^{209} have a half-thickness of 14.7 mg corresponding approximately to the reported value[1] of 0.7 Mev.

Energies of Alpha-Particles of Ac^{225}, Fr^{221}, At^{217}, and Po^{213}

The energies of the alpha-particles of these isotopes were determined with the pulse analyzer using a standard of RdTh and its daughters, whose energies are known, to establish reference points. The instrument[20] which was used has 48 electronic channels which register all the alpha-pulses within a narrow energy band; the 48 channels cover 48 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.[28]

The values obtained for the energies were 5.80±0.05 Mev for the Ac^{225} alpha-particles, 6.30±0.05 Mev for Fr^{221}, 7.00±0.05 Mev for At^{217}, and 8.34±0.01 Mev for Po^{213}. A small peak containing about 25 to 30 percent of the number of alpha-particles in the main peaks was observed at 6.05 Mev which has been attributed tentatively to fine structure in the Fr^{221} spectrum. Figure 12 is a typical pulse analyzer curve showing this small peak together with the large peaks for Ac^{225}, Fr^{221}, and At^{217}. It is improbable that this activity is due to unrecognized branching in the series. No radiochemical evidence has been found for branching decay[29] of Th^{229}, Ac^{225}, Fr^{221}, or At^{217}.

Bi^{213} does exhibit some branching decay. The bismuth activity from a solution of Ac^{225} was separated with lead sulfide carrier and purified from other activities by repeated sulfide precipitations. The lead was then precipitated as sulfate and the supernatant containing the carrier-free bismuth activity was mounted and examined in the pulse analyzer. In some cases plates were prepared by direct electrodeposition of the bismuth from 1*N* HCl solution of the actinium. In addition to the normal peak of Po^{213} at 8.34 Mev a small peak at about 6.0 Mev was observed. The sample was followed for a period of several half-lives and both peaks decayed with the Bi^{213} half-life of 47 min. The number of alpha-particles in the shorter range peak was 2.0±0.2 percent of the total in both, based on 15 determinations. An experimental curve is shown in Fig. 13. Although the energy of this small peak is close to that mentioned above attributed to Fr^{221}, the number of alpha-particles from the Bi^{213} is insufficient to account for more than a fraction of the total in that peak.

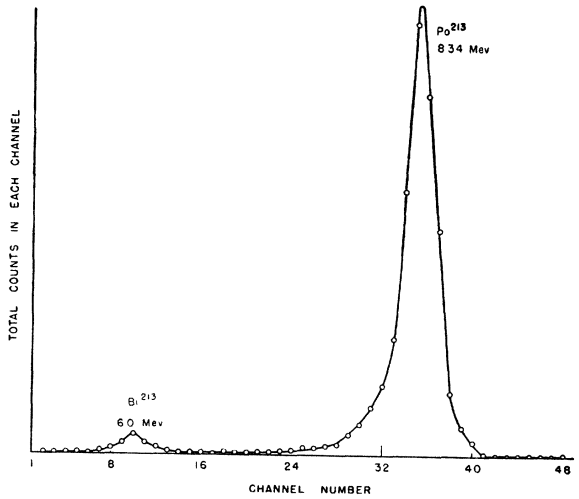

FIG. 13. Pulse analyzer curve of alpha-particles from Bi^{213} and daughters. The small peak at 6.0 Mev containing two percent of the total alpha-particles is due to alpha-branching of Bi^{213}.

[28] We are indebted to B. Weissbourd and J. Mech for assistance with the pulse analyses of our numerous samples.

[29] Systematics of decay of the heavy elements [Perlman, Ghiorso, and Seaborg, Phys. Rev. 77, 26 (1950)] suggest that At^{217} and Fr^{221} may not be beta-stable. There is also the suggestion that At^{217} may have an as yet unobserved alpha-group of higher energy.

With the quantities of material available at the time of these experiments, attempts to isolate the Tl^{209} activity were unsuccessful. Since then one of us[30] has been able to characterize the isotope as decaying with a 2.2-min. half-life by emission of a 1.8-Mev beta-particle. Gamma-rays if present constitute a maximum of one percent of the Geiger activity.

[30] French Hagemann, Phys. Rev. **79**, 534 (1950).

PHYSICAL REVIEW VOLUME 79, NUMBER 3 AUGUST 1, 1950

Interaction of 12- to 13-Mev Neutrons with Deuterons*†

G. L. GRIFFITH,** M. E. REMLEY, AND P. G. KRUGER
University of Illinois, Urbana, Illinois

(Received March 22, 1950)

By use of a CD_4-filled high pressure cloud chamber, the angular distribution of recoils from 12- to 13-Mev neutrons has been studied. The angular range 0° to 140° in the center-of-mass system was covered by these measurements. The distribution indicates a rather large asymmetry in the forward and backward directions with a minimum at about 60° in the center-of-mass system. The existence of a $D(n, 2n)p$ reaction was evident by the presence of tracks between 60° and 90° in the laboratory system which were too long to be accounted for by either deuteron or proton elastic recoils.

I. INTRODUCTION

THEORETICAL predictions which have been developed up to the present time concerning the angular distribution of neutrons elastically scattered by deuterons show a marked dependence on the type of nuclear forces assumed. Massey and Buckingham[1] have made extensive calculations concerning this problem below 11.5 Mev, and Verde[2] has recently published calculations concerning this problem for neutron energies up to 20 Mev. Since 1937 there have been several experimental determinations made of the angular distribution of $n-d$ elastic scattering.[3] Unfortunately, the discrepancies existing among these various experimental determinations of the angular distribution and the theoretical predictions are such that no definite conclusions can be drawn concerning the nature of the nuclear forces involved in $n-d$ elastic scattering. Rosenfeld[4] has presented an excellent summary of the experimental and theoretical work done on this problem up to 1948. The importance which this problem may have in the eventual understanding of the nature of nuclear forces makes it reasonable to accumulate more experimental evidence concerning $n-d$ elastic scattering.

II. APPARATUS

The high pressure cloud chamber used by Laughlin and Kruger[5] to measure $n-p$ scattering has been used to investigate $n-d$ scattering. A thin deuterium gas target bombarded by 10-Mev deuterons from the University of Illinois cyclotron served as a source of 12- to 13-Mev neutrons. The cloud chamber was filled with CD_4 and used D_2O and isopropyl alcohol as the vapor mixture. This served as a source of deuteron scattering centers as well as a detector of the deuteron recoils. Details of the cloud chamber, neutron source and collimator, and the resultant neutron spectrum may be found in reference 5.

The cloud chamber was surrounded by a temperature-controlled water-cooling system which was maintained at 21°C.

An Illex No. 3 Universal shutter was mounted directly in front of the camera lens to determine the exposure time. In case of shutter failure, caused by the excessive strains of solenoid operation, the shutter could be replaced without disturbing the camera lens position. The use of two General Electric A-H6 mercury arc lamps for chamber illumination, a coated $f/2.0$ Ektar 45 mm lens, and Linagraph Ortho 35 mm film gave satisfactory exposures at $f/3.5$ and 1/20 of a sec.

Neutrons were introduced into the cloud chamber only during expansion by placing a deuteron beam shutter made of sheet tungsten ahead of the thin deuterium gas target. At each expansion this beam

* This report is part of a thesis submitted in partial fulfillment of the requirements for the degree of Doctor of Philosophy at the University of Illinois, 1950. A preliminary report of these data was given in Phys. Rev. **77**, 748 (1950).

† Assisted by a joint program of the ONR and the AEC.

** Now at the Westinghouse Research Laboratories, East Pittsburgh, Pennsylvania.

[1] H. S. W. Massey and R. A. Buckingham, Phys. Rev. **71**, 558 (1947). R. A. Buckingham and H. S. W. Massey, Proc. Roy. Soc. **A179**, 123 (1941).

[2] Mario Verde, Helv. Phys. Acta. **22**, 339 (1949).

[3] Kruger, Shoupp, Watson, and Stallman, Phys. Rev. **52**, 678 (1937). H. Barschall and M. Kanner, Phys. Rev. **58**, 590 (1940). Coon, Davis, and Barschall, Phys. Rev. **70**, 104 (1946). J. Coon and H. Barschall, Phys. Rev. **70**, 592 (1946). J. Darby and J. Swan, Nature **161**, 22 (1948). J. Coon and R. Taschek, Phys. Rev. **76**, 710 (1949).

[4] L. Rosenfeld, *Nuclear Forces* (Interscience Publishers, Inc., New York, 1948), Chapter XIV.

[5] J. Laughlin and P. G. Kruger, Phys. Rev. **73**, 197 (1948). J. Laughlin, Ph.D. thesis, University of Illinois, 1947.