The 4n+1 radicactive series

English, Albert, C.

How to cite:

English, Albert, C. (1941) *The 4n+1 radicactive series*, Durham theses, Durham University. Available at Durham E-Theses Online: http://etheses.dur.ac.uk/9124/

The 4n+1 radioactive series.

by

Albert C. English.

Thesis submitted to Durham University for the degree of ~~PhD.~~ M. Sc.
1947.

Acknowledgements.

The work presented in this thesis was done at the Montreal laboratory of the National Research Council of Canada in the course of duties as an employee of the Department of Scientific and Industrial Research. Thanks are due to Dr.J.D.Cockroft, director of the establishment, for permission to use the work for this purpose and to Professor F.A.Paneth, head of the Chemistry Department.

For helpful advice and discussions during the research I am indebted to Professor F.A.Paneth, Dr.B.L.Goldschmidt, Dr.A.N.May and many other members of this team.

I would particularly thank Dr.A.N.May and his colleagues for their help in the physical aspects of the work and permission to use some of their equipment, and wish to point out that they have made important contributions to the results reported here.

Mr.P.Demers[★] made the investigation on tracks in the emulsion of the photographic plate that first showed the presence of four α-disintegrations in the series after the radium isotope.

Mr.E.P.Hincks and Mr.T.E.Cranshaw,[★] working on the pulse-analyser, made the early measurements of α-particle energies.

Mr.T.E.Cranshaw measured the period of 85^{217}[ø] and made further energy measurements of the α-particles in the series[★].

Mr.J.V.Jelley[★] showed the presence of β-α- "coincidences" in the series.

Dr.A.G.Maddock[X] of the Chemistry Division made a search for an emanation in the series using a vacuum technique.

[★] Physics Monthly Reports, Dec. 1944 to April 1945.

[ø] Report MP 162

[X] Chemistry Monthly Reports, Jan. and April, 1945.

Abstract

The disintegration products of U^{233} have been investigated and the main route of the 4n+1 series outlined as follows:

$$\underset{162,000y}{U^{233}} \xrightarrow{\alpha} \underset{\sim 10^{4}y}{Th^{229}} \xrightarrow{\alpha} \underset{14d}{Ra^{225}} \xrightarrow{\beta} \underset{10d}{Ac^{225}} \xrightarrow{\alpha} \underset{5m}{87^{221}} \xrightarrow{\alpha} \underset{2.1\,.\,10^{-2}s}{85^{217}}$$

$$\xrightarrow{\alpha} \underset{46m}{Bi^{213}} \xrightarrow{\beta} \underset{?\,10^{-5}s}{Po^{213}} \xrightarrow{\alpha} \underset{3.3h}{Pb^{209}} \xrightarrow{\beta} \boxed{Bi^{209}}$$

To face page 2

4n + 1 series

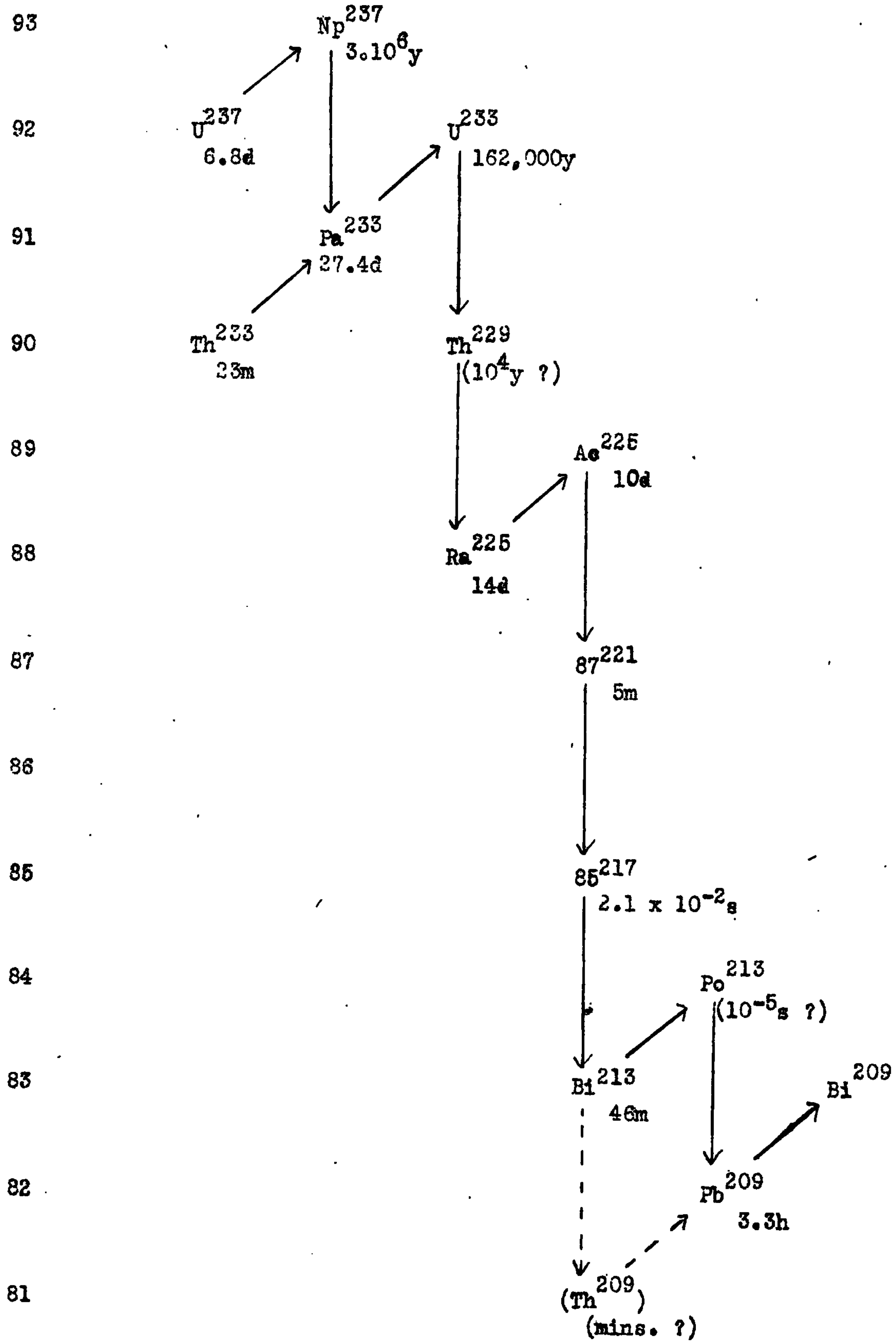


FIG. I

DIAGRAMMATIC SUMMARY.

For ease of reference the whole 4n + 1 series as it is known up to the present is charted in Fig. I. Vertical transitions are α-emissions and diagonal, β-emissions. Full lines denote the main sequence of the family and broken lines a branching which is not yet certain and is less than 3% of the main route.

Table I summarizes the data on the series. Figures marked (ø) are by T.E. Cranshaw.

TABLE I

		Half-life	α-energy Mev	β-energy Mev	Mode of Formation
Th^{233}	β	23.5m(5)		1.6(5)	Th^{232}-n-γ(2,3,4,5) Th^{232}-d-p(10)
U^{237}	β	6.3d(9)		0.26(8)	U^{238}-n-2n(7,8,9)
Np^{237}	α	3.10^{6}y(9) 6.10^{6}y(21)	?		U^{237} β (9)
Pa^{233}	β	27.4d(3)		0.58 (4%)(29) 0.27 (60%) < 0.27 (36%)	Th^{233} β (3,4,5) Th-d-n (10)
U^{233}	α	162,000y(6)	4.812±0.015(20)		Pa^{233} β (5)
Th^{229}	α	∼10^{4}y	∼ 5		U^{233} α
Ra^{225}	β	∼ 14d		very low < 0.05	Th^{229} α
Ac^{225}	α	10d	5.81		Ra^{225} β
87^{221}	α	5m	(6.1? (0) (6.33		Ac^{225} α
85^{217}	α	2.1 10^{-2}s (0)	6.97 (0)		87^{221} α
Bi^{213}	(α)? β	46m	∼ 6 ?	∼ 1.3	85^{217} α
Po^{213}	α	(? 10^{-5}s)	8.28(0)		Bi^{213} β
Pb^{209}	β	3.3h(15)		0.70(13)	Po^{213} α Pb^{208} -d-p (11,12,13,14) Pb^{208}-n-γ } (15) Bi^{209}-n-p }

INTRODUCTION

Three radioactive series are known in nature, the thorium series (mass number 4n where n is an integer), the uranium series (4n + 2) and the actinium series (4n + 3).

In 1923 and 1924, at a time when the mass assignment of only the thorium and uranium series was known for certain, Widdowson and Russell (19,26) deduced that there were probably only four independent disintegration series of which three were known, and they predicted for the fourth, the 4n + 1, a route passing through an isotope of 89 and the "missing" elements 87 and 85 to end in Bi^{209}. An unsuccessful search was made for an emanation of the unknown family.

A few members of the new series had been prepared by 1940 and Turner(18) used the regularities in the distribution of isotopes to make some quite accurate predictions on the disintegration sequence.

In Br 499, August 1944, N. Feather, using the regularities of Geiger-Nuttall relationships for the isotopes of an element, critically examined the 4n + 1 series and deduced probable life-times and energies for the possible disintegration products after U^{233}.

Of the 4n + 1 series, no radioactive member has been found in nature but several have been prepared artificially. The first was Th^{233} in 1935 by Fermi et al.(1), a β-emitter of 23 minutes half-life formed in the reaction $_{90}Th^{232}$ $\underline{(n,\gamma)}$ $_{90}Th^{233}$. Other workers have investigated this substance (24,25,2,3) and it has been produced by the reaction $_{90}Th^{232}$ $\underline{(d,p)}$ $_{90}Th^{233}$ by Gofman and Seaborg (10).

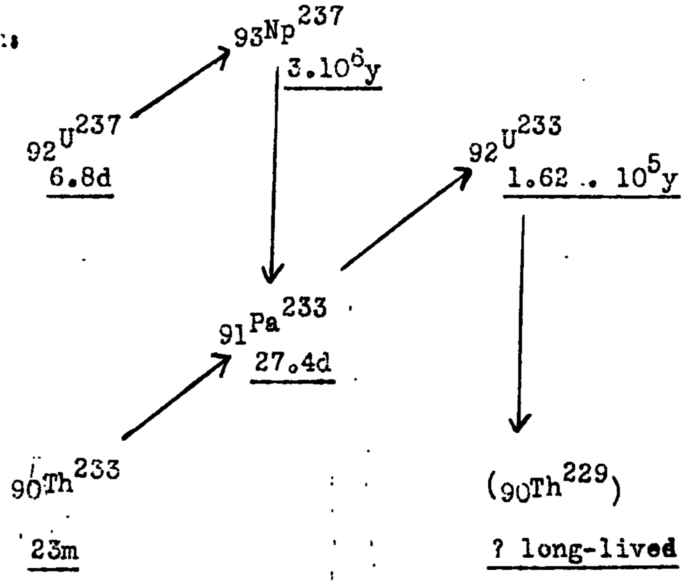
Daughter of Th^{233} is Pa^{233}, 27.4 day β-emitter which was discovered by Meitner, Strassman and Hahn (2). The doubts which later arose about these two substances from the discovery of fission by Hahn and Strassman were cleared up by the researches of V. Grosse, Booth and Dunning (3) and Seaborg, Gofman and Kennedy (4). Gofman and Seaborg (10) have shown the production of Pa^{233} by the process ${}_{90}Th^{232}(\underline{d,n})\ {}_{91}Pa^{233}$.

U^{233}, derived from the decay of Pa^{233}, was first separated and examined by Seaborg, Gofman and Stoughton (5) and it is on the disintegration products of this substance that the work in this report is based, the production of milligram quantities of U^{233} by the irradiation of thorium with neutrons in a pile having made possible the investigation of the main part of the 4n+1 series. U^{233} is α-active and the latest figure for the half-life is 162,000y (6).

Another radioactive product of the 4n+1 type is U^{237} discovered by Nishina et al (7) and McMillan (8). It is produced in the reaction ${}_{92}U^{238}(\underline{n,2n})\ {}_{92}U^{237}$ and is a 6.8 day β-emitter.

The daughter of ${}_{92}U^{237}$, investigated by Wahl and Seaborg (9), is ${}_{93}Np^{237}$, an α-emitter (and therefore a parent of Pa^{233}) of $3 \cdot 10^6$y half-life.

The upper part of the 4n+1 series as it was known up to the time of the investigation given in this report may, therefore, be summarized in the following diagram:



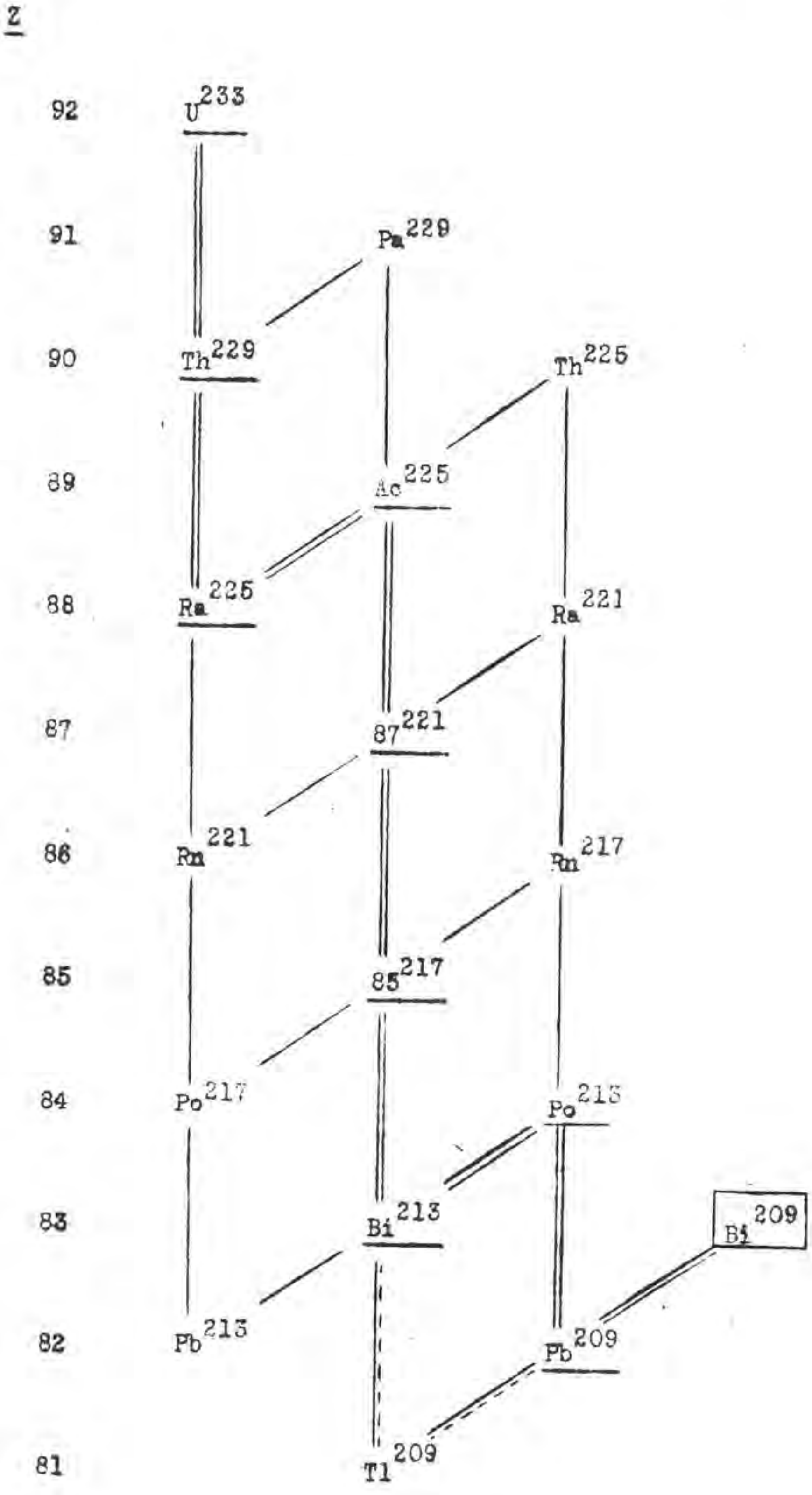

Z
92
91
90
89
88
87
86
85
84
83
82
81
U233
Pa229
Th229
Th225
Ac225
Ra225
Ra221
87221
Rn221
Rn217
85217
Po217
Po213
Bi213
Bi209
Pb213
Pb209
Tl209

FIG. 3

In addition to these substances, another 4n + 1 body has been made. Pb^{209} is a 3.3 hr β-emitter discovered by Thornton and Cork (11). It is produced in the reactions:

$$_{82}Pb^{208}(\underline{d,p})\ _{82}Pb^{209}\ (11,12,13,14),$$

$$_{82}Pb^{208}(\underline{n,\gamma})\ _{82}Pb^{209}\ (15),$$

$$_{83}Bi^{209}(\underline{n,p})\ _{82}Pb^{209}\ (15).$$

Pb^{209} decays to Bi^{209}, the stable isotope of natural bismuth.

The disintegration products of U^{233}.

The purpose of the work reported here was to trace the route of the 4n + 1 series beyond U^{233}. The search for the disintegration products had to be guided by the fact that the daughter of U^{233} is long-lived since there is no obvious growth of α- or β-activity in the substance, and the problem was, therefore, the detection and extraction of very small activities from a comparatively highly active sample of U^{233}.

With the usual assumption that α- and β-emissions are the only transitions in the series, the map of the probable alternative routes of disintegrations following U^{233} has been drawn in Fig. 3, where the limits given are the same as those used by Feather (Br 499) and are dependent mainly upon considerations of β-stability of nuclei. Frequent reference to the diagram will be made in this report and the route finally derived from the evidence is shown by double lines.

U^{233}

The sample of U^{233} used in this work was extracted by Dr. Goldschmidt and colleagues[X] in October 1944 from a number of thorium carbonate slugs irradiated in the Site X pile. The uranium, about 23 mg, contained in some tens of grams of thorium, was passed on to me for final purification. A diethyldithiocarbamate extraction in the presence of tartrate and at pH 6 was first performed to separate the main bulk of the thorium, and then a fluoride precipitation to take out the few milligrams carried over. The purification was continued by a fluoride precipitation with 1 mg of La and a final peroxide precipitation.

The precipitate was dissolved in the minimum of nitric acid and made up to 25 ml. Total α-counts in the solution was 238.10^6/min in the efficiency of our α-counter which is about 50%. For a half-life of 162,000y (6) for U^{233} this would mean about 22.8 mg.

In the early part of the work, 10 ml of this solution or about 9 mg was used and later, 12.5 ml or about 11.4 mg.

The isotopic purity of this sample of U^{233} has not been measured but presumably it is very similar to that of other specimens obtained in the same way for which a recent figure is 95.8 ± 0.5%.[*]

No β-counts were visible in the end-on Geiger-Mueller counter in a sample of 2000 α-counts/min.

X Chemistry Monthly Reports, October 1944.

* MUC - AJD - 59. (Dempster) March 1945.

PRELIMINARY NOTE ON INTERFERENCE BY THE NATURAL THORIUM FAMILY

Two months after the beginning of this work, information was received* from Seaborg's team that the U^{233} extracted from neutron bombarded thorium contained a certain percentage of the isotope U^{232}, an α-emitter of about 30 years half-life (10) which is also produced in the irradiation, possibly by a U^{233} (n, 2n) reaction. ${}_{92}U^{232}$ decays to ${}_{90}RdTh^{228}$, 1.9y α-emitter of the natural thorium series and parent of ThX and the rest of the series down.

The figure given for U^{232} contamination was one α-particle from U^{232} in 2000 from U^{233}. The half-life of Th^{229}, the daughter of U^{233}, is very long since samples of U^{233} show no change in counting rate in a long time so that the ratio between activities in the thorium series and the new series after the thorium isotope can, conceivably, be of the order of unity.

It will be shown that extracts of a radium isotope from our sample of U^{233} always contain ThX, 3.6 day parent of six radioactive shorter-lived products, a fact which complicates the investigation and necessitates some delay before reliable work can be done on the series.

The early work was performed in ignorance of the above facts and it is fortunate that the results are not invalidated simply because the time during which some samples were kept for decay measurements, before further examination, was sufficiently long (over ten days) to reduce the ThX contamination to a fraction of the new series activity.

RADIUM AND ACTINIUM ISOTOPES.

Extraction of a Radium Isotope.

The search for a disintegration product of U^{233} was begun at the radium isotope. Because of the considerable difficulties involved in the use of

---

* Letter from Huffman to Cockcroft - E.J.R.H. 52, Jan. 25th, 1945.

the usual $BaSO_4$ method due to strong adsorption effects and the recalcitrant nature of the precipitate, it was decided to use the precipitation of $Ba(NO_3)_2$ in fuming nitric acid solution.

The method was tested and found to be most efficient in the separation from uranium. The fraction of uranium carried down by 1 mg. of $Ba(NO_3)_2$ from tracer quantities of U was less than 1/5000 so that two or three cycles would be sufficient for the elimination of several mg. of U^{233}.

The $Ba(NO_3)_2$ precipitation is recognized as a reliable separation for radium from most of the elements of the top of the periodic table except lead. In addition to uranium, actinium and thorium are not carried down, a fact confirmed for tracer quantities during this work since neither the radiothorium known to be present in the U^{233} solution nor the actinium isotope shown to be present in the 4n+1 series have appeared in the separation.

Protactinium in one experiment followed the barium nitrate precipitate to the extent of 10%. Element 87, being an alkali metal, can be expected to remain in the solution and an emanation could not interfere. The method, therefore seems to be reliable for radium in the presence of, at least, elements from 86 to 92 with some care needed in the case of $_{91}Pa$.

For the first extraction attempted, about 18 days after the purification of the U^{233}, 10 ml. of the solution containing about 9 mg. U^{233} was evaporated with 2.5 mg. $Ba(NO_3)_2$ to a volume of about 0.1 ml. with nitric acid in a 3 ml. centrifuge tube and treated, cold, with

To face page 10

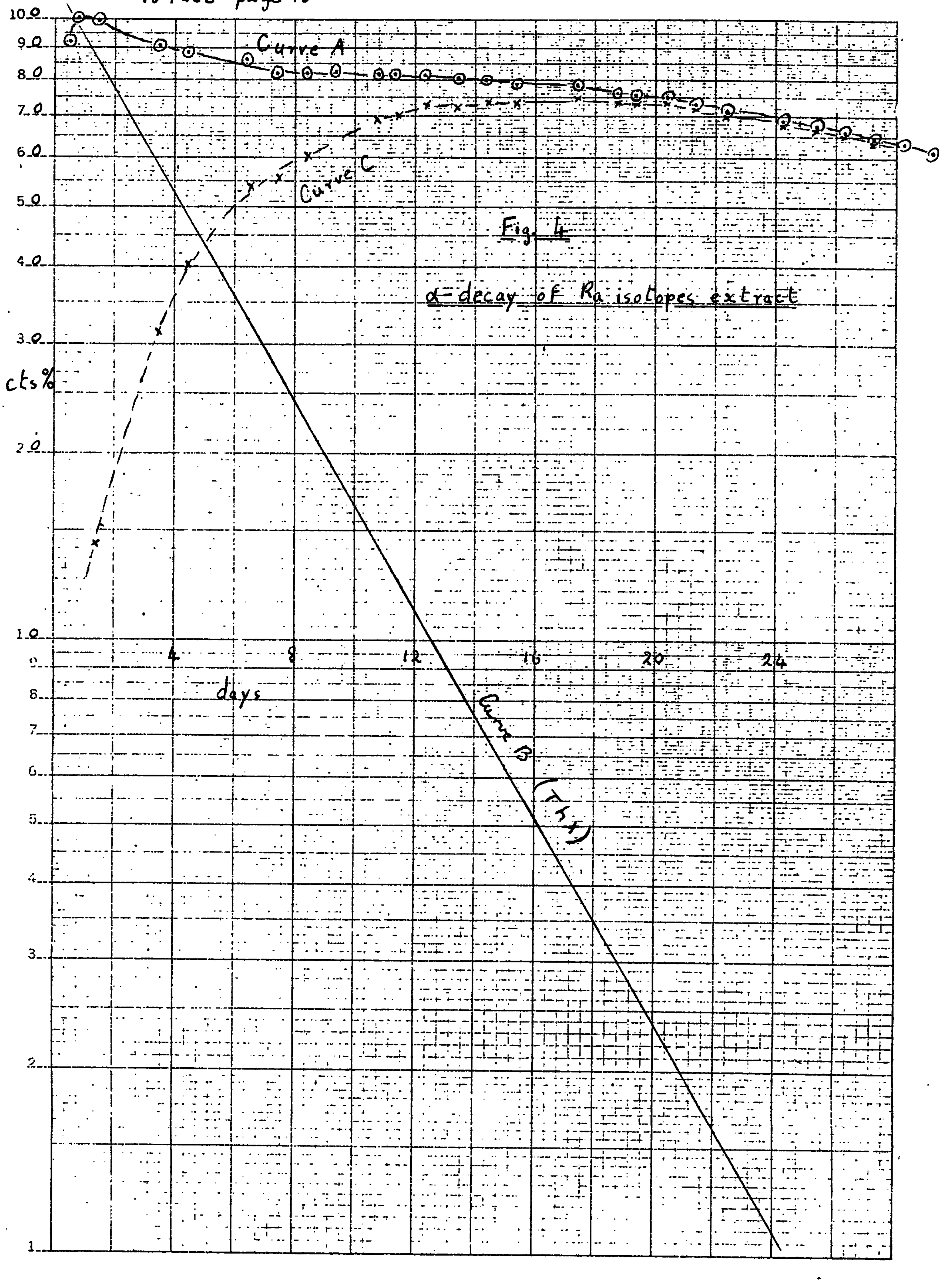


Fig. 4

α-decay of Ra isotopes extract

0.5 ml. fuming nitric acid. The precipitate was separated, washed three times with portions of fuming nitric acid, redissolved in water and transferred to a clean tube.

The process was repeated and the first filtrate from the second precipitation was found to contain only 2000 α-counts/min., indicating that contamination of the precipitate by U^{233} should now be about 1/5000th of 2000 counts or less than 1 count/min. The precipitate was washed twice, transferred to a gold counting-tray and evaporated to dryness with nitric acid. Yield of $Ba(NO_3)_2$ in the operations was in this case 80%.

This became the standard procedure for the subsequent extractions made at intervals of a few weeks.

First evidence of radioactive disintegration products of U^{233}.

The first barium extract obtained showed an α-count of 270/min and exhibited a complex decay curve having periods of the order of days.

As previously explained, a fresh extract of $Ba(NO_3)_2$ from our sample of U^{233} ($+U^{232}$) probably contains ThX. The α-activity decay curve of one of the later, more active extracts is shown in Fig. 4, Curve A, where it is clear that there is a shorter lived component which may be ThX and, from the shape of the curve, some other portion which is not a simple exponential decay since there is a distinct inflexion in the curve.

This was the first qualitative evidence that some other activity besides the possible ThX could be extracted from the specimen of U^{233}.

Detection of four related α-disintegrations.

When the first extract was ten days old, a small portion was examined by Mr. P. Demers by deposition in the emulsion of a photographic plate.*

---

* Physics Monthly reports - Dec. 1944 and Jan. 1945.

He was able to show the presence of four track 'stars' in the emulsion of the plate from four α-particles originating in the same nucleus, and that there was no period between the first and last α-emission which was of the order of one day or greater. Demers found that the energy distribution of the four α-particles was not the same as that of the ThX series particles although we now knew that probably up to 20% of the stars were due to ThX. If these four α-emissions belong to the new series and follow a radium isotope, it would seem from Fig. 3 that all the required α-particles at the end of the series have been found.

Mr. E.P. Hincks and Mr. T.E. Cranshaw measured[*] the energies of the four α-particles in the high pressure ionisation chamber and pulse analyser and the latest measurements by Cranshaw and myself give energies of 5.81, 6.33, 6.97 and 8.28 for the four main peaks seen in the pulse analyser. These energies are different from any of the three known families.

Detection of an actinium isotope in the series and the β-activity of the radium.

From the evidence given up to this page, it is not clear whether the radium isotope extracted is α- or β-active or both and the importance of the ThX contamination has not been established. The following experiment was the first to indicate the solution to these problems and the resolution of the decay curve given in Fig. 4 (A).

A barium nitrate extract was allowed to decay for 12 days and then reprecipitated by the fuming nitric acid method. It was found that only part of the α-activity came down with the precipitate while the main activity in the filtrate could be carried down by a precipitation of lanthanum fluoride, indicating an actinium or thorium isotope (see Fig. 3) and no other element from at least 86 to 92. This fraction showed a simple exponential decay of about 10 days while the barium nitrate fraction showed a regrowth

---

[*] Physics monthly reports - Dec. 1944, Jan. 1945.

of α-activity.

To differentiate between the two possibilities of actinium and thorium, a precipitation of the activity on ceric iodate in N nitric acid was attempted and it was found that only a small percentage appeared with the precipitate showing that the substance is an actinium rather than a thorium isotope. It is known that actinium can be electrolysed onto the cathode in weakly acid conditions and this phenomenon has been observed with the new actinium during a long series of electrolyses of a nitric acid solution. After several hours, the acid had been reduced to such an extent that the actinium was able to deposit.

A specimen of the actinium examined in the large ionisation chamber and pulse analyser showed the presence of all four of the new α-emitters originally detected in the radium isotope. As will be shown when the series is considered as a whole, the Geiger Nuttall relationship between energy and half-life indicates a period of some days for the lowest energy α-emitter (5.81 Mev) and since this is always present in a fresh precipitation of the new actinium, it probably belongs to this member. There is further evidence later.

If we may anticipate the more definite proof that these new activities are really in the 4n + 1 series by the identification of the already known Pb^{209} as a member, there is now substantial evidence for believing that the radium and actinium isotopes concerned are Ra^{225} and Ac^{225}, that Ra^{225} is at least partially β-active and that Ac^{225} is α-active with a period of about 10 days. From Fig. 3, only these isotopes of Ra and Ac are available as the progenitors of four consecutive α-particles and any abnormal route outside the limits of Fig. 3 is made most unlikely by the presence of Pb^{209}. The

To face page 13

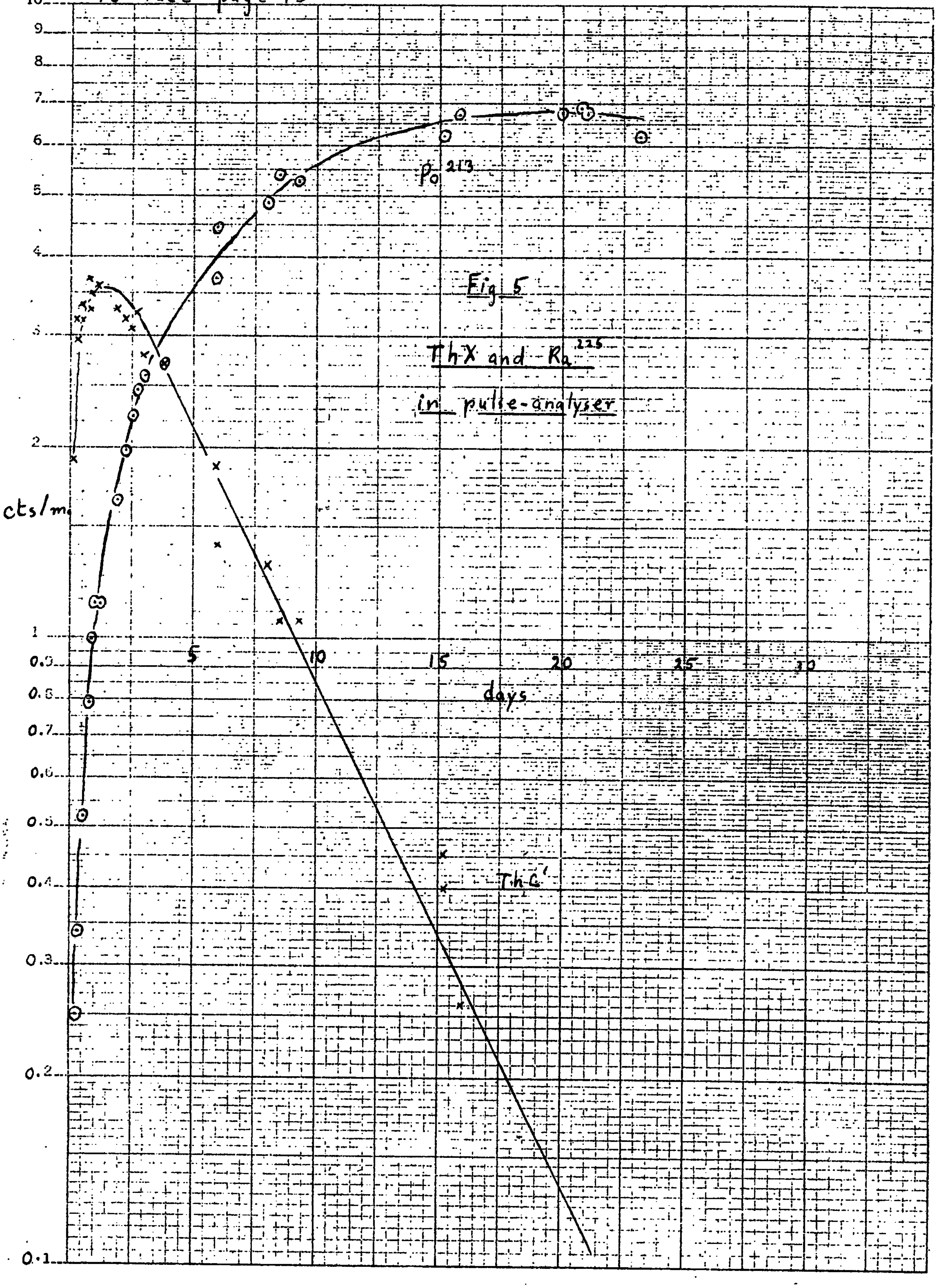


Fig 5

ThX and Ra^{225}

in pulse-analyser

names of Ra^{225} and Ac^{225} will be used from now on to simplify the discussion.

Since the growth of Ac^{225} and products from Ra^{225} is not clearly visible in the α-decay curve given in Fig. 4 (A) it is possible that it is almost completely masked by the decay of ThX. Confirmation of this was obtained in the following experiment where the exact nature of the radium isotopes extract is determined.

α-Analysis of the $Ba(NO_3)_2$ extract.

A part of a fresh $Ba(NO_3)_2$ precipitate from the U^{233} solution, containing about 50 α-counts/min, was spread on a piece of platinum foil about 2 cm^2 and examined in the large ionisation chamber and pulse analyser. The energy range was chosen which includes the α-particle of ThC' at 8.78 Mev and the highest energy member of the new series at 8.28 Mev (later shown to be Po^{213}), and the variation in the α-distribution with time was followed.

At the beginning only the peak of ThC' was present and this showed a preliminary growth as the 10.6 hr ThB attained equilibrium and then decayed with approximately the period of ThX, 3.6 days (Fig. 5).

The high energy α-partcle of the new series was not visible at the start but grew in slowly with a period of the order of days, reaching the maximum in about 18 days (Fig.5). Since we will show that the longest period between the first α-particle of Ac^{225} and this α-particle of Po^{213} is 46 minutes, there seems no doubt that the growth seen here is that of Ac^{225} not originally present in the sample and, at this stage, it seems not unlikely that the Ra^{225} is almost completely β-active since we may expect Rn^{221}, Po^{217} and Pb^{213} (Fig. 3) to be quite short-lived (Feather, Br 499) and therefore to provide no long-lived barrier before Po^{213}.

To face page 14

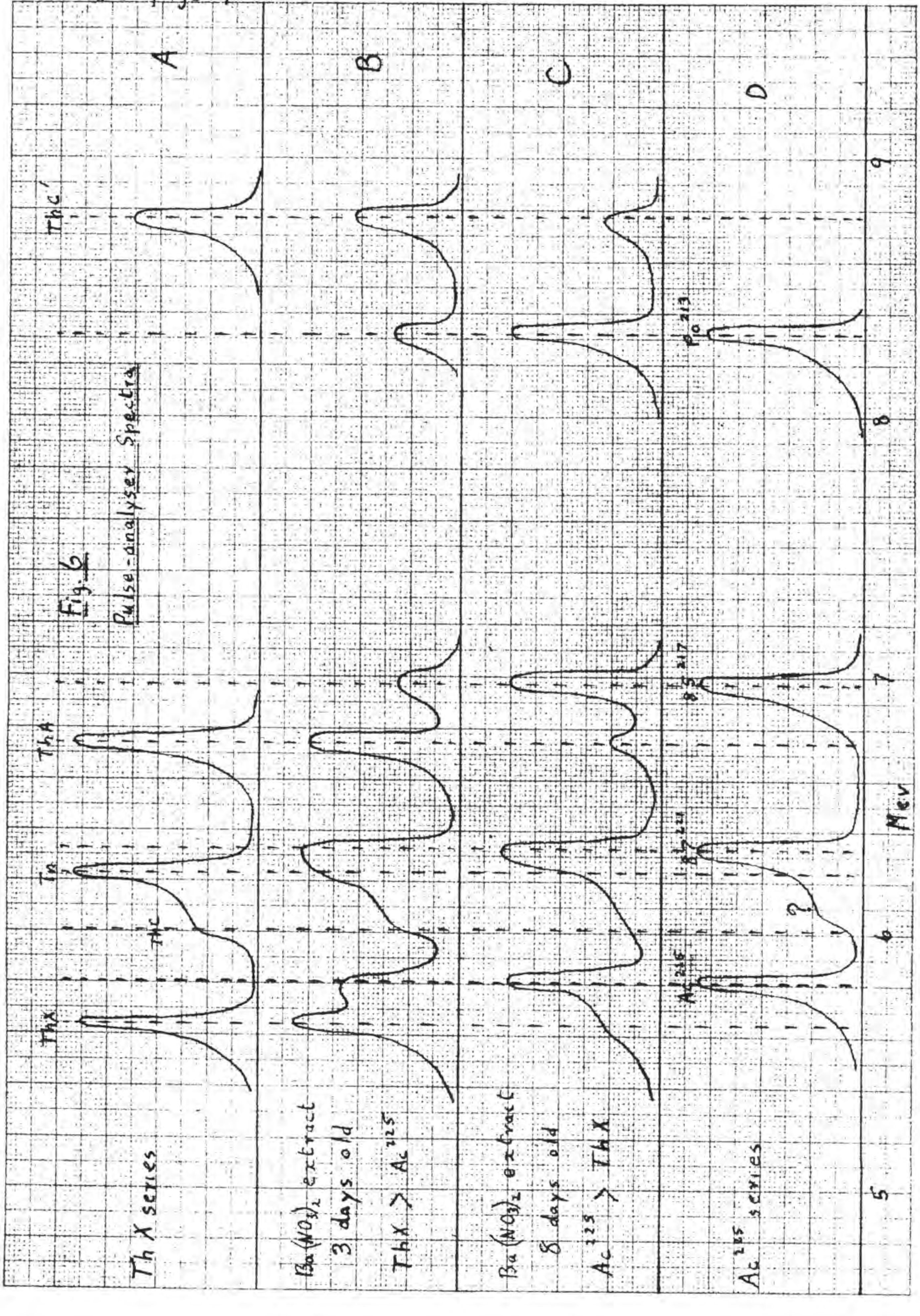


Fig. 6

Pulse-analyser Spectra

It is now possible to attempt an analysis of the decay curve A in Fig. 4. Assuming all the original α-activity to belong to ThX and products, a correction curve B may be drawn and the difference between the two curves plotted as curve C. The resultant shape is seen to be much the same as obtained in the pulse analyser and expected from the growth of Ac^{225}.

In the examination in the pulse analyser of the $Ba(NO_3)_2$ extract, some measurements were made over the whole energy range of the four new α-emitters and those of ThX and products and the transformation of the activity spectrum with time can be seen in Fig. 6 where a comparison is made between,

(A) the spectrum of a sample of ThX,
(B) the extract of radium isotopes after 3 days,
(C) the extract after 8 days and
(D) a pure sample of Ac^{225}

The drawings are a somewhat qualitative combination of a large number of measurements, and for clarity and future reference the peaks have been identified according to our present convictions.

Confirmation of Ra^{225} β-activity.

An extract of radium isotopes on barium nitrate, in which the ThX had decayed to a very small amount, was dissolved in dilute acid and the Ac^{225} removed by a lanthanum fluoride precipitation. The filtrate was evaporated with hydrochloric acid several times to remove nitric acid, 2 mg of lead added and precipitated with hydrogen sulphide for the purpose of removing the 46 min and 3 hr activities (Bi^{213} and Pb^{209}) which are considered later in this report. The barium nitrate was then separated by the fuming nitric acid technique as a final purification and transferred to a gold tray.

To face page 15

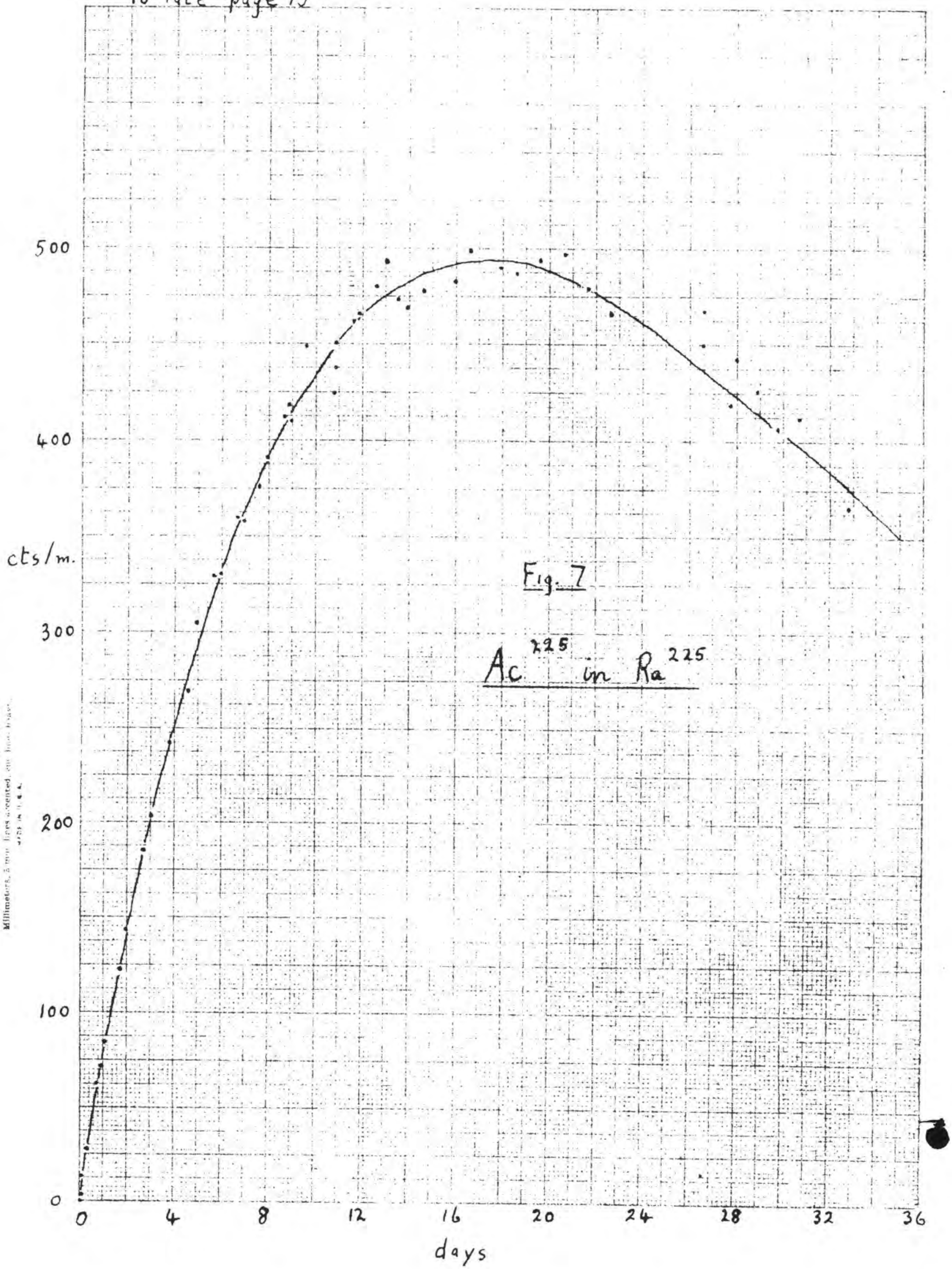


Fig. 7

Ac^{225} in Ra^{225}

The α-count of the sample in the ordinary α-counter immediately after the separation was 13 counts/min and this seems to be completely accounted for as residual ThX.* The growth of α-activity in the sample is shown in Fig. 7 in which the correction for residual ThX has been made.

Thus it is possible to prepare a radium extract, showing very little α-activity if any, from which grows the α-active actinium isotope. In a later section, further work on this sample is reported and the properties of Ra^{225} are examined in more detail, but it may be mentioned here that the β-activity of Ra^{225}, up to the present shown merely by the absence of α-activity and the growth of an actinium, is not easily detected since it has a very low energy.

EMANATION

Since Ra^{225} is β-active, the main sequence does not pass through Rn^{221}, nor, as will be seen later, through Rn^{217} (Fig. 3). A search has been made for an emanation which may be reported here as contributing to the evidence.

---

* The extract of Ra^{225} and ThX, when first separated from the U^{233} on about 2 mg of barium nitrate, had shown a maximum α-count of 3,900/min. (ThX and products). Before being used in this experiment, it had undergone three precipitations at various times so that it had probably lost a fraction of its weight. α-activity in the sample due to ThX and products at the time of this experiment (26 days after the first extraction) would, therefore, be somewhat less than

$$3900 \times e^{-\frac{(0.693 \times 26)}{3.64}} = 28 \text{ counts/min.}$$

and there is a further factor 3/4 since the last α-emitters, ThC + C', would not be present after a purification of the radium isotope,

$$= 21 \text{ counts/min.}$$

13 counts/min = 60% of the amount expected from total original ThX. (All counts in 50% geometry).

The evaporation to dryness of a solution of Ra^{225} would cause the loss of an emanation if one existed and the regrowth of the activity would be visible if the period were sufficiently long. Experiments of this kind gave completely negative results setting a lower limit to the life-time of an emanation at about 2 minutes.

Dr. A.G. Maddock* made an unsuccessful search for an emanation using a vacuum technique believed to be efficient for a lifetime greater than 10^{-1} sec.

None of these attempts would have been sufficiently sensitive to detect a 1% branching of Ra^{225}.

LEAD ISOTOPE

Detection of Pb^{209}.

It is most convenient now to consider the work done at the end of the series on the isotopes of lead and bismuth since the evidence on the intermediate members may be considered somewhat meagre without reference to the whole series.

Pb^{209} is already known as a 3.3 Hr β-emitter (11,12,13,14,15) having been produced in several artificial radioactive reactions from Pb^{208} and Bi^{209} so that its detection as a disintegration product of the radium and actinium isotopes extracted from U^{233} makes Fig. 3 and the deductions therefrom more convincing.

A number of sulphide precipitations made on solutions of the series during the investigation on the bismuth isotope (next section) were examined in the β-counter and it was found that after the decay of the bismuth, a residue remained, free from short-lived α-activity, which decayed with a

---

* Chemistry Division monthly reports - Jan. and April, 1945.

To face page 17

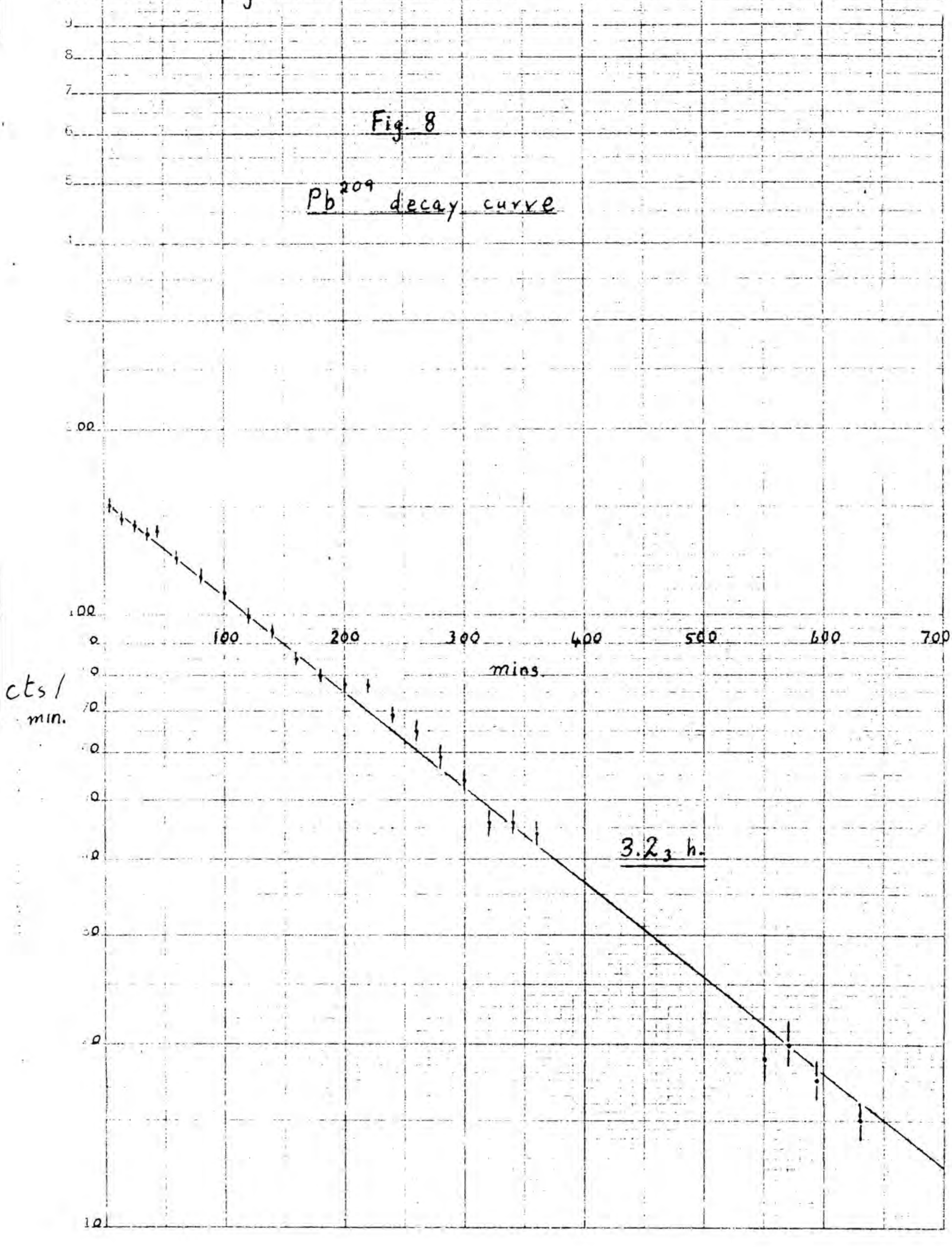


Fig. 8

Pb^{209} decay curve

period of approximately 3 hours.

During experiments on another member of the series, an isotope of element 87, activities were volatilised at red heat from Ac^{225} and condensed on counting trays. The β-activity was found under these conditions (agreeing with the behaviour of lead) and more clearly since the condensed material was singularly free from long-lived contamination of Ac^{225} so that in these cases, after sufficient time, the β-radiation was unaccompanied by any α-particles and must, therefore, come after all the known α-emitters.

It remained to separate the activity, in particular from the bismuth isotope which is both β- and α-active and has a period of 46 minutes, by a recognised method for lead and to measure its half-life with some accuracy.

2 mg of lead nitrate was added to a nitric acid solution of Ac^{225} containing about 50% alcohol and the precipitation effected by drops of dilute hydrochloric acid. The crystalline lead chloride was separated and washed, counted first in the α-counter and then its decay followed in the β-counter.*

The corrected decay curve is shown in Fig. 8 and gives a figure of 3.2 hours for the half-life.

This experiment also indicates the absence of a Pb^{213} (Fig.3) of half-life greater than a few minutes.

---

* The contamination of the sample was 40 α-counts/min (10 per member) remaining constant during the measurement and the correction to be applied to the β-counts for this contamination (∼ 20 counts) plus the background of the counter was 35 counts/min from an initial count of 190/min.

BISMUTH AND POLONIUM ISOTOPES

Detection of a bismuth isotope in the series.

It has been found possible to separate by various methods from a solution of Ac^{225} a substance which emits both β and α particles in approximately equal proportions and decays with a period of 46 minutes. On examination by the pulse -analyser, it is found that the only main α-peak present is one at 8.28 Mev which, by the Geiger Nuttall rule, should be extremely short-lived. This indicates that the 46 minute substance is β-active and that its daughter is α-active with a very short lifetime.

The chemical properties of the 46 minute substance have been examined very carefully since its identification is most important in defining the route of the series.

Chemical identification of the 46 minute activity.

The activity is coprecipitated with lead or bismuth sulphide in acid solution and it is volatilised at a dull red heat from a barium chloride precipitate and with more difficulty from lanthanum oxide. These properties do not clearly differentiate between Pb, Bi and Po which are the elements leading to the last α-particles of the series. (Fig.3)

A solution of Ac^{225} was made 1 N in nitric acid and salted with a fraction of a milligram of lead and bismuth carriers. An electrolysis performed at a potential of four volts between small gold electrodes, with vigorous stirring, deposited the activity on the cathode with the bismuth. Lead deposited on the anode and showed no α-activity; nor did any grow in subsequently as would be the case if Pb^{213}, having a suitable half-life, were present in any quantity. In solutions of the series uncontaminated by ThX, the only α-activity ever observed in electrolysis under conditions of 1 N acid was the 46 minute activity.

A silver electrode in a normal solution of hydrochloric acid takes up a potential which allows the deposition of polonium but not bismuth. In a solution of thorium containing some thousands of counts/min of ThC a silver foil collected no activity in five minutes while a solution of polonium (RaF) containing a few thousand counts/min deposited considerable activity in this time. In contact with a solution of Ac^{225} for 30 minutes, no activity deposited on the silver foil showing that the 46 minute substance is not an isotope of polonium.

A platinum foil in contact with a solution saturated with hydrogen gas reaches a potential sufficient to deposit both bismuth and polonium on its surface, and to a lesser extent, lead. From a solution of Ac^{225}, the 46 minute substance was deposited on the platinum and examination with the pulse-analyser again showed the presence of only one main α-peak at 8.28 Mev.

Bismuth metal foil displaces polonium from solution. A piece of bismuth was agitated in a solution of thorium nitrate in conditions under which hydrogen polarised platinum collected 5,000 α-counts/min of ThC in a few minutes whereas the bismuth showed only 10 counts/min, indicating that there is not appreciable interchange of ThC in solution with the surface of the bismuth. No activity appeared on the bismuth in contact with Ac^{225} solution again pointing to the absence of a sufficiently long-lived polonium.

On lead foil, which displaces ThC from a solution of thorium nitrate, the 46 minute activity deposited indicating either a bismuth or a polonium.

A precipitation which is most reliable as a test for a polonium is that of tellurium by stannous chloride. On testing the method with a solution of thorium nitrate, it was found that an appreciable percentage of ThC (Bi^{216}) was carried in the precipitation in the absence of bismuth hold-back carrier but that addition of a mg of bismuth was sufficient to reduce this to negligible proportions. The method was applied to a solution of Ac^{225} and no activity appeared with the tellurium.

As a final confirmation of the identity of this activity, an attempt was made to prove that it is an isotope of Bi by a fractional precipitation procedure. Since the period is comparatively short, no long fractional crystallisation could be attempted and the activities available do not permit great accuracy.

A method was chosen, therefore, which would allow reasonable speed of operation and effect good discrimination between bismuth on the one hand and lead and polonium on the other.

Both lead and polonium oxalates are soluble in acid solution while bismuth oxalate precipitates. A precipitation of bismuth oxalate in the presence of tracer polonium carried 20% of the activity. Thus it might be expected that a fractional precipitation of bismuth oxalate would divide polonium in the ratio 10% to 90% between precipitate and filtrate if the bismuth ratio were 50% to 50%.

In conjunction with a sulphide precipitation in acid condition performed on both the precipitate and filtrate from a fractional oxalate separation, it should be possible to distinguish very clearly between bismuth and all other elements at least from 81 to 89.

A sample of BaCl containing Ac^{225} was dissolved in normal hydrochloric acid and approximately 1 mg of bismuth as nitrate added to the solution.

Oxalic acid was added and a precipitate of bismuth oxalate formed slowly. (Presumably some Ac^{225} was carried by the precipitate but would not interfere finally).

The precipitate was separated quickly in the centrifuge, dissolved in drops of concentrated hydrochloric acid and diluted somewhat. The original centrifugate and this solution were then treated with hydrogen sulphide and the precipitates of bismuth sulphide washed, transferred to weighed trays and dried on a hot plate. The decay of the α-activity of each was followed and the samples weighed afterwards.

The α-activity of the samples was used rather than the β-activity partly because the backgrounds of our β-counters are high and their efficiency low, and partly because complications would arise from the presence of Pb^{209}, the three hour β-emitter, in one fraction. Self absorption of the α-rays in the sample would be expected to increase the error of the experiment but since the two portions were not too dissimilar in weight and the distribution on the trays about the same, the effect should not be great.

Fraction A received only a sulphide precipitation
" B " an oxalate and a sulphide precipitation

Wt. of A - 0.73 ± 0.02 mg.
Wt. of B - 0.65 ± 0.02 mg.

Ratio of weights - 1.12 ± 0.06

α-count of A at zero time - 45 ± 5 counts/min.
α-count of B " " " - 50 ± 5 counts/min.

Ratio of counts - 0.9 ± 0.2

$$\frac{\text{Ratio of counts}}{\text{Ratio of weights}} = 0.8 \pm 0.2 \text{ (all errors given are extremes)}$$

The theoretical value for a bismuth isotope is unity while other elements from 81 to 89 should give values of 9 or more or alternatively not appear at all. The result is as good as can be expected under the circumstances and some correction for self-absorption of α-particles in the heavier fraction would tend to improve it.

The following table summarizes the reactions of the 46 minute activity.

TABLE III

46 minute activity

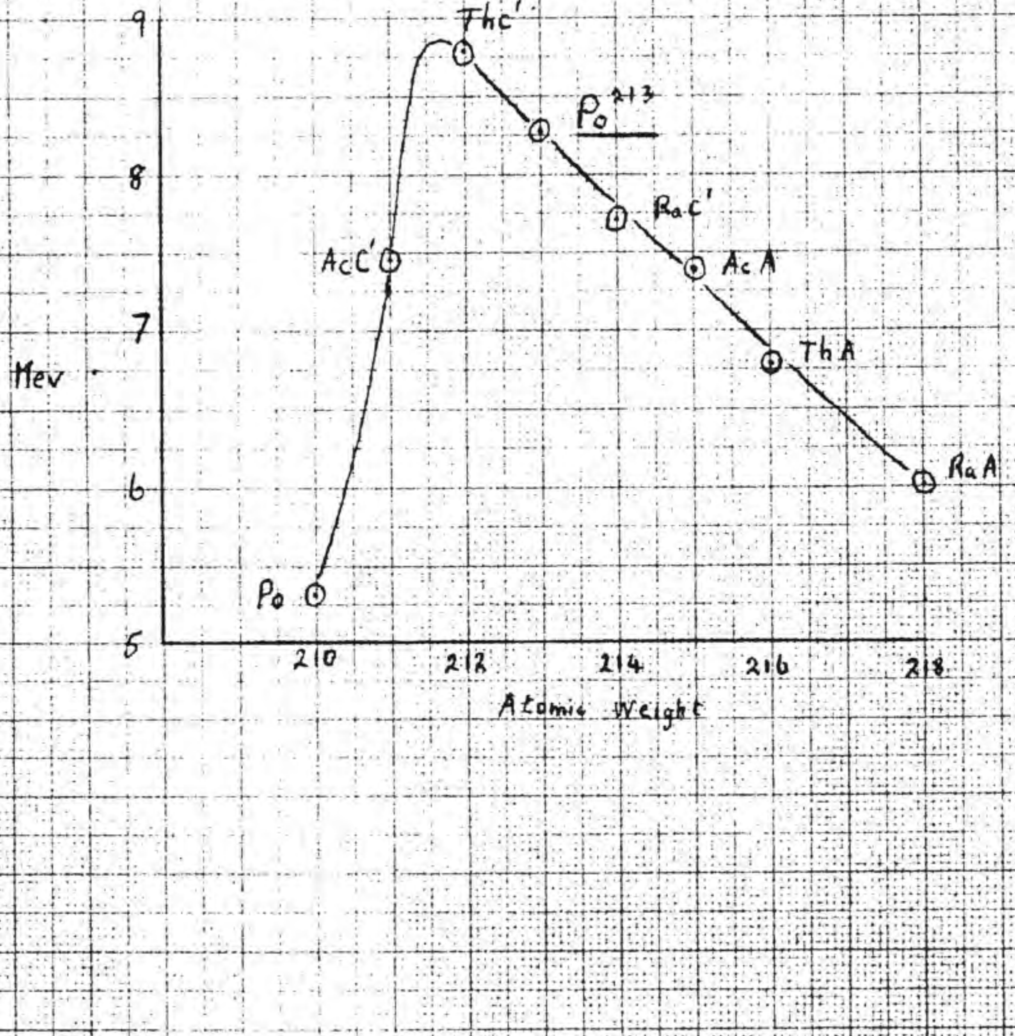
Method	Activity Separated	No Separation	Element indicated (of Bi, Pb & Po)
electrolysis cathode electrolysis anode	X	 X	Bi or Po, not Pb
Ag electrode + HCl		X	not Po
Pt + H_2 electrode	X		Pb, Bi or Po
Pb electrode	X		Bi or Po, not Pb
Bi electrode		X	not Po
H_2S precipitation	X		Pb, Bi or Po
Te by $SnCl_2$		X	not Po
"Isotopy test"			Bi

It seems to be fairly well established that there is a bismuth isotope in the series, β-active and leading to the last α-emitter in the series, (the other two after Ac^{225} are both sufficiently short-lived to be seen if derived from the 46 minute bismuth) and there seems little doubt that

To face page 23

Fig. 9

α-particle energy of Po isotopes



the isotope is Bi^{213}, parent of Po^{213} (fig. 3). Further work, on the β-radiation of Bi^{213} and the possibility of branching, will be given later.

Dr. A.N. May pointed out that β-α "coincidences" could be expected in the series from Bi^{213} and Po^{213} and Mr. J.V. Jelley confirmed this experimentally.*

Polonium^{213}

Dr. A.N. May's section has provided physical evidence which makes the identification of Po^{213} much more certain. Cranshaw's measurement of the energy of the last α-particle of the series gives 8.28 Mev and Fig. 9 (due to Dr. May, unpublished report) shows that this energy fits very well for Po^{213} in the graph of the various isotopes of polonium plotted against the energies of α-particles.

87 AND 85 ISOTOPES

The order of the four α-particles

It is useful here to note the evidence on the order of the α-disintegrations. Cranshaw[X], in his work on the period of the substance we now identify as 85^{217}, photographed α-particle pulses on the screen of an oscilloscope using an electrical circuit whereby the trace of the oscilloscope was triggered by a pulse, so that another α-particle following the first within a period of 1/20th of a second would be seen displaced a certain distance along the trace. He showed that the α-particle of 6.97 Mev followed shortly after that of 6.33 Mev.

The work reported above on Bi^{213} shows that the α-particle of 8.28 Mev comes last in the series and the Geiger Nuttall rule and the experiments of the next section indicate that the α-particle of 5.81 Mev belongs to Ac^{225}.

---

* Physics monthly report - March 1945.

X M.P. 162 and Physics Monthly reports / February, March and April 1945.

Thus the order of the α-particles seems to be 5.81, 6.33, 6.97 and 8.28; and they must be assigned in order to Ac^{225}, 87^{221}, 85^{217} and Po^{213} (Fig. 3). Cranshaw's figure for the half-life of the 6.97 disintegration, 85^{217}, is 2.1×10^{-2} sec so that this member will be practically inseparable from 87^{221} which precedes it.

Evidence on 87^{221}.

The α-decay of Ac^{225} leads to 87^{221}, the second α-emitter after the radium isotope.

During the early work, when a search was being made for isotopes of lead and bismuth at the end of the series, a sample of barium chloride containing Ac^{225} was used in volatilisation experiments. Besides the bismuth activity found, there was indication of an α-activity with a period of the order of a minute which was volatilised at about 900°C and could be condensed on a cool surface.

The method of volatilisation has proved most convenient in working with this short-lived activity since the separation can be done with great speed and the sample is immediately ready for examination.

In the later work, the activity was volatilised from lanthanum oxide films on platinum trays by the following procedure. A clean gold tray was fitted into a hollow in a copper block so that there was good thermal contact and the platinum tray containing the lanthanum oxide and Ac^{225} as an adherent layer was laid upside down on top. The shape of the trays prevents the oxide from touching the gold tray. The upper tray was heated to any desired temperature with a small oxy-gas flame and the volatilised activities condensed on the cooled gold tray.

When specimens were intended for pulse-analyser measurement, a flat foil was substituted for the lower tray.

In the ordinary α-counter, the condensed activity is seen to diminish rapidly at first and then more slowly with the 46 minute period of Bi^{213} (controlling Po^{213} α-particles) which is volatilised too. No α-activity remains after the Bi^{213} has decayed, Ac^{225} being involatile.

Several samples, thus prepared, have been examined in the large ionisation chamber and pulse-analyser. It was found possible to get a specimen into the chamber and ready for counting in about ten minutes and it is found that all the α-energies are present except the lowest at 5.81 Mev (Ac^{225}). The highest energy α-peak is controlled by the 46 minute Bi^{213} and decays with that period while the two remaining peaks at 6.33 and 6.97 decay together very quickly (the period is about 5 minutes) and disappear. (Fig. 16 in the second section on branching of Bi^{213} shows this phenomenon).

From this it seems possible to volatilise the second α-emitter of the four which is believed to be 87^{221} from previous evidence.

The volatility and subsequent condensation of the activity under the conditions of the experiment eliminate elements from 86 to 92 except 87. The volatility of element 87 may be roughly inferred from the properties of the other alkali metals. The chlorides have lower boiling points as the atomic weight increases so that 87 chloride might be expected to boil at about 1150°C. Information on the behaviour of alkali metal oxides when heated in air is scanty but there is an increase in volatility with the atomic weight and the volatility of 87 from lanthanum oxide seems not improbable.

One isotope of element 87 is known already, AcK (87^{223}) a β-emitter of 21 min half-life, discovered by Mlle M. Perey (23) who found that the activity

To face page 26

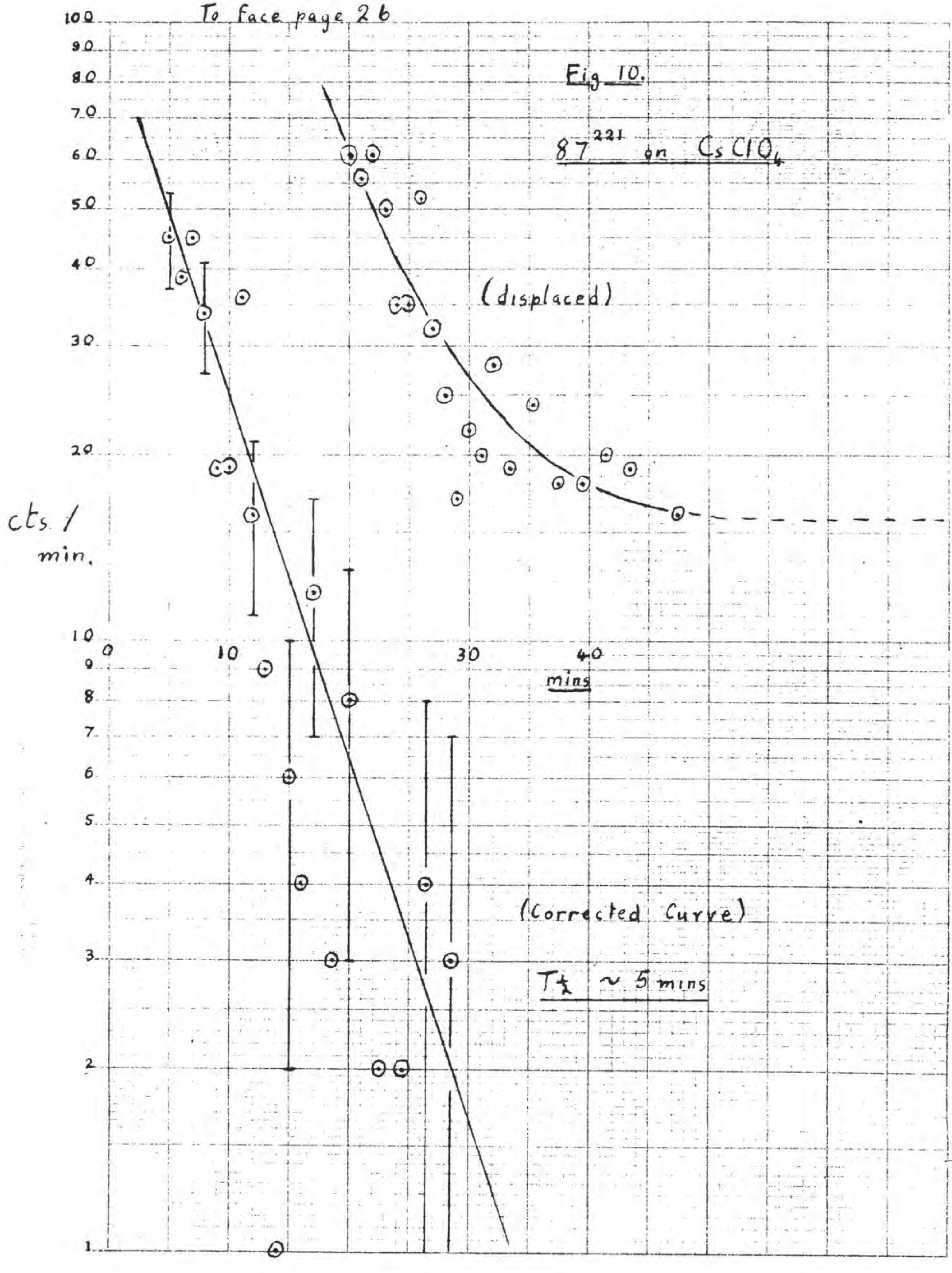


Fig 10.

87^{221} on $CsClO_4$

grew from Ac^{227} and showed chemical properties similar to caesium. One of the methods used was the precipitation of AcK by caesium perchlorate.

It has been found possible to precipitate the 5 minute activity of the new series on caesium perchlorate.

A sample of Ac^{225} in lanthanum, together with 2 mg of caesium, was taken to fumes with a few drops of perchloric acid and cooled to a clear solution. On adding a cc. of alcohol, a crystalline precipitate of caesium perchlorate was rapidly formed, was centrifuged and transferred to a gold tray. No time was taken to wash the precipitate which separated cleanly, and the transfer was facilitated by using a lusteroid centrifuge tube from which the bottom could be cut with a razor blade. The time from precipitation to the start of counting was less than five minutes.

Decay of the sample is shown in Fig. 10 with the corrected decay curve after substracting long lived activity. Arrangements have been made to test the volatility of AcK in the near future as a check on this property of the new 5 min. activity but the evidence at the moment is quite convincing, especially in the light of the total knowledge on the series.

THORIUM ISOTOPE

Thorium^{229}.

Not much attention has, as yet, been paid this member of the 4n + 1 family for several reasons. Any extraction of a thorium isotope will, of necessity, contain radiothorium which has a half-period 1.9 years, so that it is impossible, at present, to obtain a pure specimen of Th^{229}. Since the work on the series beyond the radium isotope has proved most fruitful, for in this case it is not very long before the natural thorium family disappears, it was good policy to leave the thorium isotope in the U^{233} solution so that

To Face page 27

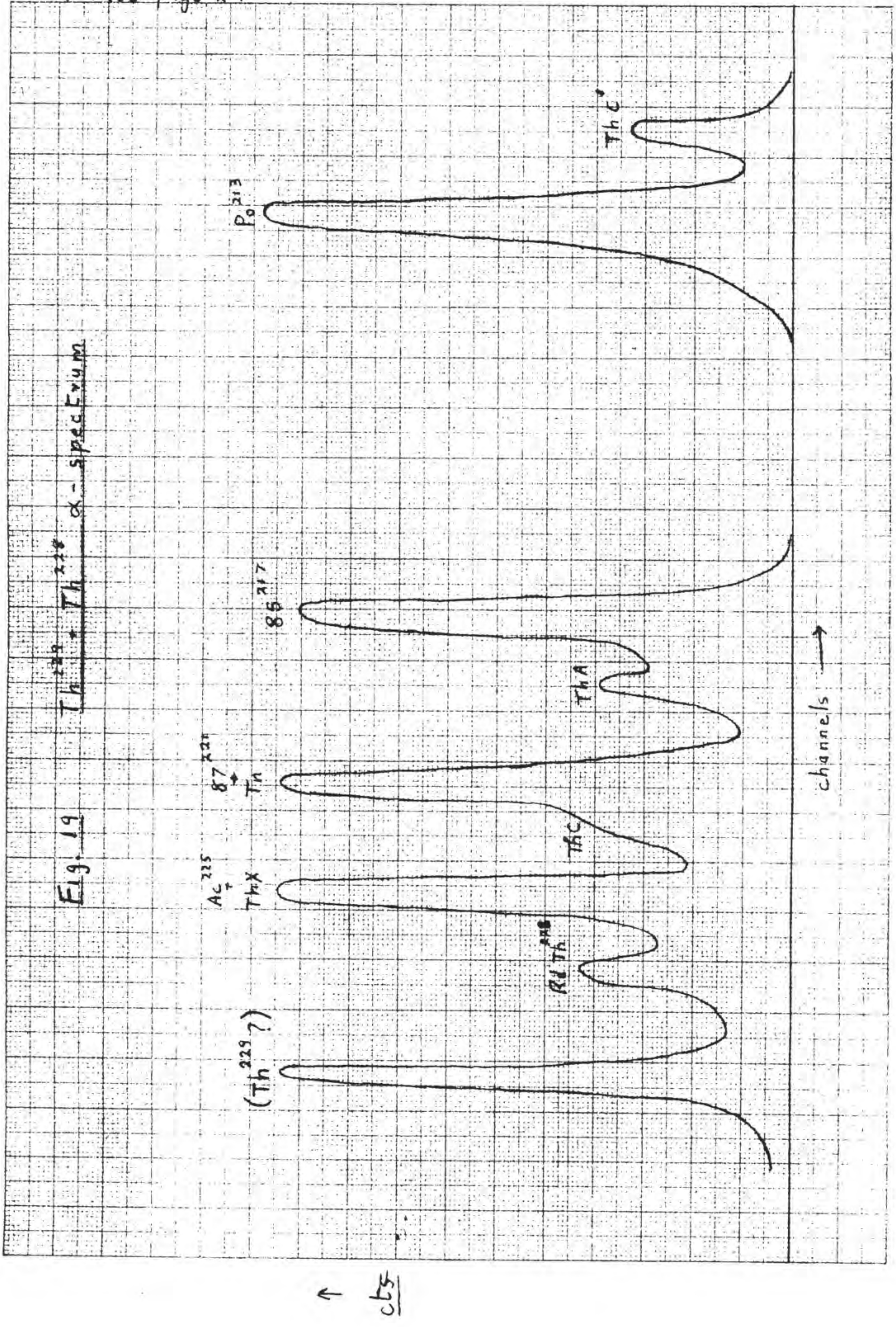


Fig. 19 $Th^{229} + Th^{228}$ α - spectrum each extraction of Ra^{225} gave a higher activity and allowed new advances. From the beginning of the work to the time of writing, the yield of α-counts at the maximum of the Ac^{225} growth curve has changed from about 200/min to 4000/min for the four members.

One extraction of the thorium isotope has been made, by a method which was designed so that the minimum of reagents need be added to the U^{233} sample, using the radiocolloid effect for separating radioactive tracer amounts of thorium from milligram quantities of uranium.

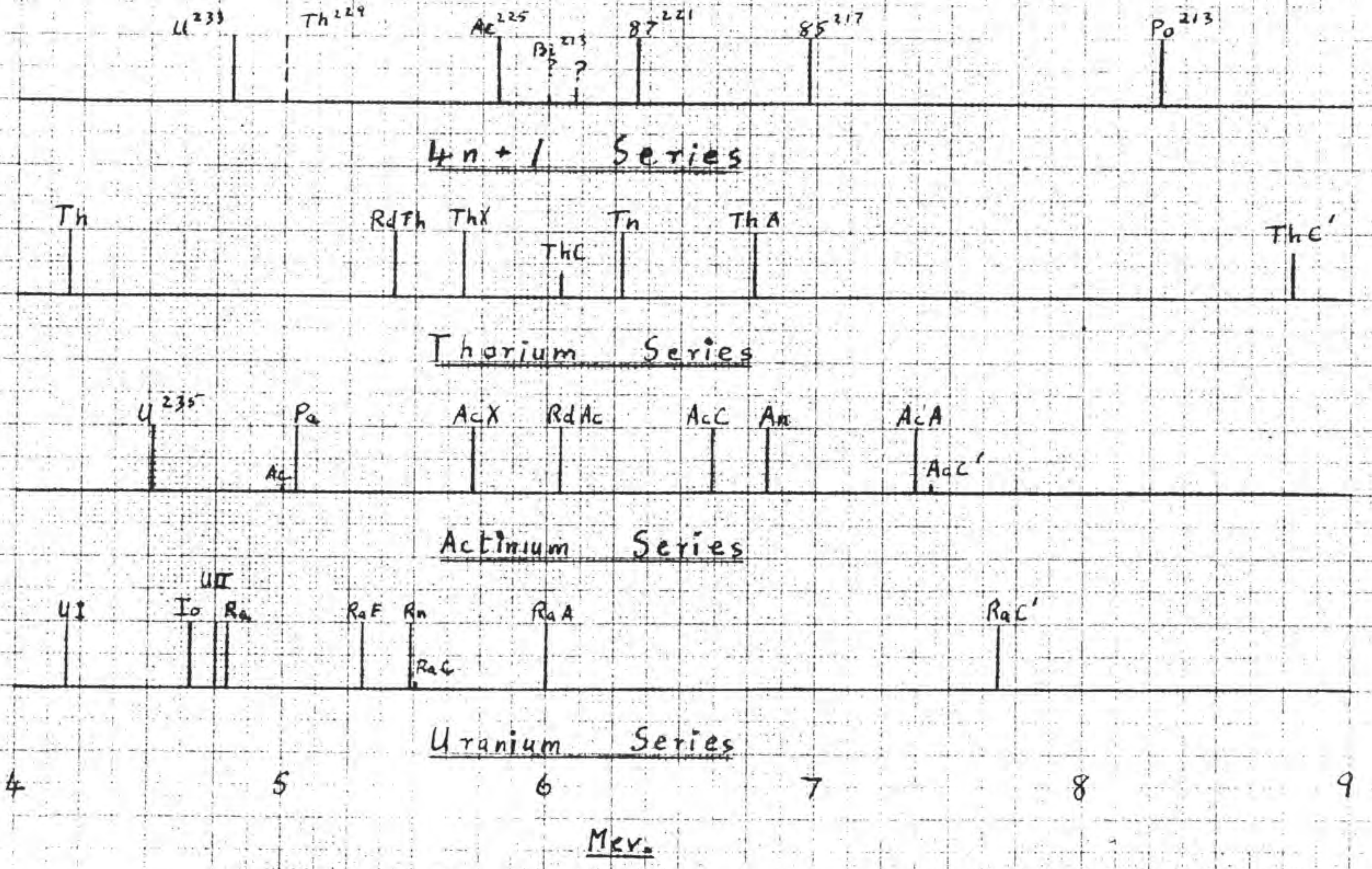
The nitrate solution of about 9 mg of U^{233} was evaporated to dryness on the water-bath twice and then redissolved in distilled water and stood hot for several hours. Under these conditions, thorium is adsorbed on the walls of the glass vessel. In this extraction, however, a small precipitate formed at this stage, possibly from impurities in the fuming nitric acid which had been added at various times to the U^{233} sample and it was suspected and later confirmed that this precipitate contained some U^{233}. The solution was separated by centrifuging and the precipitate and adsorbed layer dissolved in nitric acid. A lanthanum oxalate precipitation was then made producing a sample containing about 900 α-counts/min and on this a further radiocolloidal separation gave a purified fraction of 300 counts/min.

At the time of the experiment, the resolving power of the pulse-analyser was poor and examination of part of the final sample by Mr. Hincks showed only a broad α-peak at about 5 Mev and it was seen that more energetic α-emitters grew in the sample. Later, when the pulse-analyser was enhanced by Cranshaw's "non-linear" amplifier, I examined a portion of the thorium extract now containing almost equilibrium amounts of the disintegration products of both Th^{229} and $RdTh^{228}$. The spectrum obtained is shown in Fig. 19

To face page 28

Fig. 21

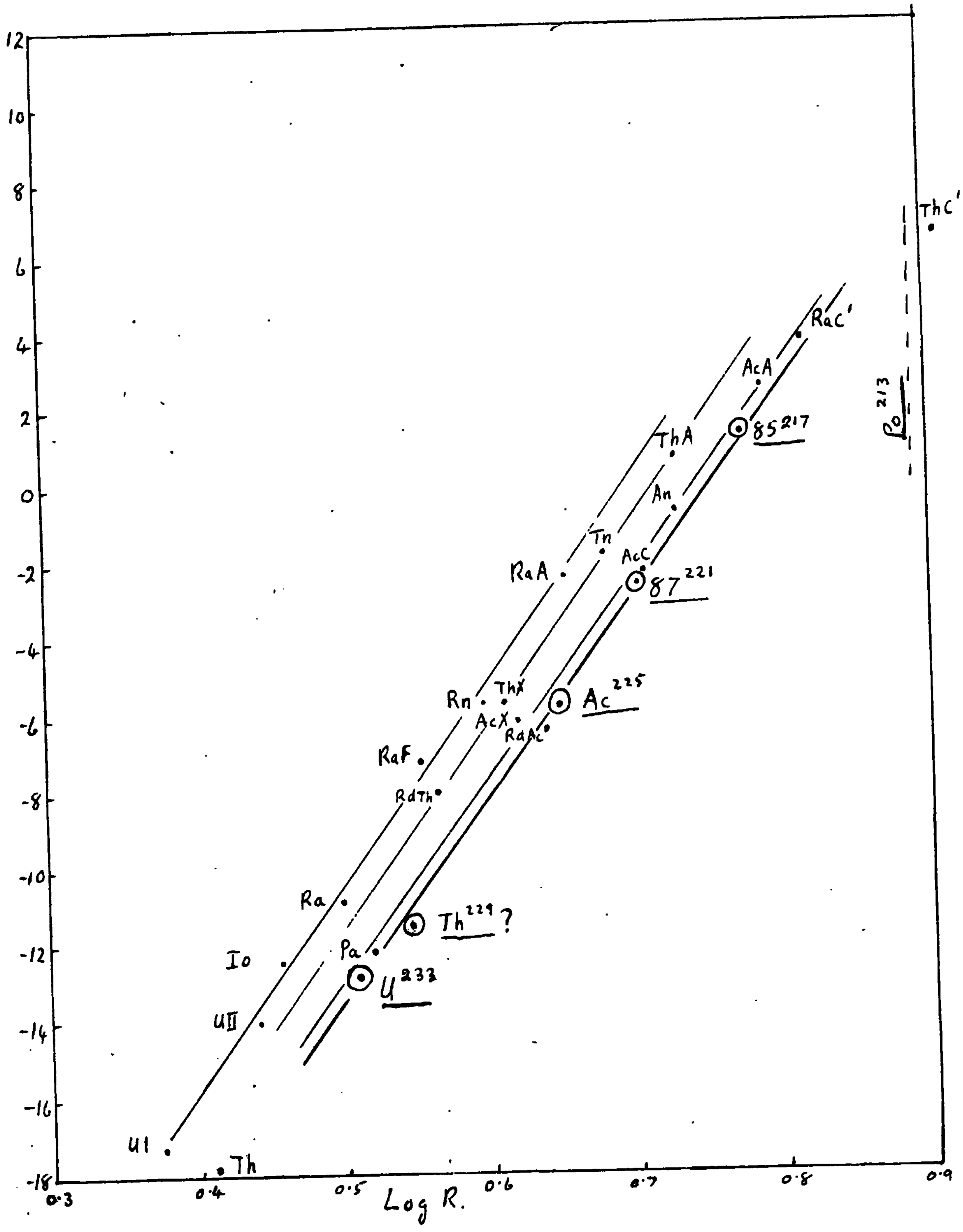
α-energy spectra of radioactive series



To face page 28

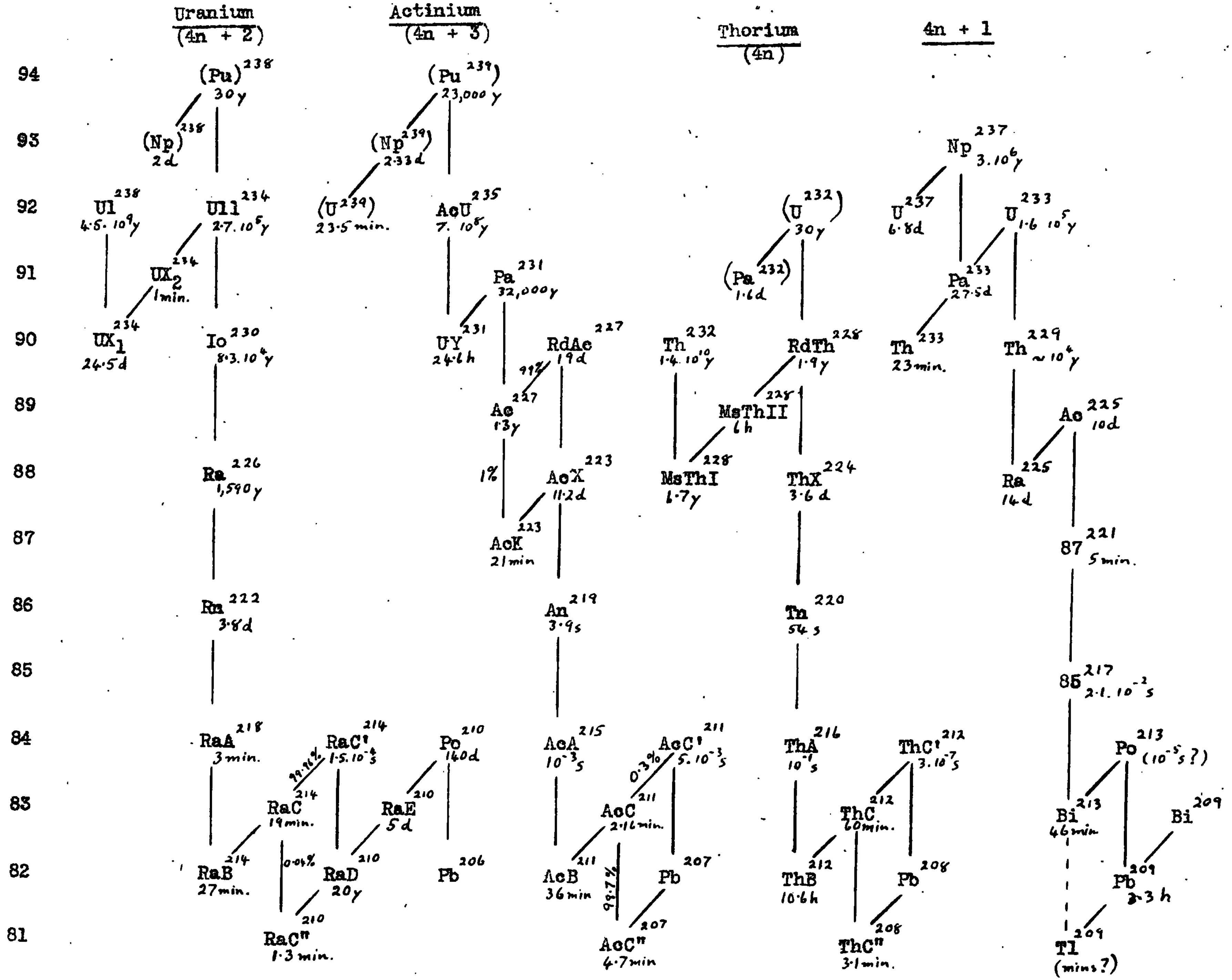
Fig. 22

Geiger-Nuttall curves



To face page 28

THE FOUR RADIOACTIVE SERIES FIG. 20



in somewhat idealised form where it will be seen that in the high energy region the two peaks of Po^{213} and ThC' are visible and indicate the relative proportions of the two families; lower down ThA and 85^{217} are identified. As far down as ThX, the distribution is similar to that of Fig. 6C since in both cases the ratio is in favour of the 4n + 1 series.

In addition to all α-peaks previously identified, there are two others lower than ThX, the smaller of which corresponds in position to the energy of radiothorium (Th^{228}) and is commensurate with the visible peaks of ThA and ThC', while the lowest at about 5 Mev corresponds in size to the rest of the 4n + 1 series peaks and almost certainly belongs to Th^{229}.

The α-activity of Th^{229} is not in doubt since it is required by the existence of Ra^{225} (Fig.3) and 5 Mev is to be expected if the half-life is about 10^4y as calculated in a later section.

THE FOUR RADIOACTIVE SERIES COMPARED.

It is quite important, in presenting the evidence for the 4n + 1 series, to provide a general comparison of the four series. For this purpose the following diagrams are given.

a) The disintegration routes of the four series are mapped in Fig. 20 Some artificially produced members have been included in the natural series and are indicated by brackets.

b) The α-energy spectra are compared in Fig. 21.

c) Geiger-Nuttall curves are plotted in Fig. 22.

SUMMARY AND DISCUSSION.

Working on milligram quantities of U^{233}, it has been found possible to investigate its disintegration products and to outline the main route of the 4n + 1 series.

The crucial evidence in defining the sequence of the disintegration and the isotopic assignments is as follows, where reference to Fig. 3 will make the arguments clearer.

A radium isotope has been extracted which is β-active and predecessor of four consecutive α-disintegrations.

From the radium grows an actinium isotope of period 10 days which displays the same four α-disintegrations and is inseparable from the first of these which has an energy appropriate to the period. The α-activity of the actinium isotope is further confirmed by the evidence on its disintegration products some of which cannot be formed if it is β-active.

One of the products of these radioactive substances is a lead isotope which is β-active with a period measured as about 3.2 hr and from which apparently no further α- or β-activity is derived. This is undoubtedly the isotope Pb^{209}, already known as 3.3 hr β-emitter which decays to inactive Bi^{209} (see appendix I).

This evidence indicates that the use of Fig. 3 is legitimate and that the radium and actinium isotopes are Ra^{225} and Ac^{225}.

The fourth product which is clearly defined is a bismuth isotope which is β-active with period 46 minutes followed by a substance of extremely short life which disintegrates with emission of a high energy α-particle, the last in the series. These facts can only be correlated with the substances Bi^{213} and Po^{213} and there is confirmation on the Po^{213} isotope provided by the work of Dr. A.N. May's section in the fact that the energy of the α-particle associated with the bismuth isotope fits very well for Po^{213} when compared with the other known isotopes of polonium.

The sequence of the disintegrations is now defined by these main points as that shown in double lines in Fig. 3 and confirmation is obtained in the second α-emitter which should be an isotope of element 87 and which is shown to behave like an alkali metal.

From these particular data it is seen that the sequence is unlike any of the known series and its identification as part of the 4n + 1 series, apart from the fact that the activities were derived from U^{233}, is made certain by the detection of a lead isotope with an activity and half-life similar to Pb^{209} already made by direct artificial methods.

The present information on the disintegration products of U^{233} is now summarized as follows.

1) <u>Th^{229}</u>

Owing to the fact that our sample of U^{233} contains also some of the isotope U^{232}, it has not been possible to extract Th^{229} free from the Th^{228} (RdTh). A thorium extraction has been made and it was shown that the four α-emitters from Ac^{225} on were not initially present but grew in slowly (together with the disintegration products of RdTh). In the pulse-analyser, an α-peak has been distinguished at about 5 Mev which is probably due to Th^{229}.

The half-life has been roughly calculated from the yields of the subsequent disintegration products and their periods, giving a figure of about 10^4 years.

2) <u>Ra^{225}</u>

This substance is shown to be β-active with a period calculated from the growth of its daughter as 14 ± 2 days. The β-radiation is of very low energy, probably less than 0.05 Mev.

3) Ac^{225}

The actinium isotope is α-active and decays with a half-life of 10 days. The latest energy measurement of its α-particle gives 5.81 Mev.

4) 87^{221}

The half-life of 87^{221} is measured as 5 ± 1 mins and the energy of its α-particle 6.33 Mev. From an irregularity in the shape of the peak in the pulse-analyser, first noticed by Cranshaw, it seems probable that the α-emission displays fine structure since a small peak is visible at 6.1 Mev.

5) 85^{217}

Cranshaw obtains for the half-life of this substance, 2.1×10^{-2} sec. and 6.97 Mev for the energy of the α-particle.

6) Bi^{213}

This substance is mainly β-active with period of 46 mins. A rough figure for the energy of the β-rays derived from absorption measurements is 1.3 Mev.

There seems to be branching in the α-direction of less than 3% with the energy of the α-particle at about 6 Mev.

7) Po^{213}

The polonium isotope is a very short-lived α-active body (? 10^{-5}s) and is inseparable from the Bi isotope. Cranshaw's measurement of the energy of the α-particle is 8.28 Mev.

8) Pb^{209}

The last radioactive member of the series is Pb^{209} which is already known as a β-emitter of 3.3 hr half-period.

9) Tl^{209}

This is believed to be present in the series from a branching of Bi^{213} less than 3% of the main route. Activities available are too small to work on this member which is probably β-active with a half-life of a few minutes.

The 4n + 1 series in nature.

No known member of the 4n + 1 series has a period sufficiently long to allow the independent existence of the family in nature but there is undoubtedly some small production through the reactions U^{238} (n,2n) U^{237} and Th^{232} (n,γ) Th^{233} brought about by spontaneous fission neutrons, of which the second reaction is the more important. From measurements of the spontaneous neutron emission of U and Th, Pose (30) estimates the half-life for spontaneous fission of these two elements as $3.1 \cdot 10^{15}$ years and $1.7 \cdot 10^{17}$ years, making the assumption that one neutron is produced per fission. On the assumption that three neutrons are emitted per fission, the U half-life would be about 10^{16} years, the generally accepted value at present, and the Th half-life, $5 \cdot 10^{17}$ years. Since the neutron emission of thorium is probably about 50 times less than that of uranium, the production of the 4n + 1 series by the second reaction is hindered. The best conditions would obtain in a mineral of mixed thorium and uranium. It has been estimated (27) that uranium deposits containing of the order of a few percent of thorium might contain one part in 10^{15} of U^{233}.

Already the presence of elements 93 and 94 in various uranium minerals has been investigated by Seaborg and Perlman (22) and Garner, and Bonner and Seaborg (27) who obtained indication of a concentration of about one part in 10^{14} of 94^{239} in pitchblende and carnolite. The α-activity extracted could be due to other isotopes of 93 or 94 including those of the 4n + 1 series but 94^{239} seems the most probable, derived from the absorption of neutrons in U^{238}.

Wahl (28) reported the detection of a mass-line 237 in the mass-spectrographic investigation of some minerals of the tantalum-niobium group containing uranium, thorium and rare earth metals. However, he also mentions the mass-line 205 (supposing it to be the end-product of the hypothetical new series) which is the main isotope of thallium and it is significant that the difference between 237 and 205 is 32, the mass of two oxygen atoms. On the present knowledge, it seems improbable that concentrations greater than one in 10^{15} of the 4n + 1 series exist in nature.

The new information on the series enables us to make a more intelligent search and it is immediately apparent that no consideration need be given the short-lived members after Ac^{225} nor the isotopes of uranium or thorium.

One of the most promising approaches to the problem seems to be at the radium and actinium isotopes. Of the known radium isotopes, Ra^{226}, AcX^{223}, $MsThI^{228}$, ThX^{224}, and $NpRa^{225}$, the last is unique in the production of an α-emitting actinium isotope. It should be possible, by the ordinary radium extraction technique and very careful purification by the fuming nitric acid method, to obtain from a mineral a barium nitrate extract containing only radium isotopes. After about 10 days when the Ac^{225} reaches about 90% of the maximum on the growth curve from Ra^{225}, a preliminary separation of ceric iodate is done, if necessary, to eliminate radiothorium grown from MsThI and then a precipitation and purification of cerous or lanthanum fluoride to extract any Ac^{225} present. If MsThII is present in any quantity a few days would be sufficient to reduce it to negligible proportions and the ceric iodate precipitation could be conveniently transferred to this point for removal of the radiothorium. The final fluoride extract should contain only Ac^{225}, and its disintegration products which increase the α-activity by a factor of four.

The sensitivity in terms of activity should be very similar to that of Seaborg and Perlman and a yield equivalent to their "order of 10^{-5} microgram of 30,000y 94^{239}" would mean an order of 10^{-4} microgram for U^{233} or 10^{-3} microgram for Np^{237}.

NOMENCLATURE.

The nomenclature of the 4n + 1 series is, at the time of writing, under discussion with Seaborg's team who also have been working on the series, although the results up to December 1944 differ somewhat from those reported here except in the case of Pb^{209} where agreement is reached. (CC 2636, Ghiorso, Hagemann, Katzin and Studier).

In CC 2636, use is made, "for the sake of simplicity", of the name "neptunium series" and the name has since been widely used in this laboratory. It is hoped that this will be accepted as the name of the 4n + 1 series.

A detailed nomemclature for the family has been discussed at this laboratory and will be the subject of a later note if and when it is accepted by the other contemporary workers on the series.

To face page 35

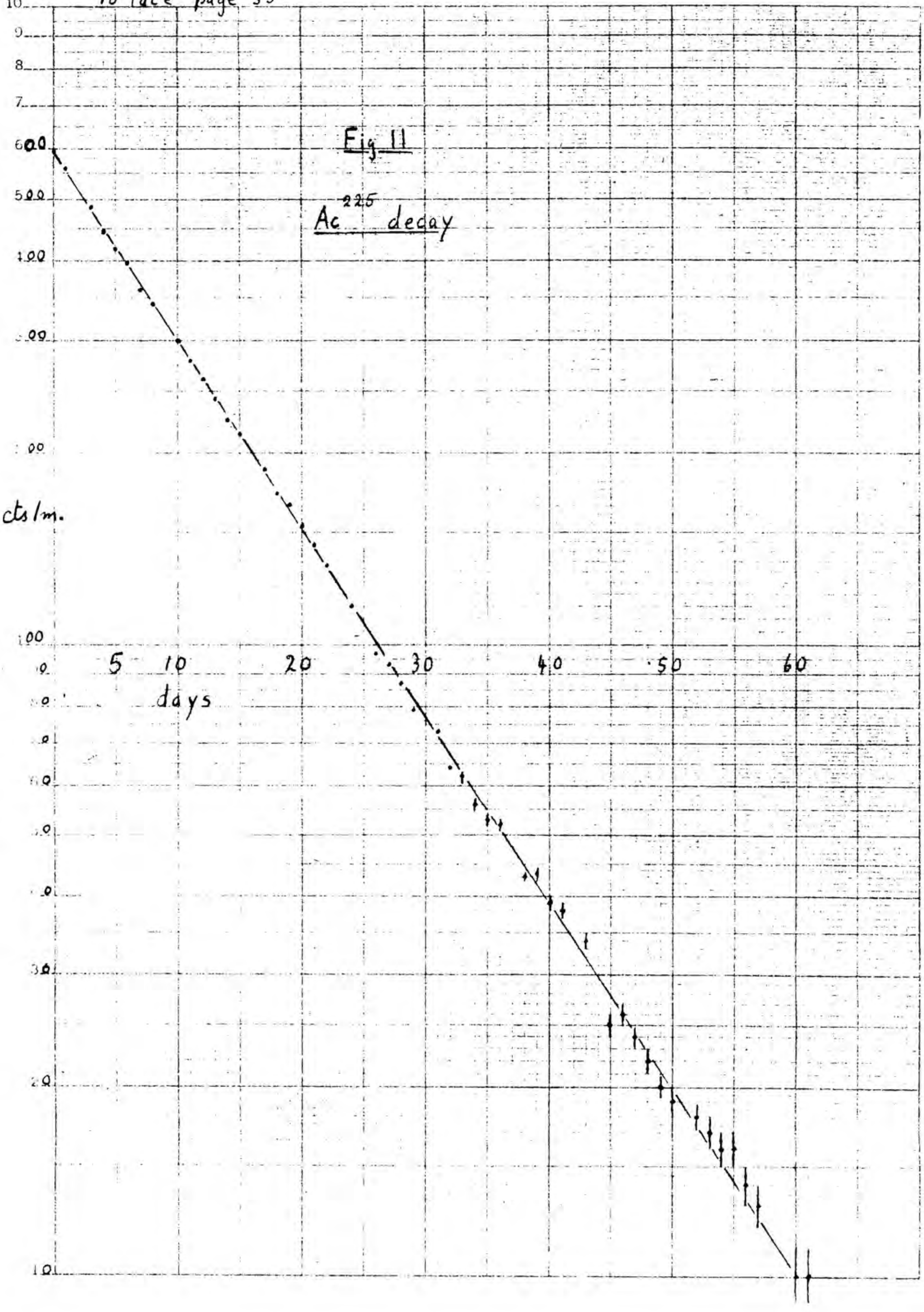


Fig 11

Ac^{225} decay

APPENDIX I

Ac^{225} half-life Measurement.

For the decay period of Ac^{225}, a lanthanum fluoride extract from Ra^{225} was purified by reprecipitation, evaporated on a platinum tray with nitric acid, and ignited. The sample was counted regularly in the ordinary α-counter and the decay curve given in Fig. 11. has been corrected for a long-lived contamination of 7 cts/min. On this measurement the half-life obtained is 10.0 ± 0.25 days.

Ra^{225} half-life.

The direct measurement of the period of Ra^{225} is difficult since, as will be shown in the next section, its own β-radiation is very soft and that of its disintegration products hard, and the period of Ac^{225} is related such that the approach to the transient equilibrium is very slow, about 200 days for 99% of the equilibrium amount (calculated from the result obtained following).

Using the measured half-life of Ac^{225} given above it is possible to estimate the half-life of Ra^{225} from the curve given in Fig. 7. Theoretical curves for various half-life values of Ra^{225} have been compared and the best fit obtained for 14 days with an error of, at most, ± 2 days.

Ra^{225} β-rays.

β-ray absorption curves have been taken on samples of Ra^{225} containing Ac^{225} and on Ac^{225} alone in the ordinary Geiger-Müller counter having a 6 mg/cm^2 mica window and it is found that the shape of the absorption curve seems to be the same whether Ra^{225} is in the sample or not.

As previously reported (p.14), a sample of Ra^{225} on barium nitrate, contaminated by only a small amount of ThX, has been purified from its disintegration products for the purpose of showing the growth of α-activity from Ra^{225}, and in this sample a search for β-activity was made. The sample was counted in the alpha counter, the thin-window Geiger-Müller counter and in the windowless counter.

The following calculations are made first to show roughly the activity to be expected in the purified Ra^{225}.

Immediately before this purification, the α-count on the sample was 890/min. and this represented 9 days growth of Ac^{225} after the previous treatment of the radium isotope.

Using the values of 14 days and 10 days as the half-lives of Ra^{225} and Ac^{225} respectively and the equation

$$\frac{N_2^t \lambda_2}{N_1^t \lambda_1} = \frac{\lambda_2}{\lambda_2 - \lambda_1} \left(1 - \frac{e^{-\lambda_2 t}}{e^{-\lambda_1 t}}\right)$$

representing the relationship between the numbers of atoms of Ra^{225} (N_1) and Ac^{225} (N_2) in a sample, originally pure Ra^{225}, after time t, on the 9th day,

$$^{*}\ \frac{\text{cts } Ac^{225}}{\text{"cts" } Ra^{225}} = \frac{N_2^t \lambda_2}{N_1^t \lambda_1} = 0.59$$

Thus the expected β-activity of the Ra^{225} in the geometric efficiency of the α-counter would be

$$\frac{890}{4 \times 0.59} \simeq \underline{380} \text{ cts/min.}$$

taking no account of self-absorption.

A check on this value is obtained from the growth curve of α-particles in the sample after the purification (Fig. 7). For this, t = 18 days,*

---

* (In both of these calculations, the corrections for counting efficiencies are left out since they cancel).

cts Ac^{225} $(N_2^t \lambda_2) = \frac{500}{4}$ and "cts" Ra^{225} at the purification =

$$N_1^0 \lambda_1 = \frac{(N_2^t \lambda_2) \quad (\lambda_2 - \lambda_1)}{\lambda_2 (e^{-\lambda_1 t} - e^{-\lambda_2 t})}$$

$$= \underline{385} \text{ cts/min.}$$

Thus about 400 β-cts/min. could be expected in the purified barium nitrate precipitate.

The α-count immediately after purification was 15 cts/min. and this has been shown to be probably due to ThX (p.15). In the thin window counter, there were 37 cts/min., a value too low for the Ra^{225} and higher than would be expected for contamination by other β-activity. The absorption half-thickness of this activity was about 9 mg/cm^2 Al and it is possible that it is due to a small contamination by Pa^{233} of which there is some quantity in the laboratory at present.

About six hours after the purification, the sample was counted in the alpha- and thin-window beta-counters and in the windowless counter. In the latter, all ionising particles are counted. Table II summarises the data from measurements taken at 6 hrs. and 18 hrs. after the purification. From the count obtained in the windowless counter are subtracted those of the α-counter and the thin-window counter after allowing for geometric efficiencies, which allowance is not very accurate in the case of the thin-window counter.

Table II

(see next page)

To face page 38

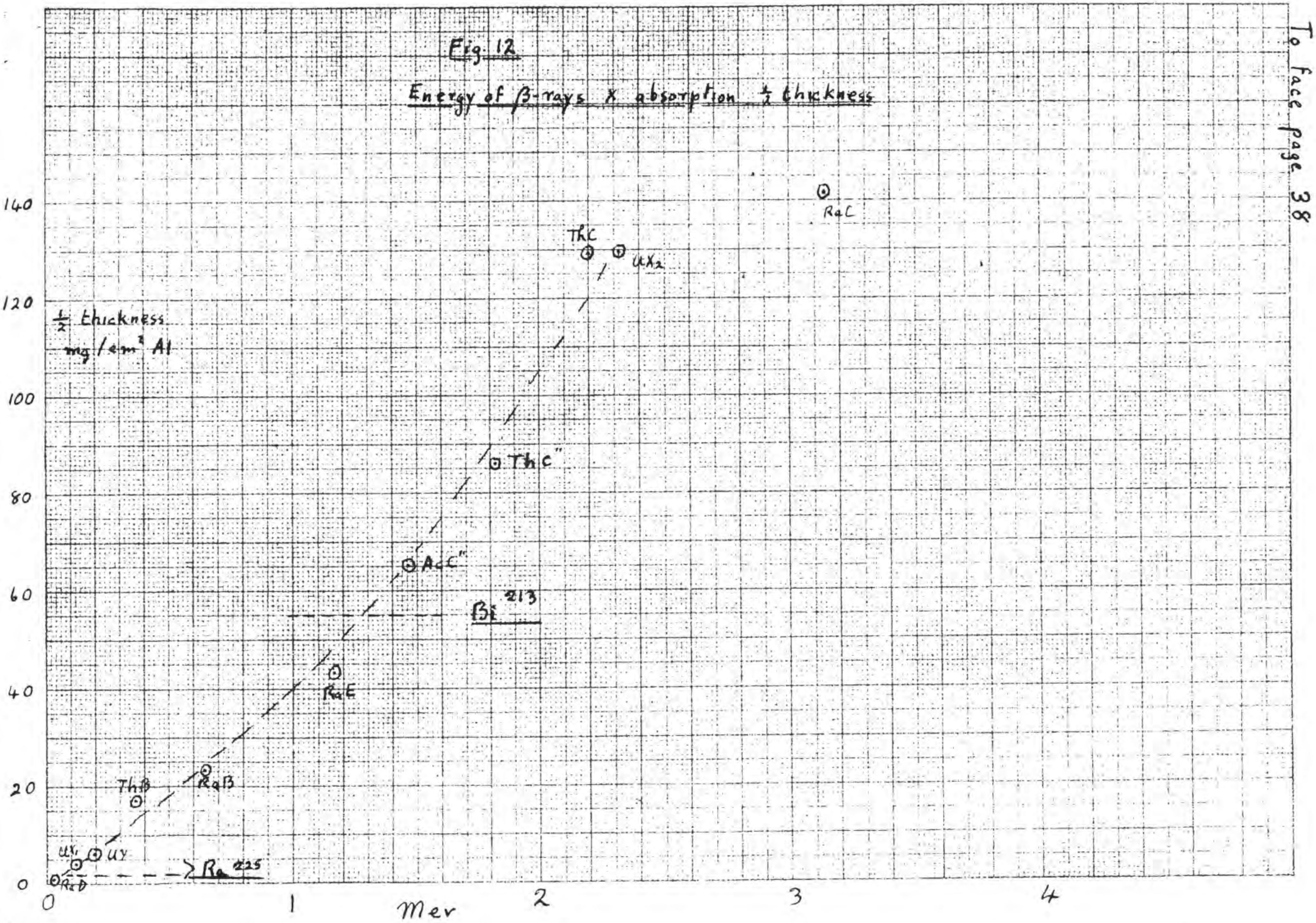


Fig. 12

Energy of β-rays X absorption ½ thickness

TABLE II

	at 6 hrs.		at 18 hrs.	
Cts/min. in windowless counter.		278		316
Correction for α's	20		43.5	
Correction for thin-window β's	61		78.5	
Background on windowless counter	47		64	
	128	278	186	316
∴ cts obtained only in windowless counter		150		130
Corrected to α-counter efficiency		260		225

Thus about 2/3 rds of the roughly calculated counts for Ra^{225} are found in the windowless counter which are not counted through a window of 2 mg/cm^2 mica.

Some idea of an upper limit for the β energy of Ra^{225} may be obtained from this fact.

In Fig. 12. is plotted the energy of β-radiation of a number of radioactive substances[*] against the absorption half-thickness in aluminium[**]. A half-thickness of 2 mg/cm^2 Al corresponds to 0.05 Mev so that it seems safe to say that the energy of Ra^{225} β -emission is less than this.

---

[*] Table of Isotopes, Seaborg, Rev. of Mod. Phys. 16, 1 (1944).
[**] Handbook of Chem. and Phys., 1943, Chemical Rubber Publishing Co.

To face page 39

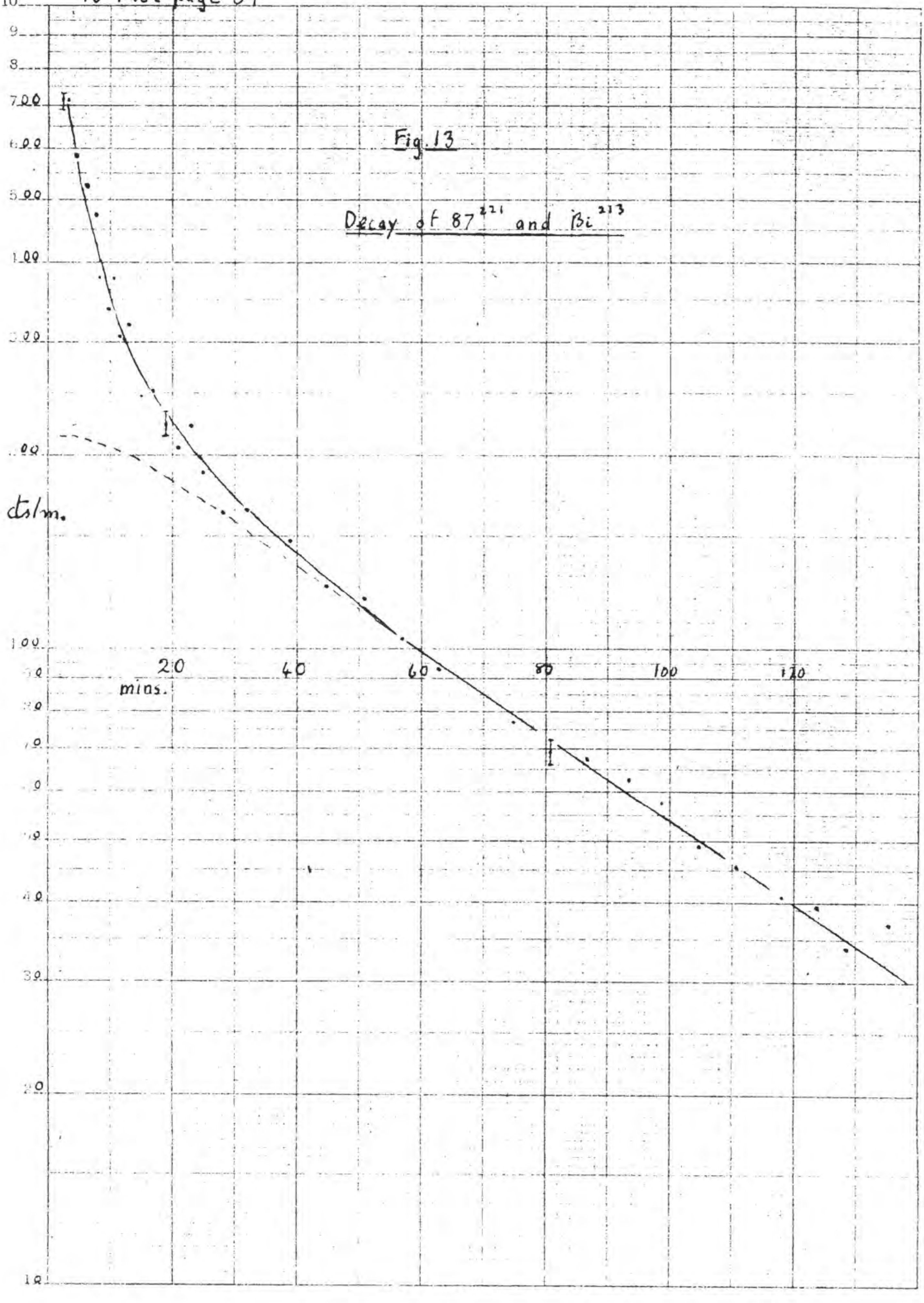


Fig. 13

Decay of 87^{221} and Bi^{213}

α-Branching of Ra^{225}.

More careful consideration may be given the evidence on α-branching of Ra^{225} which is contained in the work on the purified sample of Ra^{225} reported above and in the first section of this report.

The sample contained by calculation, about 400 cts/min. (50% eff.) of Ra^{225} and, when first purified, showed only 13 α-cts/min. (50% eff.) which can be completely accounted for as residual ThX. If they are due to alpha-branching of Ra^{225}, at the most it is a 3% branching (allowing no alpha particles for further short-lived products) and probably would be about 1% branching (allowing two short-lived products). (cf. Feather speculations on lifetimes (17)).

The evidence, however, indicates that branching is much less than 1%. It will be remembered that in the pulse-analyser examination of a fresh extract of radium isotope from the U^{233}, there was no definite sign of branching though the method is less sensitive than the previous experiment.

Period of 87^{221}.

For the half-life measurement of this short-lived body, samples were obtained by volatilisation as previously explained (p.24) and they contained varying amounts of the 46 minute Bi^{213}. A decay curve of such a sample in the ordinary α-counter is given in Fig. 13 and the line representing Bi^{213} activity (Po^{213} α-particles) is projected for the purpose of correcting for the decay of 87^{221}.

There is little point with such low activities in attempting a careful analysis of the decay curve and the correction was calculated

To face page 40

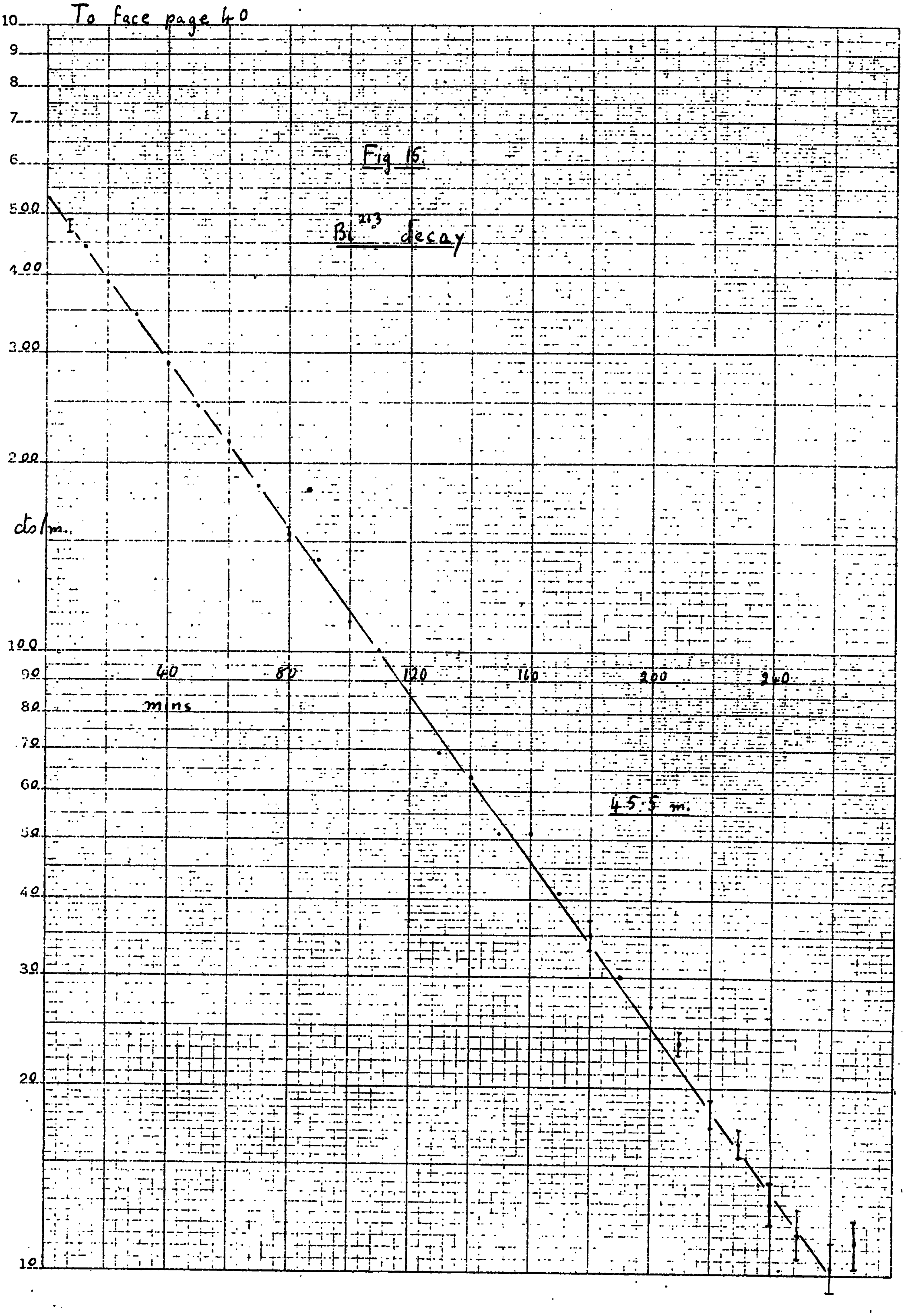


Fig 16.

Bi^{213} decay

To face page 40

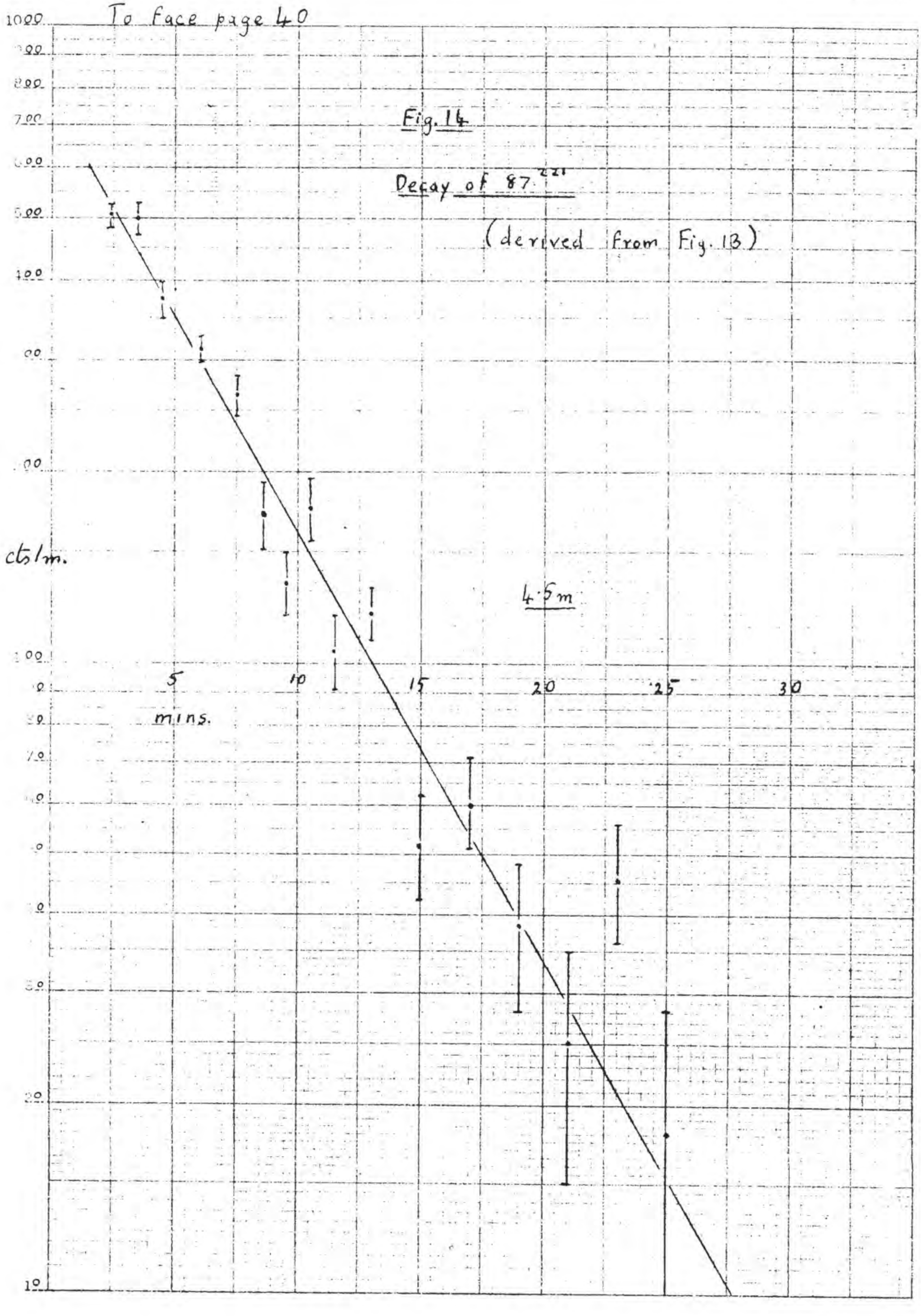


Fig. 14

Decay of 87^{221}

(derived from Fig. 13)

by the following steps. The 46 minute decay line was drawn in and projected to the time of the first count. Subtraction gave a rough figure for the α-particles of 87^{221} and 85^{217} and reduction in the ratio of 5 (the approximate half-life of 87^{221}) to 46 x 2 provided an approximate figure for the α-activity of Bi^{213} (Po^{213}) which was formed from the decay of 87^{221}. Repetition of the original subtraction using the reduced value of original Bi^{213} gave a final figure for the 87^{221}α-particles which was used in calculating the small growth curve superimposed on the decay of Bi^{213} already present in the sample. Since this secondary correction is small, the error due to the approximation is negligible in this case.

The corrected decay curve for 87^{221} is drawn in Fig. 14. On two experiments the half-life is estimated at 5 ± 1 min.

Bi^{213} half-life.

The most accurate measurements of the period of Bi^{213} have been made on sulphide precipitates having about 450 alpha cts./min. initially so that the decay could be followed for about five half-lives. Two experiments gave 45.0 - 46.0 mins. and 45.8 - 47.0 or 46 ± 1 min.. A decay curve is given in Fig. 15.

β-particles of Bi^{213}.

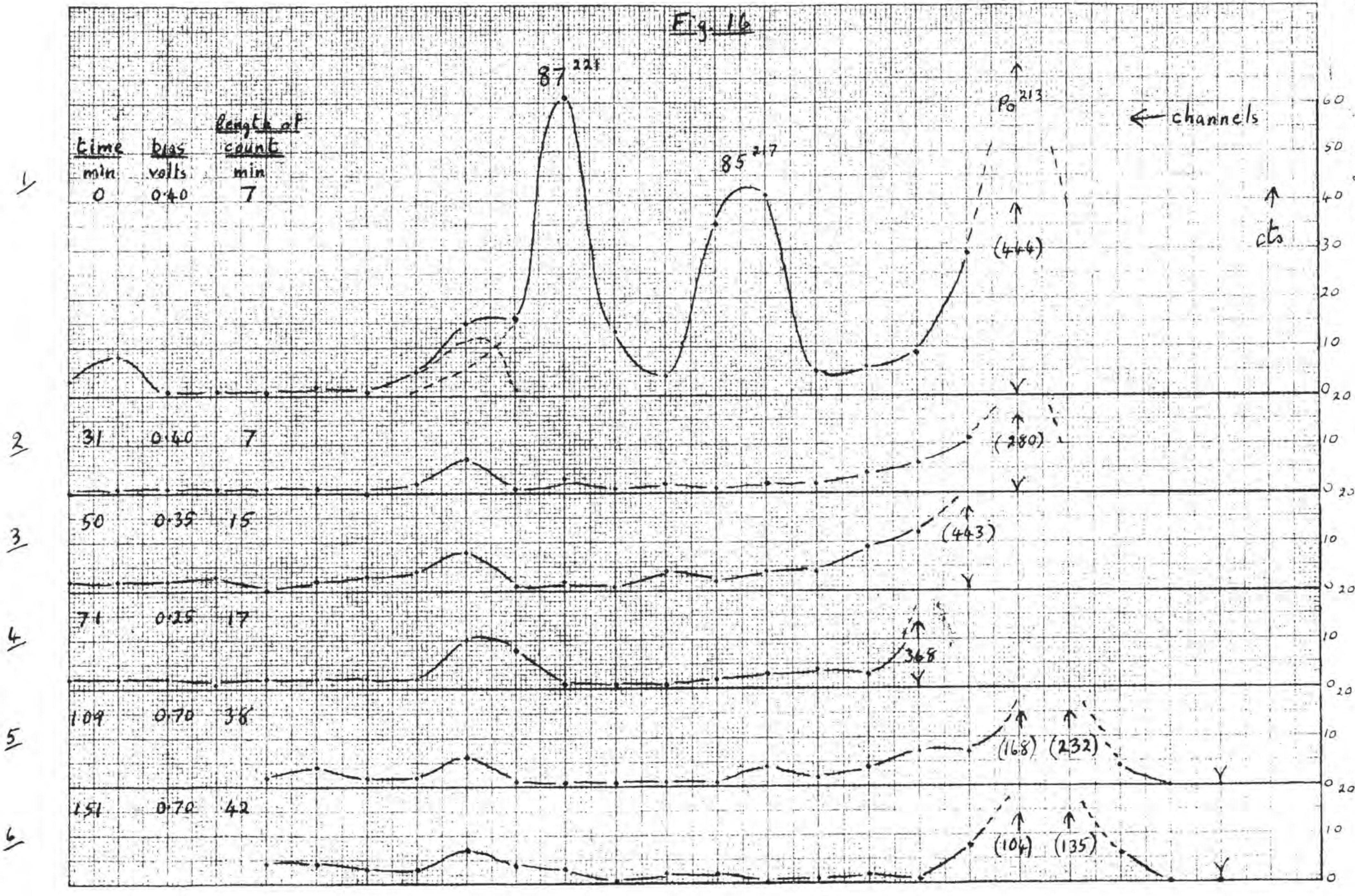
See section on Pb^{209} later.

Branching of Bi^{213} to Tl^{209}.

The corresponding members of the other three radioactive series, RaC, ThC and AcC all exhibit branching in some degree, and also RaE (Bi^{210}) according to a recent report by Broda and Feather (31). An experiment is described here which seems to find its most reasonable

- To face page 41

Fig. 16



interpretation in a branching of Bi^{213}.

A volatilisation experiment was performed on a sample of Ac^{225} as described under 87^{221} and the specimen introduced quickly into the large chamber of the pulse-analyser. Counting was started about 12 minutes after the ignition and the three main α-energies were found as usual, 87^{221}, 85^{217} and Po^{213} (controlled by Bi^{213} which had volatilised), as shown in Fig. 16, No. 1 where counts are plotted as ordinates and the pulse-analyser channels in order as abscissae.

It was ascertained that there was no contamination due to the thorium family by looking for the high energy α-particle of ThC'. After thirty minutes, six half-lives of 87^{221}, the two short-lived α-peaks had dissappeared (Fig. 16, No. 2), and a series of measurements was then made as shown in Fig. 16, Nos. 2, 3, 4, 5, and 6. It will be seen that in all the graphs, in addition to the large number of counts representing the α-particle of Po^{213}, there is a small peak at an energy somewhat lower than that of 87^{221}. Since the peak was small, there was a danger that it was merely an abnormal fluctuation in the stray counts on, or a maladjustment of, a certain channel and to obviate this, the bias was varied so as to move the peak into different regions of the 20 channels. The graphs have been displaced, depending on this bias voltage used, in accordance with the relation that 0.13 volts change in bias moves an energy band through two channels at the particular amplification used in this experiment. The peak is then seen to be quite real and seems to remain so long as the high energy peak is present.

Two hours after the last measurement in Fig. 16, another run was

To face page 42.

Fig. 17

taken when the high peak had dwindled to a count of about 40 obtained in 180 minutes and the lower peak had also disappeared. In Fig. 17 a comparison is made between (a) the sum of the measurements in Fig. 16, 2, 3, 4, 5 and 6 where the total time comes to 119 minutes, between 45 minutes after zero time and 3.5 hours, (b) the 180 minute measurement just mentioned started at 5.5 hrs. after zero time and (c) a background run on the empty chamber in the plotting of which the scale of the ordinates has been reduced to compensate for the longer count so that a visual collation may be made.

That the α-peak is real there is no doubt; its explanation requires more careful consideration. It is mentioned later in the section on α-particle energies that there is a small α-peak slightly lower in energy than the main peak of 87^{221} seen in the pulse-analyser, the small peak containing about 20% of the counts in both. The position of the small α-peak under investigation appears to be near to the smaller of the two in 87^{221} but they can hardly be the same since the former constitutes less than 3% of the main activity if it is a branching of Bi^{213}.

As considered under "α-particle energies", it is not probable that there is appreciable branching at Ac^{225}, 87^{221} or 85^{217} but even if there were branching, the possible products, isotopes of thorium, radium, or emanation, would not have appeared in this volatilisation experiment except as being produced from the 87^{221} which was condensed. Bearing in mind that this new small peak remained as long as the 46 minute high energy α-particle so that its production from 5 minute 87^{221} would lead to 10 times less activity, and also the fact that the branching, if any, at the first three α-emitters is small, there is a difference of an order of magnitude between the effect expected by branching previous to Bi^{213} and the observed effect. This is entirely dependent on the arguments given under "α-particle energies" for if 87^{221} were branched as much as 20% the effect could be explained assuming Ra^{221} or 86^{217} (Fig.3) have a period of about 50 minutes. The situation will be clarified when a high activity sample is available on which a search may be made for Ra^{221} and from which a large sample of pure Bi^{213} may be separated and examined.

Tl^{209}.

We may expect thallium^{209} to have a high energy β-emission and a half-life of only a few minutes so that with the present activities it would be almost impossible to do any work on it.

β-particles of Bi^{213} and Pb^{209}.

The activities available at present are too small to make accurate measurements of the energy of β-radiation from the series but a rough idea may be obtained from absorption coefficients.

Freshly extracted samples of the radium isotope give absorption curves showing the penetrating radiation from ThC and as the sample ages and the ThX decays, the slope changes and approaches the steady value of the β-rays from the new family. The measurements were made on the usual β-counter which has a mica window 6.5 mg/cm^2 so that the soft radiation from Ra^{225} would not be visible, a fact which has been substantiated by the similarity between absorption curves in the presence and absence of Ra^{225}.

Fig. 18 shows one of the curves obtained on a specimen of Ra^{225}

To face page 44

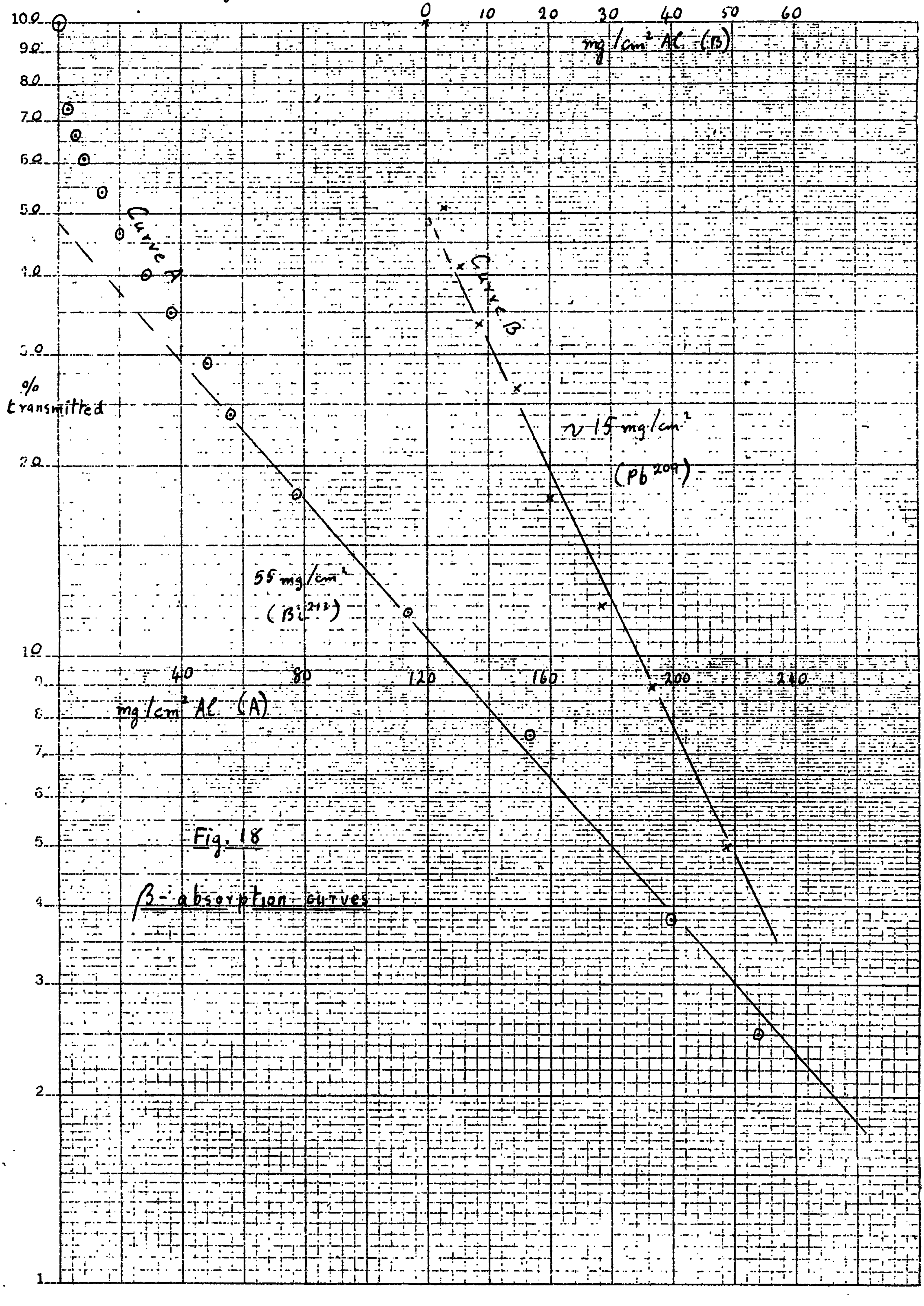


Fig. 18

β-absorption curves

18 days old and analysis of the graph demonstrates the presence of two definite components. Curve A gives the experimental points showing the two radiations combined and curve B the softer component plotted separately by the subtraction of the projection of the hard component. Qualitatively, it has been shown that the more energetic part belongs to Bi^{213} and the other, of half-thickness about 15 mg Al/cm^2, to Pb^{209}, the value being approximately correct for the reported energy of Pb^{209} β-emission, 0.7 Mev(13).

The Bi^{213} radiation has an absorption half-thickness of about 65 mg Al/cm^2 which, in comparison with other radioactive substances (Fig. 12), indicates an energy of roughly 1.3 Mev.

Period of Th^{229}.

The half-life of Th^{229} is very large so that its estimation requires a comparison between the activity grown in a certain time with the known amount of U^{233} parent and this would be complicated by the presence of radiothorium^{228}, by the growth of subsequent members in both families, and by the probable contamination by Ac^{225} in some of the chemical separations. A direct measurement of this kind has not yet been attempted but a rough calculation may be made from the extraction of Ra^{225}. Unfortunately, the activity of Ra^{225} cannot be measured easily owing to the weakness of its β-radiation and the presence of ThX, and it is necessary to go one step further to Ac^{225}. The following calculation is, therefore, somewhat devious and cannot be expected to give more than an order of magnitude. The sequence to be considered is:

$$\underset{\alpha}{U^{233}} \longrightarrow \underset{\alpha}{Th^{229}} \longrightarrow \underset{14\ d\ \beta}{Ra^{225}} \longrightarrow \underset{10\ d\ \alpha}{Ac^{225}}$$

1) To calculate counts of Ra^{225} from measured counts of Ac^{225}.

(Refer to Sections on Ra^{225} and Ac^{225} for a full explanation)

The growth of Ac^{225} from Ra^{225} is described by the following equation,

$$N_3^t = N_2^0 \frac{\lambda^3}{\lambda^3 - \lambda^2} \left(e^{-\lambda^2 t} - e^{-\lambda^3 t} \right)$$

where N_3 and λ_3 are the number of atoms and disintegration constant respectively of Ac^{225} and N_2 and λ_2 those of Ra^{225}.

Working in days throughout and leaving out the unnecessary correction for counting efficiency,

$$\lambda_2 = \frac{0.693}{14} = 0.0495 \qquad \lambda_3 = \frac{0.693}{10} = 0.0693$$

(half-lives of Ra^{225} and Ac^{225} - 14 days and 10 days)

Measured counts of Ac^{225} and short-lived daughters on the 11th day (= t) after extraction of Ra^{225} = 2300/min. (after correction for ThX)

$$\therefore \lambda_3 N_3^t = \frac{2300}{4} \times 60 \times 24 \text{ counts/day}$$

$$\lambda_2 N_2^0 = \text{counts of Ra}^{225} \text{ at } 0t = \frac{N_3^t (\lambda_3 - \lambda_2)}{\left(e^{-\lambda_2 t} - e^{-\lambda_3 t} \right)}$$

$$= \frac{2300 \times 60 \times 24}{4 \times 0.0693} \times \frac{0.0198}{0.113} = 1454 \times 60 \times 24 \text{ counts/day}$$

$$= 1454 \text{ counts/min.}$$

Efficiency of Ra^{225} extraction is 80% $\therefore$ Ra^{225} counts = 1813/min.

A correction of 10% is necessary here for self-absorption of the α-particles measured in the 2 mg of Ba(NO$_3$)$_2$.

$\therefore$ Ra^{225} = 1994 counts/min. on the 94th day after U^{233} purification.

2) To calculate the period of Th^{229}.

U^{233} - 119.10^6 counts/min. in the sample = Qo, the equilibrium count of Ra^{225}.

Th^{229} - λ_1 disintegration constant.

Ra^{225} - λ_2 d. constant, Qt = 1994 counts/min.

t = 94 days. $\lambda_2 = \frac{0.693}{14} = 0.0495, \lambda_2 t = 4.65$

$$\frac{Qt}{Qo} = 1 - \frac{(\lambda_1 e^{-\lambda_2 t} - \lambda_2 e^{-\lambda_1 t})}{\lambda_1 - \lambda_2},$$

(λ_1 is small compared with λ_2),

$$= 1 - e^{-\lambda_1 t} + \frac{\lambda_1}{\lambda_2} e^{-\lambda_2 t}.$$

Expanding the first exponential when the squared term is negligible

$$\frac{Qt}{Qo} = \lambda_1 t + \lambda_1 \frac{0.0096}{0.0495}$$

$$\lambda_1 = \frac{1994}{119 \,.\, 10^6 \,.\, 94.2} = 1.78 \,.\, 10^{-7}$$

$$T = \frac{0.693}{1.78 \,.\, 10^{-7}\ 365.3} = 1.07 \,.\, 10^4 \text{ yrs.}$$

Roughly 10^4 yrs half-life of Th^{229}

α-particle energies.

Brief reference has already been made to the work of Dr. A. N. May's section on the measurement of α-particle energies of the new series.

Demers, from the photographic plate, reports energies of 5.8, 6.2, 6.9, and 8.2 Mev.[*]

Hincks and Cranshaw[**] in the early pulse-analyser work reported 5.8, 6.3, 6.9, and 8.3 Mev.

Recent values obtained by Cranshaw under better conditions in the pulse-analyser are 5.81, 6.28, 6.97, and 8.28 Mev.

In collaboration with May and Cranshaw, I made some further measurements with the pulse-analyser in which the two lower energies were compar-

---

[*] Physics monthly report, Dec. 1944.
[**] Physics monthly report, Jan. 1945.

ed with those of ThX and Tn which have values of 5.68 and 6.28 Mev, quite close to Ac^{225} and 87^{221}.

The technique was modified in an effort to obviate certain sources of error, viz.,

a) the opening of the ionisation chamber in order to interchange standard and sample,

b) the difficulty, when standard and sample are counted in the chamber together, of separating the mingled peaks without distortion.

c) the drift of the amplifiers of the pulse-analyser.

(a) and (b) were overcome by depositing very thin layers containing ThX and Ac^{225} on opposite sides of a hinged metal plate which was mounted on the bottom electrode so that by tilting the chamber the plate could be reversed and each side examined separately.

Since the interchange was easily performed, a larger number of comparison could be made and (c) was largely neutralised.

The values obtained as the mean of 10 comparison were:

Ac^{225} - $5.80_5 \pm 0.01$ Mev

87^{221} - $6.32_6 \pm 0.015$ Mev

where errors are the extreme deviations of the individual results. In these measurements, the assumption is made that there is no fine structure in the α-emission and that the peaks obtained in the pulse-analyser are the same shape. A reservation must be made in the case of 87^{221} which is discussed immediately following but the result above is not affected since it applies only to the main peak observed at 6.33 Mev. More details on these measurements are not given since it is expected that further work by Dr. A.N. May's section will soon supersede the present figures.

The α-particles of 87^{221}.

Cranshaw, in his measurements of α-particle energies in the pulse-analyser, discovered an irregularity in the α-peak of 87^{221} at 6.33 Mev. There seems to be a small peak of 6.1 Mev (Cranshaw) which contains about 20% of the number of α-particles in the normal main peak while the main 87^{221} peak at 6.33 Mev contains the remaining 80% (see Fig. 6, D and Fig. 16, I).

The possibility that this is due to branching somewhere in the series must be considered. The fact that the peak contains about 20% of the activity of a single α-emitter seems to eliminate Bi^{213} since this has been shown to branch less than 3%, and all three substances, Ac^{225}, 85^{217} and Bi^{213} seem unlikely since a 20% discrepancy has not been found in the peaks at 5.81, 6.97 or 8.28.

If 87^{221} is branched to 88^{221} (Fig.3), it should be possible to separate this radium isotope since the period will probably be convenient for chemical work. A precipitation of lead nitrate from a solution of Ac^{225} has been made in connection with Pb^{209} and gave no indication of such a substance. Such branching would probably modify the number of counts in the peak of 85^{217} producing a different peak for 86^{217} and it has been pointed out that the α-emission of 86^{217} and Po^{213} would be sufficiently close to produce α-α "coincidences" or pulses which should appear on the pulse-analyser in the region of 15 Mev. In a 50% geometry with a 20% branching, there should be 5% of the number in the Po^{213} peak taking part in coincidences. These have not been observed.

A more careful chemical search for 88^{221} will be made.

At present it seems most probable that the α-emission of 87^{221} has fine structure and Dr. May's section is considering several experiments to investigate the matter.

The assignment of Pb^{209}

One of the main points in the evidence given in this report is the discovery of a lead isotope in the series with the 3.3 hr half-life of the isotope Pb^{209} discovered by Thornton and Cork (11) in the deuteron bombardment of lead. It seems worthwhile to quote a few details from a later paper by Fajans and Voigt (14) who showed that the 3.3 hr lead was definitely produced from the isotope Pb^{208} so that the process must be Pb^{208} (d, p) Pb^{209}.

They compared the intensities of the 3.3 lead as produced in ordinary lead and in lead obtained from uraninite and correlated these with the isotopic composition of the two leads. Thus the ratios of the various isotopes in the two leads was as follows:-

Isotope	204	206	207	208
Ratio $\frac{\text{U Pb}}{\text{ord. Pb}}$	0.0007	3.99	0.249	0.00325

Since there is a great difference in the ratios, the ratio of the 3.3 hr activity need only agree in order of magnitude to establish the isotope.

In two experiments, they obtained activity ratios of 0.0035 and 0.0045 indicating a reaction on the isotope 208.

APPENDIX II

α-Counter

The arrangement was the standard equipment of Chemistry Division consisting of a parallel plate ionisation chamber, head-amplifier, linear amplifier, oscilloscope, H.T. set, discriminator and bias control, and scale of 64. Background on the chamber was kept down to one or two counts/min and a permanent standard of U^{233} gave consistent readings over many months of use. Efficiency of counting geometry was about 50%.

β-Counter

The standard equipment of Chemistry Division consisting of a Geiger-Müller counter of the end-on mica-window type mounted in a lead 'castle' and connected to a high-tension set and scale of 64. The mica window was 6 mg/cm^2 and painted with a thin layer of "Aquadag" amounting to 0.5 mg/cm^2. Background on the counter was usually about 12/min and the efficiency in the best counting position about 30%.

Thin-window counter

A Geiger-Müller counter with a mica window of 2 mg/cm^2 was supplied by Mr. N. Veall of Technical Physics Division. The geometric efficiency was about 20% and the background about 13 counts/min.

Windowless Counter

This counter, constructed by Mr. N. Veall, consists of two Geiger-Müller counters of the end-on type mounted contrapositively on a metal chamber so that the whole thing can be evacuated and filled with argon-alcohol mixture. Samples are placed inside the chamber and lie between the two counters so that counting geometry, if both counters are used, is almost 100%. Since there is no window between counters and sample (although the support of the sample obstructs one counter), α-particles and soft β-particles are not prevented from entering the counter.

Pulse-analyser and ionisation chamber (α-particles)

The "pulse-analyser" and "large chamber", a part of the equipment of Dr. A.N. May's section of Physics Division and used by them in the measurement of α-particle energies of the new series, have also been available to me in part of the chemical work when it was useful to distinguish which α-particles were present after certain treatments. A short description is given here only to make the references in this report intelligible.

The "large chamber" is a parallel plate chamber in a steel cylinder with electrodes 10 cm square, separated by 5 cm, and with a grid in front of the positive collecting electrode to eliminate the effect of the slower collection of positive ions. Samples may be laid in the centre of the bottom electrode, the chamber evacuated and filled with argon at 45 lbs/sq. in. in excess of atmospheric pressure. 1500 volts is put across the electrodes and the α-particle ionisation pulse, which is proportional to the energy of the α-particle, is passed through a head-amplifier and linear amplifier. Next in line is a "non-linear" amplifier whose function is to select, by bias control, a lower limit to the size of the pulse accepted and to amplify the overlapping portions of pulses to the required "spread" over the pulse-analyser channels.

The pulse-analyser, designed by Freundlich, Hincks and Ozeroff[★], consists of a power amplifier leading to twenty discriminating "channels" each having a scale of two and tuned consecutively in 5 volt ranges. A pulse entering the analyser after suitable amplification, affects all the

---

[★] Report not yet published.

channels up to the voltage corresponding to its energy but a cancelling circuit prevents the pulse from counting in all but the highest channel to which it reaches. Pulses coming from the ionisation chamber distribute themselves along the pulse-analyser according to the energy of the α-particle.

A graphical plot of the number of counts in each channel against the channels in order provides a picture of the distribution of α-particle energies in a sample in the chamber. It is therefore possible to work with a mixture of several α-active substances and to observe changes in each separately. The amplification of the "non-linear" amplifier can be varied from the inclusion of energies extending over about 4 Mev within the channels to about ten times greater "spread" when required.

REFERENCES

(1) Fermi, Amaldi, O. D'Agostino, Pontecorvo, Rasetti and Segre, Proc. Roy. Soc. A 149, 522 (1935).

(2) Meitner, Strassman and Hahn, Z. f. Phys. 109, 538 (1938).

(3) v. Grosse, Booth and Dunning, Phys. Rev. 59, 322 (1941).

(4) Seaborg, Gofman and Kennedy, Phys. Rev. 59, 321 (1941).

(5) Seaborg, Gofman and Stoughton, Reports A 192, June 1942, and CC 126.

(6) Hyde, Hageman and Katzin, Report CC 2636, Dec. 1944.

(7) Nishina, Yasaki, Ezoe, Kimura and Ikawa, Phys. Rev. 57, 1182 (1940).

(8) McMillan, Phys. Rev. 58, 178 (1940).

(9) Wahl and Seaborg, Report A 151.

(10) Gofman & Seaborg, Report A 352 (CN 332), October 1942.

(11) Thornton & Cork, Phys. Rev. 51, 383 (1937).

(12) Krishnan and Nahum, Proc. Camb. Phil. Soc. 36, 490 (1940).

(13) Fajans and Voigt, Phys. Rev. 60, 619, (1941).

(14) " " " " " " 626, (1941).

(15) Maurer & Ramm, Z. f. Phys. 119, 602 (1942).

(16) Ghiorso, Hageman, Katzin and Studier, Report CC 2636 (333 MLC 1907)

(17) Feather, Report Br 499, Aug. 1944.

(18) Turner, Phys. Rev. 57, 157 and 950 (1940).

(19) Russell, Phil. Mag. 46, 642 (1923).

(20) T.A.P.R. 2/45

(21) Cunningham, Report CN-991.

(22) Seaborg & Perlman, Report A 146.

(23) Mlle. M. Perey, Comptes Rendus, 208, 97 (1939).

(24) Curie, Halban and Preiswerk, Comptes Rendus 200, 1841 (1935)

(25) Hahn and Meitner, Naturwiss 23, 320 (1935).

(26) Widdowson and Russell, Phil. Mag. 48, 293 (1924)

(27) Garner, Bonner and Seaborg, Report A 278, Aug. 1942.

(28) Wahl, Science 93, 16-17 (1941)

(29) Elliott, Physics Monthly Reports, March 1945.

(30) Pose, Z. Physik 121, 293 (1943).

(31) Broda and Feather, Report Br 615, June 1945.

4n + 1 Series

INDEX

Radium and Actinium Isotopes

Bismuth and Polonium isotopes

87 and 85 isotopes

Appendix I

/sw