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The Disintegration Constant of Thorium and the Branching Ratio of Thorium C

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In age determinations of minerals in which thorium predominates, some inconsistencies have appeared when the value used for the disintegration constant of thorium is that adopted by the International Radium-Standard Commission (1930). This value differs from the original Geiger-Rutherford value by 30 percent. The present research was undertaken to determine the disintegration constant of thorium by the method of counting alpha-particles. Easily weighed sources were used. The material was obtained from a thorite from the same lot that was used by Geiger and Rutherford. The thorite contained only a small amount of uranium and correction for the ionium present was made. Various checks on the counting are given. The half-value period of thorium was found to be $1.39(10)^{10}$ years and the branching ratio, thorium C′ to thorium (C+C′), is 0.663. A method of impounding the thoron, yet permitting alpha-particles to escape, is described.

INTRODUCTION

THE determination of the age of radioactive minerals demands several constants but mainly the constants of disintegration of uranium and of thorium. Radioactive minerals from the same locality should yield the same age determination whether the minerals are those containing chiefly thorium, e.g. the monazites, or are those having uranium in predominance, e.g. the uraninites. Considerable disagreement, however, is found when the value of the disintegration constant of thorium used is that corresponding to the half-value period of $1.8(10)^{10}$ years, which is the value adopted by the International Radium-Standard Commission in their report of 1930.[1] The first determination of the disintegration constant of thorium was made by Geiger and Rutherford[2] with a value corresponding to $T=1.3(10)^{10}$ years, the use of which value gives better age agreement but it differs from the accepted value above by 30 percent. The disagreement in age determinations is not due to the uranium constant.

Geiger and Rutherford obtained their material as thorium oxide from a thorite and counted scintillations of the order of 3000. They began counting about five weeks after the material was chemically separated from the mineral and, therefore, in their material there was nearly a practical radioactive equilibrium between thorium and its isotope, radiothorium. Mesothorium was separated. Because the chemical analysis showed a very small amount of uranium, no correction was attempted for the isotope ionium. Other older determinations[3] were made by McCoy ($T=1.8(10)^{10}$ yr.), Heimann ($T=1.5(10)^{10}$ yr.), and by Meitner ($T=2.3(10)^{10}$ yr.) who used chemically purified thorium oxide and studied the ionization produced by the radiations. The original source of the material is not stated nor the condition of radioactive equilibrium and no correction is made for ionium.

The value of the constant adopted by the International Commission is based on the deductions of Kirsch,[4] who used the data of a chemical analysis of a certain Ceylon thorianite, by Hönigschmid,[5] in which the atomic weight of the lead was found to be 207.21, i.e., that of ordinary lead. It can be shown[6] on the assumption that the original quantities changed in no other way than by radioactive disintegration of the radioactive substances present that, approximately, $\lambda_{\text{Th}} : \lambda_u = \text{Th D/Th} : \text{Ra G/U}$ and

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[1] J. de phys. et rad. **2**, 273–289 (1931); Physik. Zeits. **32**, 569–581 (1931); Phil. Mag. (7) **12**, 609–632 (1931).

[2] H. Geiger and E. Rutherford, Phil. Mag. (6) **20**, 691–698 (1910).

[3] H. N. McCoy, Phys. Rev. **1**, 401–403 (1913); Bertha Heimann, Wien. Ber. IIa, **123**, 1369–1372 (1914); L. Meitner, Physik. Zeits. **19**, 257–263 (1918).

[4] G. Kirsch, Wien. Ber. IIa **131**, 551 (1922); Naturwiss. **11**, 372 (1923); Physik. Zeits. **31**, 1017–18 (1930); Naturwiss. **18**, 1054 (1930).

[5] O. Hönigschmid, Zeits. f. Electrochem. **25**, 91–97 (1919).

[6] *The Age of the Earth. Physics of the Earth.* Vol. 4, N.R.C. Bull. 80 (1931).

λ_{Th} can be obtained in terms of λ_u and the various data. A critique of this question has been given by one of us in *The Age of the Earth*[6] and it need be here only pointed out that the Ceylon thorianites, found in geological river beds, are very uncertain as to the condition of remaining "unaltered primary minerals." It is very doubtful that the accurately obtained chemical data may be used in the above approximate relation to obtain an accurate value for the disintegration constant of thorium.

A more recent value has been obtained by Fesefeldt[7] ($T=1.3(10)^{10}$ yr.) by using the Geiger-Müller counter in counting alpha-particles from quite small sources of thorium oxide obtained from monazite sand.

In view of the variations in the values of the constant by different observers, the difficulties in the check on the age determinations of monazites and uraninites from the same locality and the fact that ionium correction has not been made in any of the determinations of the disintegration constant of thorium, we have undertaken this task, at the request of the National Research Council's committee, to consider all corrections and possible checks and to determine the constant by counting the alpha-particles. The final results from thorite material give the half-value period of thorium, $T=1.39(10)^{10}$ yr. and also the branching ratio of thorium C as 0.663.

MATERIAL

It is essential, for several reasons, that a complete history of the material used be known. Thorium is obtained from minerals in which it is in radioactive equilibrium with all its disintegration products. In the chemical separation of thorium not only thorium is separated but also its isotopes radiothorium and ionium—the latter, if any uranium is also present in the mineral. The amount of ionium depends on the uranium content and is in equilibrium with all the products of uranium. The radiothorium is in equilibrium with the thorium at the time of the chemical separation of both from the mineral but since it disintegrates to half-value in 1.9 years and is also formed from thorium through the disintegration of mesothorium whose half-value period is 6.7 years, the actual amount of radiothorium (and all subsequent products of the series) will decrease with time and reach a minimum (see curves, Fig. 2) relative to the amount of thorium, in 4.8 years, after which it increases again and becomes 50 percent of the thorium equilibrium amount in 9.0 years, 75 percent in 16.8 years, etc. If it is not known when the thorium was chemically separated from the mineral one does not know the contribution due to the radiothorium unless the experiments are so refined that a differentiation based on the ranges of the alpha-particles can be made.

The ionium correction can be made if the uranium content of the mineral is known, or, more accurately, if the radium content is measured by the radon method against some radium standard solution.

It is therefore essential to start with a mineral and, preferably, with one having a small uranium content. This was done by Rutherford and Geiger and by Fesefeldt. Our mineral was thorite and came from the same lot that Geiger and Rutherford used.[8] The chemical work was done by Dr. Jeanne Henry Cornet of Brussels while sojourning in New Haven. Special care was taken to separate out quantitatively the thorium, uranium and radium. The thorium was then changed to thorium oxide and samples of this were tested for the molecular weight which showed the thorium oxide to be pure. The radium was dissolved and several samples were made to measure the amount of radium by the radon method. The radium-standard solution of the laboratory was previously prepared by careful gamma-ray measurements and was checked against a solution from the Reichsantalt (Berlin). The radium (mineral) activity is also the activity of the ionium. The analysis of the thorite gave for the important constituents as follows, in percent: thorium 42.21, uranium 0.52, iron 2.64, silicon 27.38.

The specimens used in the experiments were prepared on aluminum disks 8 cm in diameter in the form of quite uniform layers but varying in thickness for the different samples. The pro-

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[7] Hans Fesefeldt, Zeits. f. Physik **86**, 605–610 (1933).

[8] Two samples were given to one of us by Geiger in 1910 from a lot collected by B. B. Boltwood. Although we are not certain, we believe that they were Norwegian thorite specimens.

cedure was as follows. The oxide was ground to fine powder in an agate mortar wet with a small amount of chloroform. It was then transferred to a test tube containing about 20 cm^3 of chloroform and after shaking it was decanted into a second test tube where it left behind a small quantity of oxide in larger granules. It was found unnecessary to repeat with a third test tube but by further shaking all thorium oxide was found to be held in suspension before pouring it over the aluminum disk. A special circular metal vessel was made with inner walls curved and with a recess in the base for the aluminum disk so that the surface of the disk and the bottom surface of the vessel formed a plane. A weight with three legs having needle-point feet, which came on the disk near its circumference, held the disk in place. Enough chloroform was placed in the dish to cover the disk about 1 mm before the oxide from the test tube was poured over. In this manner waves were diminished, striations ordinarily found in the deposited layer were obviated and very uniform layers were obtained. The material was then left undisturbed, protected from dust, etc., until the chloroform evaporated and then the metal dish, cleaned of its contents in the meanwhile, was heated and the disk with the layer of thorium oxide was placed in it on a suitable ring to evaporate any small quantity of liquids that might still remain in the oxide. The specimen was then weighed to determine its surface density, the aluminum disk, cleaned and dried by warming, had been previously weighed. A suitable cylinder with a knife-edge end was placed centrally on the material and disk and the outside rim of oxide was carefully removed. After warming the specimen it was weighed again. Several such performances, with decreasing but known size of the cylinder, were done for the purpose of obtaining data on the uniformity of the density of the layer and for obtaining a size suitable for the number of channels to be used in a grid which geometrically reduced the total number of alpha-particles counted to the desired amount. The mean surface densities (mg of oxide per cm^2) obtained were plotted against the distance from the center to get a curve for a slight correction in the last mean density obtained for the material of an area corresponding as nearly as possible to that part of the grid to be used in the experiment.

Since thoron gas is one of the products of the material a method had to be devised to impound it by a covering thin enough to allow the alpha-particles to emerge. Various substances were tried but the best results were obtained with celluloid and varnish. Fesefeldt used celluloid alone but somewhat thicker than we desired. We found that very thin films were very pervious to thoron. However a thin layer of varnish on a double layer of thin celluloid gave satisfactory results in nearly all cases. The films were produced as follows: 1 g of celluloid was dissolved in 200 cm^3 of amylacetate. A quantity varying between 0.1 and 0.2 cm^3 of this solution was dropped on the surface of distilled water (20 cm diameter) on which the celluloid film formed. A wire frame of appropriate size and shape was brought under the film and by cutting the film with amylacetate it could be folded and placed on an aluminum rim whose external diameter corresponded to that of the thorium oxide disk. After it was thoroughly dried it was dipped into a very dilute solution of waterproof (water-lite spar) varnish, 20 cm^3 in turpentine 300 cm^3 and by careful manipulation, and the use of centrifugal force, the excess varnish was moved to the outer edge of the aluminum rim where it was wiped off. It was then dried for several days and dipped in varnish again. The double celluloid film had a stopping power for alpha-particles of polonium of a fraction of a millimeter of air and when doubly varnished its stopping power corresponded to 2 or 3 mm air as was found by direct test with an auxiliary apparatus with a Geiger point counter. The inner edge of the rim was carefully attached to the prepared film by the use of undiluted varnish to prevent leaks at this place and the rim itself was similarly sealed to the aluminum disk with the inner circle of the thorium oxide under the film. Tests for thoron leak were immediately carried out in the regular apparatus and leaks of 1 atom of thoron per minute were easily detected. Generally no leak was observed. In some cases a big leak, due to damage to the film, occurred and the specimen was discarded. In two cases small leaks were observed but counts were taken after an appropriate study of the natural background count was made to correct the counts made in the experiment.

For the determination of the disintegration constant of thorium, as reported in this paper, *only* thorium oxide from the *thorite* was used. For purposes of checks and also for further data on the branching ratio, thorium oxide was also prepared from two commercial re-purified materials, both of which had been used by the late B. B. Boltwood in some of his experiments. One source was thorium nitrate (Welsbach) used by Boltwood as early as 1907 and the other an oxide (Eimer and Amend) obtained prior to 1918. Results from these materials will be discussed later.

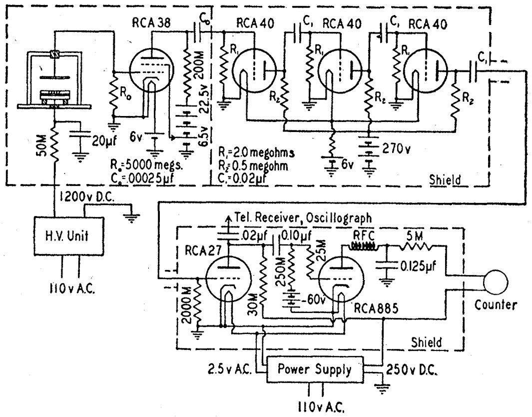

FIG. 1. Amplifier and counter circuit.

APPARATUS

Except for slight improvements the apparatus employed to count and record the alpha-particles was the same as that used in the earlier determination of the disintegration constant of uranium.[9] The alpha-particles from a specimen of thorium oxide placed in an ionization chamber passed upward through cylindrical channels in an accurately machined brass plate and produced ions in the air above. The negative ions were collected on an electrode placed about 2.0 cm from the drilled plate, since a potential difference of 1200 volts was maintained between the two. The number of ions due to each alpha-particle was sufficient to change the potential of the collecting electrode momentarily by approximately $2.5(10)^{-4}$ volt. This voltage pulse was communicated to the input of a vacuum tube amplifier whose details are shown in Fig. 1. The relay mechanism used in the uranium experiments to actuate the Cenco counter was replaced by a thyratron circuit of conventional design. This change together with the introduction of a short time-constant (pulse sharpening) coupling circuit between the first and second stages of amplification materially improved the resolution of close counts and increased the possible counting rate. Various tests indicated that counting rates as high as 150 per minute could be used safely. The accuracy of recording was checked periodically by means of a telephone receiver or an oscillograph, connected as shown in the figure.

The original drilled plate or grid (G_1) used in the uranium experiments was remeasured and found to have changed slightly in thickness, because of polishing and cleaning. With the new value of the thickness (5.110 mm) and the unchanged value of hole radius (1.962 mm) calculation showed that the number of alpha-particles emerging per channel was $N_0(0.36951)$, where N_0 is the total number of alpha-particles emitted from a square millimeter of the source. The method of calculation is given in the paper on uranium.[9] In addition to G_1 another grid, G_2, having a larger number of smaller channels, was prepared in order to check the results obtained with G_1. The second grid whose thickness was 5.084 mm had 308 channels of mean radius 0.9978 mm. The latter value is the average of direct measurement (0.9980 mm) as in the case of G_1 and calculation based on amount of metal removed by drilling (0.9976 mm). The calculated number of alpha-particles emerging per channel of G_2 was $N_0(0.02852)$. Hence the ratio of the number of alpha-particles per channel of G_2 to the number per channel of G_1 for any given source was by calculation 0.07717. The average of a large number of experimental determinations of this ratio gave 0.07708, a very satisfactory agreement.

The background or natural count of the apparatus was determined usually by placing an aluminum plate over the grid in such a way as to prevent alpha-particles from reaching the ioniza-

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[9] A. F. Kovarik and N. I. Adams, Jr., Phys. Rev. **40**, 718 (1932).

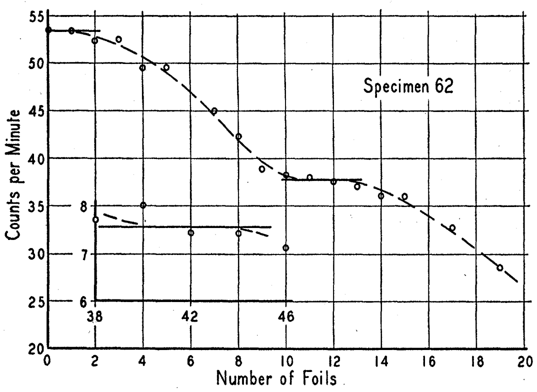

FIG. 2. Counts of particles through aluminum foils.

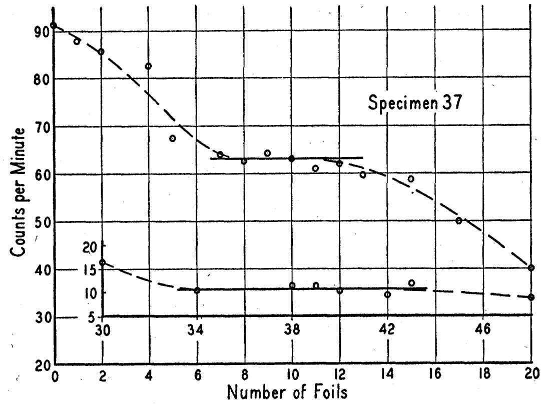

FIG. 3. Counts of particles through aluminum foils.

tion space. However, in the case of some specimens which had a slight leak of thoron it was necessary to make additional observations with the specimen removed.

METHOD OF PROCEDURE

The specimen, after reaching radioactive equilibrium, was fitted into a recess in the base of the counting chamber. One of the two grids used with a predetermined number and arrangement of channels was placed in a definite position on top of the specimen. A certain number of positions of the grid relative to the material were required to give opportunity for every part of the source to emit alpha-particles through the open channels. For grid G_2, having 308 small size channels, only two positions were required. For the grid G_1 the number varied from 4 to 18, depending on the arrangement of the selected channels. The naturals were counted at the beginning and the end and many times in between, in each series of counts, so that they were well distributed relative to other counts taken and in total had a long time count. The natural or background count depends, in case of alpha-particle counting, mainly on laboratory and electrical circuit disturbances and on the sensitivity of the apparatus. Since the latter was sometimes made fairly high, the records for the naturals show some variation. Of course, thoron leak brings in a variation which may change with conditions of the experiment.

The emerging alpha-particles belong to thorium, ionium, radiothorium, thorium X, thoron, thorium A, thorium C and thorium C′. Counts were obtained with the channels open and in the various positions of the grid over the specimen. If aluminum foils of moderate stopping power (2–3 mm air) are placed over the grid and successively increased in number, then at first the count will remain unchanged, but ultimately the shortest range alpha-particles will not be counted and in fact the straggling alpha-particles will first be eliminated. Because thorium and ionium alpha-particles (2.6 cm; 3.0 cm) have ranges with overlapping stragglers and because the next longer range alpha-particles, namely, those from radiothorium (4.0 cm) differ in range from those of thorium and ionium by nearly a centimeter of air, the counts for a certain number of aluminum foils will be again constant and form another horizontal step in the curve (Figs. 2, 3, 4) giving counts against number of aluminum foils. Were it not for straggling and for imperfect collimation due to size of the channels the drop in counts from one step to the next would be vertical and there would be as many steps as there are ranges of alpha-particles. Where two different ranges are close together so that the stragglers of one overlap with those of the other, differentiation between them is not possible.

In the present experiments we are interested in three steps, namely, the initial step in which all alpha-particles are counted and this will be referred to as the open step, the next following step, after eliminating alpha-particles of thorium and ionium, referred to as the first step, and

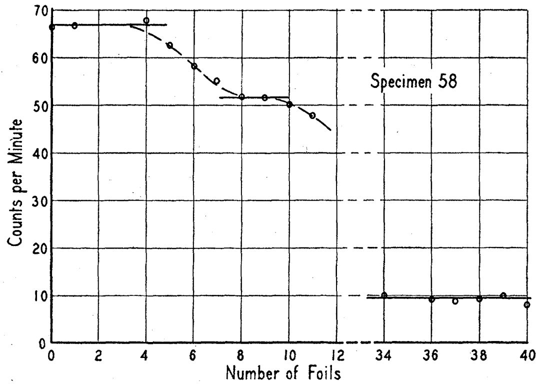

FIG. 4. Counts of particles through aluminum foils.

finally the step, referred to as the second step, when all the alpha-particles are eliminated except those of thorium C′ (8.6 cm). The count difference between the open step and the first step gives the count for the number of alpha-particles from thorium plus ionium. The net count of the second step represents the alpha-particles from thorium C′, which, with those from thorium C, would represent one-fifth of the particles given by the net count of the first step (Ra Th, Th X, Tn, Th A, Th(C+C′)).

The difference, open step less first step, when corrected for the ionium, gives the count of disintegrating thorium atoms, counted as restricted by the geometry of the grid. With the knowledge of this and the mass of the thorium involved we can obtain the value of the disintegration constant. The ratio of the net second step count to one-fifth of the net first step count gives us the branching ratio of Th C, i.e., Th C′ : Th(C+C′). The ratio of the net first step count to net open step count, when corrected for the ionium contribution, would be 5 : 6 if radiothorium were in equilibrium with thorium, as it actually is in the mineral. It will be a smaller ratio when the counts are made some time after the chemical separation of the thorium from the mineral. (See Fig. 5.)

It is, therefore, important to establish the three steps and obtain a sufficiently large count for each step and for the various positions of the grid to use all parts of the material. It is also important to have the mean date of such counts and the date of the chemical preparation, from the mineral, of the material used. Our material was prepared May 30, 1930. Because it was impossible to begin the experiments until much later it was decided to carry them on long enough to be able to check the theoretical curve through the minimum. This check is shown in curve labeled ratio r in Fig. 5. The curve drawn is the theoretical curve representing the ratio of the activity of the radiothorium and subsequent alpha-ray products, which are all in transient equilibrium, to the activity of thorium plus radiothorium and the other products. Experimentally, this ratio is the net first step count to the net open step corrected for the ionium.

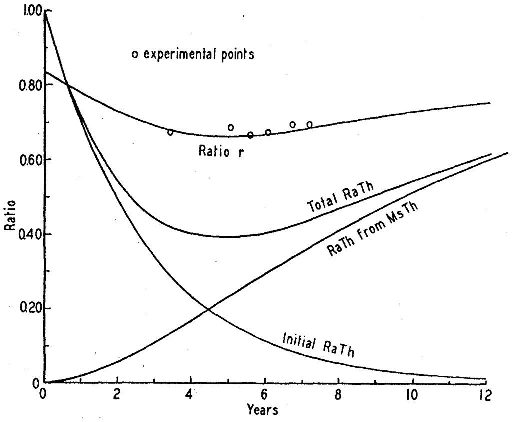

FIG. 5. Ratio of activity of radiothorium and subsequent alpha-ray products.

RESULTS AND CALCULATIONS

The data needed for the calculation are given in Table I. The mean date of the counting is given in the second column so that the age of the material used may be obtained for the calculation of the ratio $r=5\rho/(5\rho+1)$, where $\rho=$ the activity of radiothorium on that date relative to the activity of thorium or to its own equilibrium activity on May 30, 1930 when it was separated with the thorium from the mineral. The third column indicates the grid and number of channels used. The fourth column gives the mean surface density of the thorium oxide. The fifth column gives the gross count and time for each of the three important steps and the [ ] figure, where given, represents the weight assigned. The last column gives the natural or background count per minute. Generally the counts for the various positions of the grid were nearly alike and in such cases the total was obtained by adding the counts and the periods, respectively, for the various positions. In cases of largely differing counts per position, the positions were weighted alike, irrespective of actual counts.

Table II gives the data reduced to the number of alpha-particles counted per minute, per channel of the grid G_1 and per surface density of 1 mg ThO_2/cm^2. Where grid G_2 was used the calculated ratio of the number of emerging alpha-particles per one channel of G_2 and G_1, respectively, is used to make the reduction as indicated. The last column gives our estimated weight for the value in the case of each specimen. This was arrived at, on the basis of 10, considering the total counts, open and first steps, the number of positions of the grid over the specimen, the constancy of the surface density as the area was reduced and the absence (or presence) of thoron leak. Specimens 35 and 59 had leaks and special efforts were made to study the background counts to make a proper correction. Specimen 36, as its surface density indicates, was thick and some of the straggling alpha-particles did not emerge. Specimen 37 had a thick varnished celluloid film and the open count was likewise deficient so that the open step could not be obtained (see Fig. 3). In both of these cases the count for thorium was obtained from the first step averaged with the second step for which the branching ratio 0.663 was used to get the first step result. The mean value was used to obtain the open count (thorium alone) by the use of the calculated ratio r.

The ionium correction is expressed as 3.39 percent of the experimental net open count and was obtained by the radon method of measuring the radium in the thorite. After having determined the thorium plus ionium count for several specimens a preliminary determination of the thorium was obtained and the radium (=ionium) activity was expressed as percent of the combined thorium plus ionium count.

The mean weighted result for one channel of grid G_1, therefore, becomes the important figure for calculation. Other values of constants used

TABLE I. *Data used in counting alpha-particles.*

SPEC.	DATE (MEAN)	CHANNELS GRID	DENSITY $MgThO_2/cm^2$	STEP	GROSS COUNT : MINUTES	NATURAL PER MIN.
17	X·6·1933	$11G_1$	0.886	open	83351 : 3100	2.666
				1st	81672 : 4320	2.666
				2nd	1195 : 285	2.055
36	XII·9·1934	$11G_1$	3.65	open	layer too thick	
				1st	181526 : 2610[18]	2.60
		$4G_1$	3.65	1st	7976 : 300[1]	2.60
		$7G_1$	3.65	1st	16502 : 350[1]	2.60
		$11G_1$	3.65	2nd	54734 : 4843	2.60
34	III·9·1935	$22G_1$	2.656	open	88212 : 575	2.43
				1st	108993 : 1145[3]	2.43
				1st	68475 : 710[2]	3.45
				2nd	18260 : 1248[2]	2.43
		$33G_1$	2.656	open	31601 : 150	2.43
				1st	70904 : 500	2.43
				2nd	9054 : 425[1]	2.43
35*	XII·1·1935	$19G_1$	1.230	open	128598 : 2000	6.644
				1st	102865 : 2300	6.644
				2nd	33008 : 2800	6.644
37	III·12·1936	$27G_1$	1.36	(open	128722 : 1415)	2.534
				1st	100035 : 1590	2.534
				2nd	23429 : 2200	2.534
53	VII·5·1936	$36G_1$	0.352	open	42099 : 1255	2.108
				1st	38515 : 1650	2.348
62	III·4·1937	$308G_2$	0.818	open (a)	53430 : 1000[1]	2.781
				open (b)	16968 : 300[1]	2.781
				1st (a)	56427 : 1500[1]	2.781
				1st (b)	15934 : 400[1]	2.781
				2nd (total)	9081 : 1200	2.781
59*	VII·25·1937	$308G_2$	1.031	open	78798 : 1060	9.77
				1st	38638 : 730	8.87
				2nd	7337 : 640	5.85

TABLE II. *Number of alpha-particles per min. per channel.*

Spec.	open − 1st step Th + Io	3.39% (Th + Io) Ionium	Th/min/1 hole/$\sigma = \frac{1\text{ mgThO}_2}{\text{cm}^2}$		Wt.
			G_2	G_1	
17	7.982	0.271		0.7912	10
34	47.70	1.617		0.7802	8
35*	19.575	0.664		0.8092	8
53	10.443	0.354		0.7962	8
36	Deduced from 1st and 2nd steps			0.8430	4
37	" " " " " "			0.8185	8
62	15.764	0.534	$\frac{0.06045}{0.077169}$	=0.7833	10
59*	20.51	0.70	$\frac{0.062384}{0.077169}$	=0.8084	6
		Weighted mean = 0.8000			

TABLE III. *Branching ratio:* Th C′/Th(C+C′).

Spec.	$\frac{\text{net 2nd step}}{\frac{1}{2}\text{ (net 1st step)}}$	Weight
17	0.6580	10
36	0.6488	10
34	0.6636	8
35*	0.6756	8
37	0.6721	10
62	0.6685	10
59*	0.6643	6
58 commercial	0.6558	10
	Weighted mean = 0.663	

are indicated. Avogadro's number used is the recent value by S. v. Friesen[10] and 232.12 is used for the atomic weight of thorium.

The calculation of the number of alpha-particles emitted by a gram of thorium is made as follows:

0.8000 = alpha-particle per minute per 1 channel, G_1, per 1 mg ThO_2/cm^2,

$0.87884 = Th/ThO_2$,

$N \equiv$ number of alpha-particles emerging from 1 channel, G_1, per second,

$N_0 \equiv$ number of alpha-particles emitted from 1 mm² source per second,

$$N = N_0(0.3695),$$

$$\therefore \quad N_0 = \frac{0.8000/\text{min.}}{0.3695} \times \frac{1\text{ min.}}{60\text{ sec.}} \times \frac{1}{0.87884} \times \frac{10^5}{\text{gTh}}$$

$$= 4.1058(10)^3 \frac{\alpha}{\text{sec.}}/\text{gTh}.$$

The number of atoms in a gram of thorium is given by

$$\frac{6.028 \times (10)^{23}}{232.12} = 2.597(10)^{21}\text{ Th atoms per gTh}$$

and hence

$$\lambda = 4.1058(10)^3 : 2.597(10)^{21} = 1.58(10)^{-18}\text{ sec.}^{-1}$$
$$= 4.99(10)^{-11}\text{ year}^{-1}.$$

The corresponding half-value period is $T = 1.389(10)^{10}$ years. Table III gives the values of the branching ratio of thorium C for the various specimens. The last one, number 58, was a commercial (Welsbach nitrate) material. The mean weighted value of this ratio is 0.663.

DISCUSSION AND CONCLUSION

The history of the material from the thorite is completely known for purposes of checking up on the relative activity of radiothorium and its subsequent products. The experiments were carried on through the minimum value $\rho = 0.3919$ which occurred at 4.8 years and the experimentally obtained ratio $r = 5\rho/(5\rho+1)$ for various periods; i.e., the mean dates of the counting on each specimen, lie close to the curve calculated with the half-value periods of MsTh and RaTh as 6.7 years and 1.9 years, respectively. The disintegration constant is smaller than that of Geiger and Rutherford or that of Fesefeldt by 6.8 percent of their value. Since our thorite came from the same lot as Geiger and Rutherford's, it is probable that the ionium content was nearly the same in both sets of samples. If our ionium percent correction is applied to the Geiger and Rutherford half-value period ($T = 1.3(10)^{10}$ yr.) it becomes nearly $1.34(10)^{10}$ years which differs by only 3½ percent from our value—a discrepancy easily accounted for by the relatively small count they made of the alpha-particles. The analysis of our thorite gave thorium and uranium content 42.21 percent and 0.52 percent, respectively. The uranium in the thorite, in grams per gram of thorium, is, therefore, 0.0123. The radium associated with this amount of uranium is $4.18(10)^{-9}$ g. The radium separated from the thorite, measured by the radon method, was found to be $4.11(10)^{-9}$ g of

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[10] S. v. Friesen, Proc. Roy. Soc. **A160**, 424–440 (1937).

radium associated in the thorite with 1 g thorium, which is a very good check on the chemical analysis.

The method used in these experiments makes it possible to determine the count for thorium and ionium. The ionium correction has a check from the uranium content obtained by chemical analysis. The thorium count can also be obtained from first step data when the ratio $r=5\rho/(5\rho+1)$ is calculated with the knowledge of the age of the material and also from the second step with the additional knowledge of the branching ratio. This was indicated for the specimens 36 and 37. In the remaining specimens these facts were used to check the theoretical curve for the ratio r and to obtain the branching ratio. The original branching ratio was determined by Marsden and Barratt[11] as 0.65 by counting scintillations. More recently Meitner and Freitag,[12] who used the cloud chamber, obtained 0.657. Our experiments yield the value 0.663.

We also carried out long counts on two different commercial materials formerly used by Boltwood. One was Welsbach nitrate which was changed to oxide and from which radium (and mesothorium) were separated in 1930 and the other was Eimer and Amend oxide similarly treated. The radium was measured by the radon method. The complete history is not known but Boltwood's records indicate that he used some of the material in 1917 and in 1925 and may have used the Welsbach nitrate in 1905–07. Consequently, it is not known whether the radium measured was grown in some short period from the ionium or whether it was wholly or partially separated at the time of extraction from the mineral. If one assumes in the case of Eimer and Amend material that it was about 20 years old and that, when produced, the mesothorium and radium were removed, then the radium found by the radon method is the radium grown from ionium in those 20 years application of such a correction for ionium to the thorium plus ionium count gives 0.808 as against 0.8000 for the thorite material (column 4, Table II). The Welsbach nitrate material required the assumption that the radium measured actually represented the equilibrium amount in the mineral from which the material was obtained in order to give a reasonable check on the thorium count. This may be true since the material may be from monazite sand for which the radium content is of the order we measured. Under this assumption, the Welsbach material yields as a mean from two specimens 0.7939 (cf. thorite, Table II). Both materials would give fairly correct results if the assumptions made are correct. These experiments were done specifically to point out the difficulties encountered when the history of the material is incompletely known.

We used sources large enough for precise weighing. This required proper reduction of the alpha-particle count, for accuracy, which was accomplished geometrically with the channelized grid. Two grids whose channels differed widely were used. The ratio of calculated number of alpha-particles, from a given source, emerging from the two channels, respectively, received an excellent check experimentally and also the thorium count in the experiments (Table II) gives satisfactory agreement among the results obtained.

It seems to us that none of the important phases of the problem has been overlooked. Considering all the possible errors, we have concluded that the possible error in the final result for the disintegration constant is not greater than two percent and that it is smaller for the branching ratio determination.

In conclusion, we desire to express our thanks to L. Hurwitz who assisted us in some of the work, and to Dr. Jeanne Henry Cornet for the preparation of the original material as well as later preparation of some of the commercial material used for checks.

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[11] E. Marsden and T. Barratt, Proc. Phys. Soc. London **24**, 50 (1911).

[12] L. Meitner and K. Freitag, Zeits. f. Physik **37**, 481–517 (1926).