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A New Determination of the Disintegration Constant of Uranium by the Method of Counting α-Particles

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Alpha-particles emitted by a known amount of pure U_3O_8 in the form of thin films were restricted in their passage into an ionization vessel by cylindrical channels of a grid placed over the material. The method of counting α-particles alone was that of Greinacher. All α-particles entering the ionization chamber automatically registered on a mechanical counter, for which an elaborate electrical and mechanical arrangement of apparatus was used. Over 100,000 counts were made from two specimens differing in surface density in the ratio of nearly 5 to 1. The $\lambda_{\mathrm{U\,I}}$ obtained is $1.53_2(10)^{-10}$ yr^{-1} or $T=4.52_4(10)^9$ yr, a value consistent with Gleditsch's $\lambda_{\mathrm{Ra}}=4.11(10)^{-4}$ yr^{-1}, Boltwood's radium to uranium ratio of $7.40(10)^{-7}$ and branching ratio between 0.96 and 0.97.

INTRODUCTION

IN THE determination of the age of radioactive minerals two disintegration constants, in particular, are needed, namely those of uranium and thorium. For minerals containing mainly the uranium series, the disintegration constant of uranium is of prime importance while on the other hand for thorites and thorianites containing predominantly thorium, an accurate value of the thorium disintegration constant is demanded.

In the case of thorium there is a discrepancy of thirty percent in the two values usually accepted and in the case of uranium the discrepancy is from three to ten percent. Because of these rather widely differing values, the National Research Council's committee on the measurement of geological time by atomic disintegration has urged for some time the advisability of obtaining by direct laboratory methods more accurate values for these constants.

We have undertaken this work and the present paper deals with the uranium alone. The work on thorium is well under way but certain difficulties in connection with the thoron have arisen which have not yet been successfully solved.

METHOD, MATERIAL AND PROCEDURE

There are two ways in which the disintegration constant of uranium may be determined accurately. The first method was used by Geiger and Rutherford[1] when they counted the scintillations produced by the α-rays from a thin film of uranium oxide of known weight. This method is direct and involves fundamentally counting and weighing in the experimental procedure. The second method can be just as accurate as the first but it is more

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

involved. The value is given in terms of the radium disintegration constant, the radium to uranium ratio when these two members of the same radioactive series are in equilibrium—as they are in old unaltered uranium bearing minerals—and the so-called branching ratio of the actinium series to the uranium series. Since the values for the radium disintegration constant have extremes differing by ten percent, a determination by this method cannot have a greater percent accuracy than that which may be given to the radium constant. When counting scintillations was the only known way of counting α-particles the numbers counted were not always as large as the laws of probability demanded for great accuracy for various obvious reasons. Geiger and Rutherford counted only 900 scintillations from the oxide films and 2000 from the mineral films for their determination of the uranium disintegration constant. Since the first electrical automatic registration of the various kinds of rays was reported by one of us[2] using then the Geiger point counter, several new methods of attack have been developed and among these is the method of Greinacher[3] in which the ions produced by the α-particle charge up a small capacity electrode. This effect is then amplified by means of a suitable arrangement of vacuum tubes and the momentary current from the last tube is utilized in an Eindhoven galvanometer, an oscillograph, or some other indicating device. The ionization effect of the α-particle is so great for a short path in comparison to that of the β-particle or the electrons liberated by γ-rays, x-rays or cosmic rays that it is possible to adjust the sensitivity so that only the α-particles are effective. For these reasons we have adopted this method and by using a power tube as the last unit, a sensitive relay is caused to be operated by an α-ray and not by a β-ray or a γ-ray. This is important since all three types of radiations are emitted by a uranium oxide film. A second relay was made to close a local circuit containing a mechanical counter, known as the Cenco impulse counter, which automatically registered the α-rays individually as they appeared in the electric field of the ionization chamber. The number so counted can be made as large as is desirable without encountering the physical fatigue which accompanies counting scintillations.

The source was a thin film of pure U_3O_8 on an aluminum plate about 7.8 cm in diameter and 0.12 mm thick. The uranium oxide was prepared by the late B. B. Boltwood. It was a purified and many times recrystallized uranium nitrate prepared from an uraninite[4] and was labelled by him "very pure U_3O_8." The film was prepared by grinding the oxide moistened with chloroform in an agate mortar, and afterwards washing it with chloroform into a test tube making the volume of the chloroform about 20 $cm.^3$ The material was violently shaken up and quickly poured into another test tube. Particles too big to be held readily in suspension remained in the first test tube. The second test tube containing the impalpable particles was shaken

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[2] Alois F. Kovarik, Phys. Rev. [2] **13,** 272–280 (1919).

[3] H. Greinacher, Zeits. f. Physik **36,** 364–373 (1926); **44,** 319–325 (1927).

[4] The uraninite bears letters F.M.C. which no doubt refer to the exact source of the mineral. So far, we have not found in his notebooks the key to this.

and the liquid was poured over the aluminum plate placed in a brass dish in which a recess was made equivalent to the thickness of the plate. The liquid extended more than a centimeter beyond the circumference of the aluminum plate. The plate was held down by a weight on three needle points which came in contact with the plate near the circumference. On account of the quantity of chloroform used the time required for the evaporation was about one hour and the particles settled down gradually and uniformly over the plate. The time can be advantageously increased by covering the dish: thus impeding the evaporation. Under such conditions excellent films were obtained. The aluminum plate was weighed with great care before and after the deposition of the film on it, being warmed for some time previous to the weighing in each case.

The film of oxide was protected by a thin aluminum foil attached to a ring of aluminum of the same thickness as the plate. On top of this source was placed a grid through the channels of which α-particles emerged into the ionization chamber. The grid was made by drilling a machined brass plate of uniform thickness so as to produce identical cylindrical channels according to a carefully selected pattern such that if the grid was placed in successive angular positions differing by about 20° every part of the source came under some channel at some time of observation. If a strong source was used it was not desirable to have the counts per minute exceed a safe number (found by experiment) above which the mechanical device did not register accurately. In such a case a simple expedient was resorted to, some of the holes being closed by means of screw heads of appropriate size. The depth of the channel was $h=5.184$ mm and the radius was 1.962 mm. A thin aluminum foil held on a narrow brass ring was placed over the grid. This prevented ions formed in the channels from being drawn out by the electric field and thus prevented counting some α-particles which did not pass through the top opening of the channel.

The collecting electrode was 8 cm in diameter and was placed 2.5 cm from the top of the grid and approximately the same distance from the roof and the sides of the enclosing case. The case with the electrode attached could be removed readily to get at the specimen placed on an insulated table which was connected through the electromagnetic filter to the source of potential. The capacity of the electrode in this relative position to its surroundings was considerably higher than that used by other experimenters using this method of counting. We found that our amplification system and the allowance of nearly 1.5 cm as the effective range of the α-particles in the electric field made it possible to record the particles. In fact, when a polonium source was used in some of the test experiments, the collecting electrode had a diameter of 11 cm and its capacity was about 25 e.s.u. The polonium α-particles have a range considerably greater than those of U I, and the latter gave about 1.5 cm effective range in the ionization chamber. While it was possible to count the U I α-particles in this case also, it was observed that we were working near the limit of sensitivity and for this reason the capacity was reduced by making the collecting electrode smaller (8 cm). All α-particles, even those from the extreme channels used, had their total effective range entirely in the field between the collecting electrode and the aluminum foil placed over the grid.

The details of the amplifying system are indicated in Fig. 1. In order to obtain as large a signal as possible the collecting electrode of the ionization chamber was connected to the grid of a vacuum tube with small input capacity. This tube was followed by three tubes giving high voltage amplification. Following these was a power tube which actuated a sensitive relay. A less sensitive relay and finally the Cenco impulse counter followed in turn.

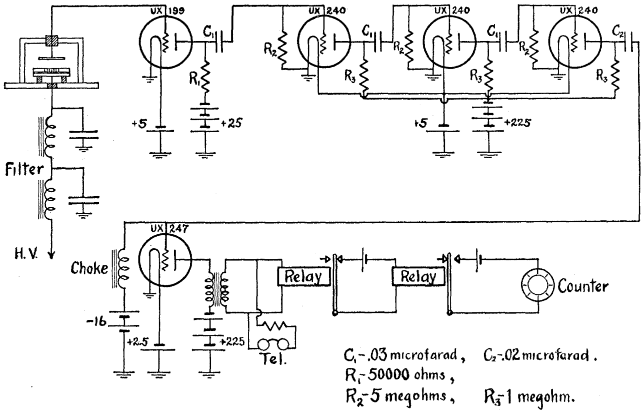

Fig. 1. Circuit diagram.

The values of the circuit constants are shown in the legend of the figure. The calculated value of the voltage amplification to the grid of the power tube is about 40,000. Hence, as the signal produced by an α-particle in the ionization chamber was of the order of $2.5 \cdot (10)^{-4}$ volt the grid swing of the power tube was of the order of 10 volts. This was ample for the operation of the relay-counter assembly.

In order to be able to determine whether the adjustment of the relays and counter were correct so that no "counts" were missed, a telephone head set with a high resistance in series was shunted across the secondary of the output transformer coupling the power tube and the first relay. This monitoring device proved a great convenience.

The counter with its relay and operating battery and the telephone head set were the only parts of the apparatus which were not housed in earthed metal boxes. Two large metal boxes, each with four compartments, were used for the apparatus. All the batteries used in the three-tube voltage amplification unit were on the floor of the compartment below that housing the vacuum tube arrangement and similarly in the case of the power tube and its sensitive relay. The large capacity condensers and choke coils, used for filtering, were on the floor of the compartment below the ionization chamber. The small dry cells used with the first stage of amplification were built compactly with the tube and placed with it.

The ionization chamber, the first stage amplification unit, the three-stage unit and the power tube unit were each separately suspended by a damping device. The damping arrangement in each case consisted of two fairly heavy boards, the upper being suspended from the roof of the box by three short elastics of good rubber tubing (sometimes several fold) and the lower board, carrying the instruments in question, being suspended from the upper board by longer rubber tubings (same material). Some experimentation was necessary before the proper lengths were found to produce the proper mechanical damping.

The boxes were covered with thick heavy sheets of hair felt. At first, vibrations produced by slamming of doors and very loud sounds of some particular vibration frequency caused disturbances but these were ultimately almost completely eliminated. The first stage of amplification was particularly susceptible to such vibrations. To insure certainty of minimum possible trouble, periods of time were chosen for experimentation—generally at night—when everything was quiet.

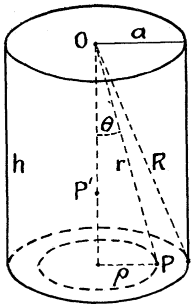

Fig. 2.

The Cenco impulse counter had to be checked for accuracy in recording α-particles. Using a uniform source, counts were obtained for the α-particles coming through various numbers of channels. The count in a unit time was found to be strictly proportional to the number of channels until the total count exceeded the mean value of about two per second. It was obvious, therefore, that in no case should the counts be a very large number per minute. Various tests indicated that it was safe to count up to 100 per minute and we arranged to use such a number of channels for a given source that about 75 per minute or less were counted.

The field in the ionization chamber was obtained from a storage battery of 4000 volts located in another room. It was found that from 800 to 1200 volts was sufficient potential difference to be used for satisfactory performance of the counting devices. Since the battery line to our apparatus acted occasionally as an antenna and since other uses were made of adjacent cells of the battery resulting sometimes in electromagnetic disturbances, we allowed the large capacity condensers to charge up and then carried on the counting for a period of ten or fifteen minutes during which the voltage of the condensers did not change appreciably. Proceeding in this manner we found we were quite free of *accidental counts* due to these extraneous causes. There were, however, possible accidental counts due to internal discharges in the condensers. These are included in the *naturals* given below.

CALCULATION OF THE NUMBER OF α-RAYS EMERGING THROUGH A CHANNEL

Since the grid is placed directly on top of the thin aluminum foil protecting the source, the source may be considered for each channel as covering the base of it. In order to calculate the proportion of the emitted rays which emerge through the circular top of the channel, we must know the solid angle subtended by the top opening at any point P of the base.

For a point P' (Fig. 2) on the axis of the cylinder at a distance x from the top,

$$\Omega_P' = 2\pi\cdot\left\{1-\left[1+\left(\frac{a}{x}\right)^2\right]^{-1/2}\right\}.$$

For P at r from O, where $x=h$, $\cos\theta=h/r$,

$$\begin{aligned}\Omega_P = 2\pi\Bigg\{&\frac{1}{2}\cdot\frac{a^2}{r^2}\cdot P_1(\cos\theta)-\frac{3}{8}\cdot\frac{a^4}{r^4}\cdot P_3(\cos\theta)\\ &+\frac{5}{16}\cdot\frac{a^6}{r^6}\cdot P_5(\cos\theta)-\frac{35}{128}\cdot\frac{a^9}{r^9}\cdot P_7(\cos\theta)\\ &+\frac{63}{256}\cdot\frac{a^{10}}{r^{10}}\cdot P_6(\cos\theta)-\text{etc.}\Bigg\}\end{aligned}$$

where P_n $(\cos\theta)$ is the zonal harmonic.

Now, if N_0 is the number of α-particles emitted from a square millimeter of the source; N, the number of α-particles emerging through the top opening of the channel; and $R=(a^2+h^2)^{1/2}$, then

$$\begin{aligned}N &= \int_0^a \frac{\Omega}{4\pi}\cdot N_0\cdot 2\pi\rho d\rho\\ &= \frac{N_0}{2}\int_h^R \Omega\cdot r\,dr\\ &= \pi a^2\cdot N_0\cdot\left(\frac{h}{a}\right)\cdot\Bigg\{\frac{1}{2}\left(\frac{a}{h}-\frac{a}{R}\right)+\frac{3}{16}\left(\frac{a}{R}\right)^3\cdot\left[\left(\frac{h}{R}\right)^2-1\right]\\ &\quad -\frac{5}{128}\cdot\left(\frac{a}{R}\right)^5\cdot\left[7\cdot\left(\frac{h}{R}\right)^4-10\left(\frac{h}{R}\right)^2+3\right]\\ &\quad +\frac{35}{2048}\cdot\left(\frac{a}{R}\right)^7\cdot\left[33\cdot\left(\frac{h}{R}\right)^6-63\left(\frac{h}{R}\right)^4+35\left(\frac{h}{R}\right)^2-5\right]\\ &\quad -\frac{63}{32768}\cdot\left(\frac{a}{R}\right)^9\cdot\left[715\left(\frac{h}{R}\right)^8-1716\left(\frac{h}{R}\right)^6+1386\left(\frac{h}{R}\right)^4\right.\\ &\quad \left.-420\left(\frac{h}{R}\right)^2+35\right]+\text{etc.}\Bigg\}\end{aligned}$$

and for the experimental dimensions:

$$a = 1.962 \text{ mm} \qquad h = 5.184 \text{ mm} \qquad R = 5.5424 \text{ mm}$$

we have

$$N = \pi a^2 \cdot N_0 \cdot [0.029824] = N_0 \cdot (0.36067).$$

EXPERIMENTAL RESULTS

Two sources were used, one having a total weight of 77.18 mg U_3O_8 with a surface density of 1.615_3 mg U_3O_8 per cm^2 and the other 17.88 mg with a surface density of 0.3759 mg U_3O_8 per cm.^{2} Counts were obtained for a time so long as to approximate to 5000 for each position of the grid. In order to show the probability variations with time, a complete record for one position of the stronger source is given in Table I.

TABLE I. *Sample record.*

Grid 0° Source: 77.18 mg U_3O_8 $\sigma = 1.615_3$ mg U_3O_8/cm^2

Count	Time	α/min.	Count	Time	α/min.
			2773	45 min.	
345	5 min.	69.0	310	5 "	62.0
283	"	56.6	320	"	64.0
309	"	61.8	319	"	63.8
300	"	60.0	334	"	66.8
333	"	66.6	306	"	61.2
302	"	60.4	325	"	65.0
307	"	61.4	317	"	63.4
302	"	60.4	297	"	59.4
292	"	58.4	325	"	65.4
2773	45 min.		5624÷90 = 62.5_3/min.		

By placing the grid in the various positions any lack of uniformity in the film is corrected, but it will be seen from Table II that the lack of uniformity could not have been appreciable. The final results for the two sources are given in Table II:

TABLE II.

77.18 mg U_3O_8 $\sigma = 1.615_3$ mg U_3O_8/cm^2; 10 channels				17.88 mg U_3O_8 $\sigma = 0.3759$ mg U_3O_8/cm^2; 34 channels			
Grid	Count	Time (min.)	Count/min.	Grid	Count	Time (min.)	Count/min.
0°	5451	70	77.87	0°	5628	90	62.53
20°	5602	75	74.69	20°	5518	"	61.31
40°	5496	70	78.51	40°	5621	"	62.46
60°	5375	"	76.79	60°	5460	"	60.67
80°	5419	"	77.41	80°	5552	"	61.69
90°	5427	"	77.54	100°	5520	"	61.33
110°	5619	"	80.27	120°	5609	"	62.33
130°	5866	75	78.21	140°	5515	"	61.28
150°	5356	70	76.51	160°	5586	"	62.07
170°	5217	"	74.53				
	54828÷710 min. = 77.22/min.				50009÷810 min. = 61.74/min.		
		Naturals =	3.71/min.			Naturals =	3.71/min.
		Net =	73.51/min.			Net =	58.03/min.

The number 3.71 per minute, labelled naturals, is a correction. It was obtained either by removing the source or by covering the grid with a thick aluminum plate which stopped all α-rays. A count of this sort was taken for about 30 minutes after each position count and sometimes also preceding it. It varied only little in value and the mean over the total time of 671 minutes was 3.71 per minute. It is due partly to contamination from the polonium source used in the original test experiments (which contamination was reduced but could not be completely removed) and partly it is due to sudden changes of charge in the condensers sufficiently great to actuate the relay.

It will be noticed that more than 50,000 counts were obtained for each specimen, i.e., over 100,000 in all. In order to average the results it is necessary to reduce the net results for each specimen to the net count per channel per minute and to a common value of surface density, e.g., $\sigma = 1$ mg U_3O_8 per cm^2 which reduction is shown in Table III. Since we desire to know the number of α-particles emitted by 1 gram of uranium per second it is necessary to perform the reduction of units.

TABLE III.

Source mg U_3O_8	$\frac{\text{Net count}}{\text{minute}}$	Channels	σ	Net count per channel per minute for $\sigma = 1$ mg U_3O_8/cm^2
77.18	73.51	10	1.615_3	4.5509
17.88	58.03	34	0.3759	4.5411
				Mean = 4.5460

$$\frac{4.5460}{1 \text{ min.} \cdot 1 \text{ mg } U_3O_8/\text{cm}^2} \times \left[\frac{1 \text{ min.}}{60 \text{ sec.}} \cdot \frac{1 \text{ mg } U_3O_8}{0.84807 \text{ mg U}} \cdot \frac{10^3 \text{ mg}}{1 \text{ gm}} \cdot \frac{100 \text{ mm}^2}{1 \text{ cm}^2}\right]$$

$$= 8.934 \times (10)^3 \frac{\alpha}{\text{sec.}} \text{ per gram U(I + II)}.$$

Since U I and U II are isotopes in radioactive equilibrium, the number of α-particles from each in a unit of time is the same. Therefore, if N and N_0 refer to the number of α-particles from U I alone,

$$N = 8.934(10)^3/2 = N_0(0.36067)$$

from which,

$N_0 = 12.385_2(10)^3$, the number of α-particles emitted by U I per second from 1 gram of U(I+II).

Using the current values of the disintegration constants of U I and of U II and values of Loschmidt's number and the atomic weight of U(I+II) we get that 1 gm U(I+II) contains

$$\frac{0.9999_3 \times 6.06_4(10)^{23}}{238.17} \text{ U I atoms} = 2.546_1 \text{ U I atoms}.$$

Therefore

$$\lambda_{\mathrm{UI}} = \frac{12.385_2(10)^3}{2.546_1(10)^{21}}\ \mathrm{sec.}^{-1} = 4.865(10)^{-18}\ \mathrm{sec.}^{-1} = 1.53_2(10)^{-10}\ \mathrm{year}^{-1}$$

corresponding to a half-value period of $T=4.52_4(10)^9$ years.

It is interesting to see what check this gives on the value of the disintegration constant of radium. Professor Ellen Gleditsch's[5] value for λ_{Ra} is 4.11 $(10)^{-4}$ yr^{-1}; B. B. Boltwood's ratio of radium to uranium in unaltered primary uraninites is $3.40(10)^{-7}$.

If we take the branching ratio to be

$$0.97\ \text{(Hahn-Meitner)},\ \lambda_{\mathrm{UI}}=1.52(10)^{-10}\ \mathrm{yr}^{-1};$$

and if we take the value

$$0.96\ \text{(v. Grosse)},\ \lambda_{\mathrm{UI}}=1.53_5(10)^{-10}\ \mathrm{yr}^{-1}.$$

Whichever ratio is taken, our value seems to be in excellent agreement with Gleditsch's value for the disintegration constant of radium. Since the $\lambda_{\mathrm{Ra}} = 4.11(10)^{-4}$ yr^{-1} gives for the number of α-particles emitted per second from a gram of radium $Z=3.49_3(10)^{10}$ it would appear that this value, rather than the high value[6] $Z=3.70(10)^{10}$, is checked by our experiments.

In conclusion we desire to express our thanks to the Bell Telephone Laboratories for a gift of one of the relays used which was made specially for us. The other relay used is also their product.

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[5] See Radioactivity, N. R. C. bulletin No. 51 (1925), and Age of the Earth, N. R. C. bulletin No. 80 (1931).

[6] Jour. de Physique [7] **2**, 273–289 (1931).