DIRECT MEASUREMENT OF ALPHA PARTICLE ACTIVITIES OF ROCKS AND THE DETERMINATION OF THORIUM

Thesis by

Russell W. Raitt

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ABSTRACT

The most important applications of the measurements of radioactivity in rocks require knowledge of only one quantity, the rate of production of alpha particles.

The common procedure has been to calculate this quantity from the radium and thorium concentrations determined by emanation methods. This procedure involves considerable experimental difficulties, and the result is dependent on an accurate knowledge of the decay constants of radium and thorium.

The method herein described measures the total alpha activity directly and simply, by counting the alpha particles emitted into an ionization chamber from a layer of finely powdered rock whose thickness is small compared to the ranges of alpha particles in the rock. Counting of the alpha particles is accomplished by means of an FP-54 electrometer tube set at a sensitivity great enough to reveal single alpha particles, which are recorded as sudden deflections of the recording galvanometer.

This method was applied to fourteen cosmic ray rocks whose radium content was determined by the direct fusion method. Measurement of the total alpha activities of these rocks has yielded their thorium content and enabled a correlation with local ionization to be made based on both the uranium and the thorium series.

I INTRODUCTION

The problem of the determination of the concentration of radioactive materials in the earth's crust has interested many investigators ever since the discovery of Elster and Geitel[1], Strutt[2], Joly[3], and others that the radioactive elements are very widely distributed throughout the rocks, soils and minerals of the earth's surface. This discovery was of considerable importance for several problems. Among other consequences of the discovery were the two major ones that radioactive energy developed within the earth is of such magnitude as to have played a significant role in the earth's thermal history and that it provided a valuable tool for the measurement of ages of rocks. The chief importance of the presence of radioactive material in the earth for our present investigation and the reason for its initiation in this laboratory lies in the fact that a very considerable fraction of the ionization produced within an unshielded cosmic ray electroscope at the surface of the ground is due to soft gamma like radiation coming from the soil and rocks beneath the electroscope. Since this radiation has about the

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[1] Elster and Geitel, Phys. Zeit. 5: 11, 321, (1904).

[2] R.J. Strutt (Lord Rayleigh), Proc. Roy. Soc. 73: 191, (1904); 77: 472, (1906); 78: 150, (1906).

[3] J. Joly, Phil. Mag. 16: 190, (1908); 22: 134, (1912).

same penetrating power as radioactive gamma radiation one might assume off-hand that it was produced by the presence of radioactive material. Some date of Cameron[4], however, gave a correlation of part of this soft radiation with the intensity of the cosmic rays proper and hence caused him to strongly suspect that part of the soft radiation was fluorescence radiation stimulated by the cosmic rays. This suspicion made it of interest to attempt the actual measurement of the concentration of radioactive elements in the material on which the electroscope was placed, in the hope that such measurements would indicate whether or not the soft radiation could be wholly caused by radioactivity.

Investigations were carried out in this laboratory by Bohn[5], Mc Millan[6], and Evans[7], culminating in the development by Evans of a satisfactory method for the determination of radium. A thorough description of the method and a discussion and criticism of the prior work in the field are given in the thesis and publications of Evans and need not be repeated here.

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4. G. Harvey Cameron, Ph D. Thesis, C.I.T. (1926).
5. J. Lloyd Bohn, Thesis, C.I.T. (1928); Jour. Frank. Inst. 210: 461, (1930).
6. E. Mc Millan, Thesis, C.I.T. (1929).
7. R.D. Evans, Thesis, C.I.T. (1932); Rev. Sci. Inst. 4: 216, 233, (1933).

II RADIUM DETERMINATIONS

Using Evan's method the writer has measured the radium content of a suite of rocks which were taken from the sites of a number of cosmic ray observations at various points throughout the world. Using an improved apparatus, but of fundamentally the same design, at Berkeley, Evans and his co-workers have also measured the radium content of the same suite of rocks, and a number of additional cosmic ray rocks for which a special technique was necessary. The results of this work on radium are to be published in a forthcoming number of the Physical Review.

Table I presents the radium measurements of the writer. Column one of this table designates the specimen number; column two gives the results of all individual runs, the designated probable errors being calculated from the deviations of the hourly measurements of ionization current; column three lists the means of the individual check runs.

The radium content is expressed in units of 10^{-12} gms of radium per gm of rock.

These rocks are described by their collectors as follows:-

C1	Gravel from top of Pike's Peak from electroscope site Sept. 9 described as "rubble" in record book.
C2	Gravel from edge of roadway on which electroscope stood in third test on Mt. Manitou, Sept. 11,1930.
C3	Glacial rubble on which electroscope sat in test from 1 h 13 m to 2:54 P.M. on Aug. 30, 1930 at Churchill, Manitoba.
C4	Glacial sand from spot on which electroscope sat in tests from 11:11 A.M. to 12:55 P.M. on Aug. 30, 1930, at Churchill, Manitoba.
C5	Gravel on which electroscope sat at Windy Point by railroad track; probably imported gravel.
C6	Igneous rock at Churchill which showed the very low activity.
C7	Samples of the rock on which the first "open" test was made on Sept. 9, 1930, on top of Pike's Peak. This is native rock on Pike's Peak in boulders a foot or more through.
C8	Samples of the underlying hard granite outcrop at Seven Sisters, Winnipeg, tested on Sept. 5, 1930. Unlike the Churchill igneous outcrop this showed high activity though on a rock in the middle of the river.
C9	Samples chipped off from the heavily weathered boulder on which the second observation on Mt. Manitou was made Sept. 11. 1930. Also like the boulder on which first test was made.
C10	Gravel from Minneapolis gravel pit. Superior Ltd., one

	mile from city limits inside, or 3 miles from loop.
C11	Minneapolis Limestone Quarry, Sept. 6, 1930.
C12	Quartzose Sandstone, Zion Canyon, Utah, 4 miles east of U.P. Lodge. Boulder at end of road.
C13	These fragments of rock were chipped from the boulder several feet in diameter on which electroscope was set to measure the local radiation from the rocks about Glen Cove, Colo. Alt. 11425 ft. on the auto highway to Pike's Peak, Sept. 19, 1932.
C14	Sample from outside Weather Bureau Sta. at Ellendale immediately underneath where No. 3 electroscope stood on the top of battery box (½ in. pine) 2 x 1 ft.
C15	Earth taken from Felts Field, Spokane, just beneath where electroscope No. 3 sat first on soil, second on one box 1 ft. high, closed top and bottom with ½ in. pine, third on two such boxes or about 26 in. high.
C16	These are samples of the limestone or marble blocks on which electroscopes were set Friday morning Sept. 9, 1932, when but 44. 6 ions were obtained at a place where the cosmic rays alone amount to 38 ions. Locale is mile 39 on the Hudson's Bay R.R.
C17	Riverside limestone taken from deep horizontal shaft in pure limestone.
C18	Kaibab limestone. Rocky point with railing perhaps 500' SE of U.P. Hotel, North Rim, Grand Canyon.
C19	Volcanic tuff from Arequipa, Peru, Nov. 1932.
C20	Surface rock from Crucero Alto, Peru, Dec. 1932.

Table I. Radium determinations by the writer.

Specimen designation	(Ra-content) x 10^{12} Individual runs	Means
C1	0.46 ± .01	
	0.47 ± .03	0.47
C2	0.36 ± .02	
	0.37 ± .02	0.37
C5	0.77 ± .02	
	1.07 ± .03	0.92
C6	0.17 ± .02	
	0.21 ± .02	0.19
C7	1.18 ± .02	
	1.33 ± .02	1.25
C8	0.26 ± .01	
	0.23 ± .02	0.25
C9	1.23 ± .03	
	1.40 ± .04	1.32
C10	0.36 ± .02	
	0.37 ± .02	0.37
C12	0.31 ± .02	
	0.34 ± .03	
	0.31 ± .01	0.32
C13	1.88 ± .04	
	1.89 ± .03	1.89
C19	1.39 ± .04	
	1.54 ± .02	1.47

The results of Evans at Berkeley, who extended the measurements to a larger number of rocks but did not make check runs on each specimen, are presented in Table II.

Table II. Radium determinations by Evans.

Specimen designation	(Ra-content) x 10^{12}
C1	0.41
C2	0.25
C3	0.33
C4	0.44
C5	0.52
C6	0.23
C7	1.52
C8	0.19
C9	1.34
C10	0.48
C11	0.60
C12	0.32
C13	1.83
C14	0.78
C15	0.68
C16	0.11
C17	0.32
C18	0.95
C19	1.75
C20	2.38

Comparison of the two sets of data gives an adequate cross-check of the method with the two apparatus and extension of this work hardly seemed justified, particularly in view of the fact that the Berkeley apparatus had undergone considerable advancement in ease and efficiency of operation. The determination of the cosmic ray rocks as a whole was therefore completed only at Berkeley. For the sake of consistency, it seems to the writer to be best in considering the rocks as a whole to use the data from the apparatus on which all of the rocks have been measured. Therefore, unless otherwise mentioned, the radium concentrations used in the succeeding pages will be those of Evans.

III THORIUM EMANATION METHODS

While the work of Evans can be said to have satisfactorily solved the problem of the determination of radium, the original goal of the determination of all the radioactive constituents of the rock was only partly attained by this success. There remained the problem of the determination of the thorium series. The actinium series, as is known from the work of v. Grosse[8] and others has a constant activity ratio with respect to uranium. From the radium measurements the concentration of all the constituents of the uranium and actinium series are readily calculated, providing of course, that sufficient time has elapsed since the origin of the rock to allow equilibrium to take place, a condition which will be true for all but a very small minority of rocks.

It has long been known[9] that the thorium series

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8. A. v Grosse, Phys. Rev. 42: 565, (1932).
9. J. Joly, Phil. Mag. 17: 760, (1909); 18: 140, (1909); J.H.J. Poole, Phil. Mag. 29: 483, (1915). H. Mache and M. Bamberger, Wien Ber. IIa, 123: 325, (1914).

is generally prevalent in the surface rocks of the earth at a concentration such that its activity is comparable to that of the uranium series. Hence the effect of the thorium series in any measurement utilizing such properties of radioactive elements as the emission of gamma rays, the production of helium and of lead, and the generation of heat, cannot be neglected if the measurements are to have more than a qualitative significance.

Difficulties with Emanation Methods.

On examination of the literature in the field of the determination of thorium in rocks,[9,10] one finds a very unsatisfactory situation with respect to methods of analysis and reliability of the results. All of the methods which antedate the work of the present investigation are similar to the method for determining radium in that they are emanation methods, that is, they depend upon removing the thoron from the rock and transferring it to an ionization chamber for measurement.

Due to the very rapid decay of thoron (half-period 54.5 seconds) it cannot be merely extracted from the rock, transferred to an ionization chamber, where its decay can be observed at the leisure of the observer.

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[10] J.H.J. Poole and J. Joly, Phil. Mag. 48: 819, (1924);
J.H.J. Poole, Phil. Mag. 3: 1246, (1927).
A.L. Fletcher, Phil. Mag. 23: 279, (1912).

Since the life of thoron is small in comparison to the time necessary to measure the very small ionization currents produced by the average equilibrium amounts of thoron in rocks, it is necessary to use some sort of flow method whereby the thoron continuously generated by the thorium X of the rock is continuously transported to the ionization chamber where it decays and its current can be measured. Such a method was devised by Strutt[11] and was used and improved by Poole and Joly[9,10] in particular, as well as by other observers[12].

In this method the rock is dissolved and the solution is either refluxed or agitated while a stream of de-emanated air, or other emanation-free gas, is passed through the solution into an ionization chamber from whence it exhausts into the atmosphere. It is clear that the ionization current is a function of the gas velocity and that there will be a certain optimum velocity at which it will be a maximum. The shape of this curve has been calculated by Poole and Joly[13] and by Evans[14] and both theory and experiment show that the ionization is uncritical to small velocity variation at the optimum velocity.

Variations in other conditions such as, reflux

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[11] R. J. Strutt, Proc. Roy. Soc. 76A: 88, (1905).
[12] Nat. Res. Council Bull. 80: Age of the Earth. (1931).
[13] J.H.J. Poole and J. Joly, Phil. Mag. 48: 819, (1924).
[14] R. D. Evans, Rev. Sci. Inst. 6: 99, (1935).

rate, emanating power of solution, absorption of thoron in the large quantities of drying agent required, have a more serious effect on the readings. Nevertheless it is possible by observing proper precautions to obtain reasonably satisfactory results for most purposes providing that the thorium X which is the direct producer of the thoron is completely dissolved and there are no precipitates or other matter in the solution to absorb the thoron or otherwise reduce the emanating power. That this ideal situation can rarely, if ever, be attained in the average rock solution is made clear from the considerable discussion on this point in the literature. It is almost if not completely impossible to obtain a clear, limpid, solution of a rock and even if it were obtained under very special conditions it is hardly likely that it would remain stable during the long period of refluxing and gas washing necessary to obtain accurate readings of the ionization current.

Obviously, the objections that apply to the use of the solution method in the determination of radium in rocks[7] are much more seriously applicable to the use of the same technique in the determination of thorium and in addition several other difficulties of a serious nature are introduced.

The natural extension of the direct fusion method to the measurement of the thorium concentration in rocks lay in the direct application of the streaming method. This could be simply done by passing a stream of inert gas at a steady rate through the furnace containing the fused rock sample and into the ionization chamber of the radon apparatus.

Since the Berkeley apparatus had received considerable development in the way of furnace design and simplification of operation this extension of the fusion method was taken up by Evans while the writer adopted an entirely different mode of approach to the problem. The results of Evans work have been recently published[14] and have demonstrated the failure of the direct fusion method to give a reliable method for determination of the thorium. The reason lies in the distillation of the rock from hot parts to the cold parts of the furnace, thus being deposited in a state of low and uncertain emanating power.

IV DETERMINATION OF TOTAL ALPHA ACTIVITY

Advantages of New Method.

The mode of attack adopted by the writer was the direct determination of the total alpha particle activity of the rock. The activity due to the uranium and actinium series is calculable from the radium concentration and the subtraction of this from the total gives the activity of the thorium series. From this figure the thorium concentration can be readily calculated with the use of the disintegration constant of thorium.

For the two major fields of application of the measurement of radioactivity in rocks, ages of rocks and the radioactive generation of heat, the value of the rate of alpha particle emission per gram of rock plays the major role.

(1) Age determinations.

The helium method for radioactive age determination[15], which is the only possible method available for ordinary rocks, determines the age of a rock specimen by dividing the helium concentration by the rate of production of helium, the latter quantity being the total rate of production of alpha particles. As the half lives of uranium and thorium are longer than 4 x 10^9 years, the assumption of constant rate of production of helium in the rock since its birth involves negligible error for rocks less than 10^9 years old. For rocks older than this, the age can be calculated using the exponential low of decay, and the error caused by lack of knowledge of the Th/U ratio

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[15] W.D. Urry, Chem. Rev. 13: 305, (1935).

will be no more than 5% for rocks of age 2 x 10^9 years, and Th/U ratios ranging between zero and infinity. This age represents the extreme upper limit for ages of terrestrial rocks and the uncertainty of 5% is probably considerably less than the experimental uncertainty involved in the application of the helium method to rocks of this great age.

(2) Heat generation.

The value of the alpha ray method for the determination of the radioactive generation of heat in rocks is found in the fact that most of the energy of radioactive decay is expended by the disintegration alpha particles[16]. The average alpha particle of the uranium series has an energy[17] of 5.29 x 10^6 electron volts while that of the thorium series has an energy of 6.00 x 10^6 electron volts. The average alpha particle for both series has 5. 59 x 10^6 electron volts energy. The total emission of energy includes also a fractional part of beta and gamma radiation. One atom of uranium I in its transformation into lead emits altogether an energy of 45 x 10^6 electron volts[18]. This gives an average energy of 5.6 x 10^6 electron volts per alpha particle of the

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[16] Rutherford, Chadwick and Ellis, "Radiation from Radioactive Substances". p. 161. Cambridge, (1930).

[17] loc. cit. p. 86.

[18] loc. cit. p. 163.

uranium series. The total beta and gamma activity of the thorium series is not known with any accuracy, but if we assume that it bears the same ratio to the alpha activity as the uranium series, we get finally for the average energy generated per alpha particle emitted by both the thorium and uranium series, the value of 5.9×10^6 electron volts. Since one electron volt is 1.59×10^{-12} ergs, we can calculate the rate of energy production in ergs by multiplying the rate of alpha-particle production by the figure 9.4×10^{-6}. The error that will be introduced by ignorance of the Th/U ratio will be small and in general less than the variations from sample to sample of the radioactivity of an average rock mass.

(3) Correlation of "Local Radiation".

If we may assume that the gamma radiation of the thorium series has approximately the same relative intensity with respect to the emission of alpha particles as that of the uranium series, then we may apply the data on total alpha activity directly to the problem of the correlation of the "local radiation" of cosmic ray observations with the radioactivity of the underlying terrain. However there are not enough data in the literature to test the validity of this assumption. And we must therefore give separate consideration to both the uranium and thorium series in this problem.

Accordingly, the final aim of the investigation became the determination of the total alpha activity of the cosmic ray rocks, whose radium content was already known. It is the purpose of this thesis to describe the method used and the results of this method as obtained on the cosmic ray rocks. These results have therefore not only tested the method but have solved the experimental problem which initiated the radioactive work at this laboratory.

Methods of Counting Alpha Particles.

By means of his duant electrometer, which he developed to a very high sensitivity, Hoffmann[19] was able to detect, by means of a considerable deflection of the electrometer needle, the sudden ionization increase caused by the entrance of an alpha particle into his ionization chamber. In subsequent years Hoffmann[20] and his co-workers[21,22,23] have improved the method

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[19] G. Hoffmann, Phys. Zeit. 13: 480, 1029, (1912).
G. Hoffmann, Ann. der Phys. 42: 1192, (1913); Elster and Geitel, Festschrift : 435, (1915); Ann der Phys. 52: 665, (1917); Ann der Phys. 62: 738, (1920); Zeit. f. Phys. 7: 254, (1921); Z.f. Phys. 25: 177, (1924); Phys. Zeit. 28: 739, (1927).

[21] M. Mader, Zeit. f. Phys. 88: 601, (1934).

[22] G. Dieck, Zeit. f. Phys. 90: 71, (1934).

[23] H. Ziegert, Zeit. f. Phys. 46: 668, (1927).

and applied it to various problems of alpha ray emission, most of them concerned with the study of distribution curves of the ionization pulses obtained from various materials in the attempt to discover new radioactive substances, or very low activities in ordinary elements. The instrument was not used, however, to obtain a quantitative measure of the concentrations of radioactive elements in rocks. In fact no experimental method for the direct determination of alpha activities of rocks antedated this investigation, to the writer's knowledge.

While the duant or Hoffmann electrometer has yielded excellent results in the hands of Hoffmann and his co-workers, it is expensive and difficult to manipulate. Furthermore, since the gamma ray and cosmic ray ionization is always a major part of the total ionization in any clean ionization chamber, the rate of drift of the electrometer at the sensitivity necessary to reveal the alpha particle kicks will be very rapid. Hence there is necessitated the added complication of introducing a compensating current by means of an induction ring the potential of which is changed at a constant rate by means of a motor-driven rheostat.

Other methods, such as the Geiger counter, have been developed which can count alpha particles and which depend for their action on the multiplication of the original ionization pulse with the ionization chamber itself by means of collision ionization or other processes. It appears to the writer, however, that the most confidence can be placed in a method which directly detects the alpha particle ionization pulse and whose results can therefore be interpreted much less ambiguously than those of other devices.

Greinacher[24] discovered that a multistage vacuum tube amplifier, the first stage of which was connected to an ionization chamber, could detect the pulses produced by single alpha particles. This method has received further development by Ortner and Stetter[25], Schmidt and Stetter[26], and especially by Wynn-Williams and Ward[27], by Ridenour of this laboratory and by Dunning[28].

This method has given excellent results and under ideal conditions would probably be adequate to measure the alpha radiation from rocks. However, in view of the complications introduced by the use of many stages, this method was discarded in favor of the use of a single stage vacuum tube amplifier using and FP-54-low-grid-current

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24. Greinacher, Zeit. f. Phys. 36: 364, (1926); 44: 319, (1927).
25. G. Ortner and G. Stetter, Phys. Zeit. 28: 70, (1927); Zeit. f. Phys. 54: 449, (1929).
26. E.A.W. Schimdt and G. Stetter, Zeit. f Phys. 55: 467, (1929).
27. C.E. Wynn-Williams and F.A.B. Ward, Proc. Roy. Soc. 131A: 391, (1931).
28. J.R. Dunning, Rev. Sci. Inst. 5: 387, (1934).

electrometer tube[29].

Vacuum Tube Electrometer

This tube has been shown by Hafstad[30] and Bearden[31] to be capable of attaining results comparable with those given by the Hoffmann electrometer in the measurement of alpha particles and protons. Furthermore the simplicity of construction and inexpensiveness of the vacuum tube set-up are very strong points in its favor.

The attainments of this FP-54 and the conditions limiting its ultimate sensitivity and the method of attaining that sensitivity are thoroughly discussed by Hafstad[30], and need not be repeated here. A sufficient sensitivity to attain the desired result of recording alpha particles is relatively easy to obtain. It is found, however, that the decline in the plate current caused by the discharge of the storage battery supplying the filament produces a galvanometer drift, which, at the necessary sensitivity, is far greater than the maximum allowable for the recording over the long periods necessary for the weak activities encountered.

Hafstad's method of reducing this drift by means

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29. Metcalf and Thompson, Phys. Rev. 36: 1489, (1930).
30. L.R. Hafstad, Phys. Rev. 40: 1044, (1932); Phys. Rev. 44: 201, (1933).
31. J.A. Bearden, Rev. Sci. Inst. 4: 271, (1933).

of a loaded counter-cell was tried by the writer and, while it was found that the drift could be reduced to any desired degree, as stated by Hafstad, nevertheless this state of affairs did not last for a very long period because the discharge curve of the counter-cell did not match that of the storage batteries. Furthermore, the long period of time required by the loaded counter-cell to reach equilibrium after an adjustment of the loading current was a considerable source of annoyance.

At this time, the balanced circuit of Du Bridge and Brown[32] appeared and was immediately adopted by the writer with a very slight modification to reduce the difficulties of obtaining a balance with the grid at "floating potential". This circuit is illustrated in Fig. 1. The galvanometer is a Leeds and Northrup type H.S. of sensitivity, the Ayrton shunt is set at 1/10. The grid leak R_9 was made by the S.S. White Dental Manufacturing Co. The variable resistances R_1 and R_6 are good quality panel-type rheostats. R_8 is a 1000 ohm potentiometer constructed from the unwound resistance element of an Electrad resistor of the "truvolt" type. Positive contact is made by means of a sliding brass collar and clamping screw.

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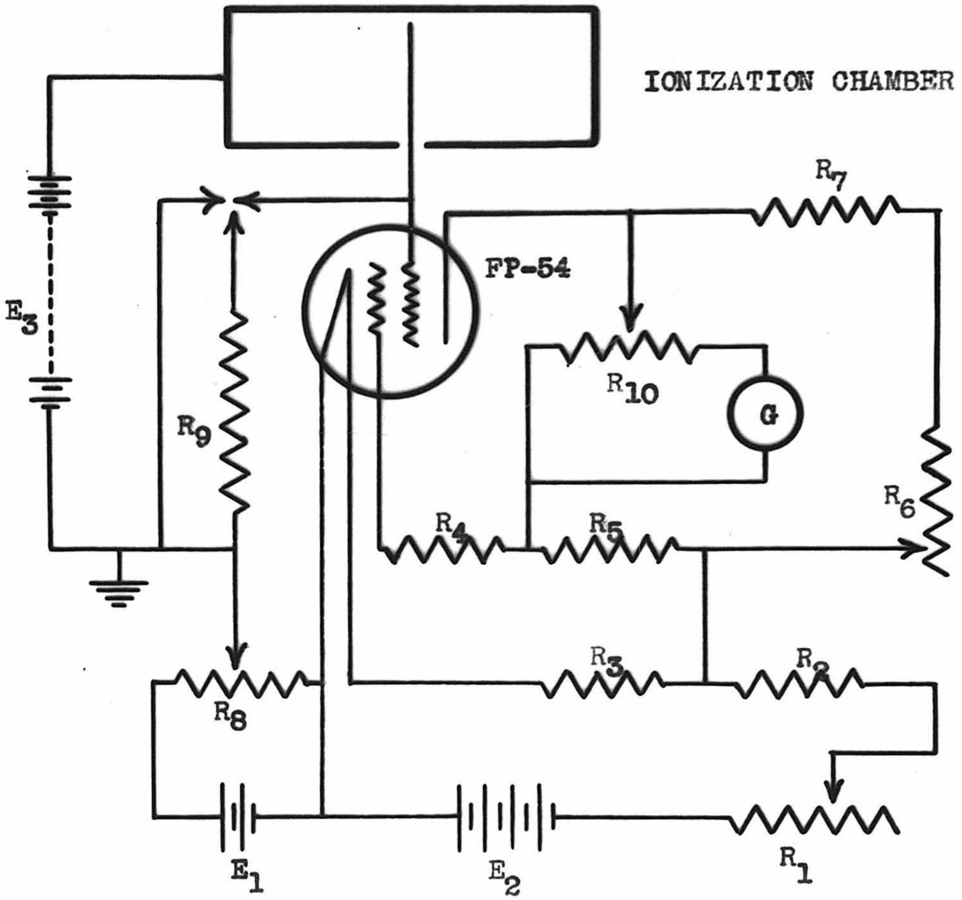
[32] L. A. Du Bridge and H. Brown, Rev. Sci. Inst. 4: 532, (1933).



R_1--12 ohms

R_2--10 ohms

R_3--40 ohms

E_1--4 volts

R_4--10,000 ohms

R_5--2,000 ohms

R_6--200 ohms

R_7--3,500 ohms

E_2--8 volts

R_8--1,000 ohms

R_9--5.3×10^{11} ohms

R_{10}--10,000 ohms

E_3--315 volts

FIG. 1. CIRCUIT DIAGRAM

All fixed resistances, with the exception of the grid resistance R_9 and the Ayrton shunt, were especially constructed for this circuit from Advance wire, and all connections, with the exception of the minimum number of sliding contacts necessary for adjustment of the circuit, were carefully soldered.

The battery, E_1 and E_2, consists of two 100-ampere-hour Exide radio batteries purchased new for this apparatus to obtain the smoothest possible delivery of current. E_3 is a bank of 7 medium sized 45 volt Eveready "B" batteries from which no appreciable current has been drawn since purchase and which are therefore very quiet in operation. The storage battery is placed in a double-walled wooden box between the inner and outer walls of which is a 1 inch layer of cotton batting. In this way, temperature fluctuations of the room, which is not thermostated as in Hafstad's work, were greatly reduced in amplitude and in rate of change.

In the first circuits constructed, ordinary wire wound resistances of the type used in radio practice were installed, and large temperature drifts as well as spontaneous instability were experienced. This trouble was diagnosed to be the result of both the high temperature co-efficient of resistance and imperfect internal connections in the resistance units. Advance wire possesses the double advantage that it can be easily soldered and has a very low temperature coefficient. On installing Advance wire resistances everywhere possible, completely adequate stability was immediately obtained. The drift of the galvanometer could be easily reduced to a very steady rate equivalent to a grid potential change of 1/10 of a millivolt per hour.

There still remained a slight temperature effect which is probably due to a temperature coefficient of the tube characteristics and could not be entirely removed without thermostating the entire apparatus. This was not necessary as the temperature effect was not serious.

Records were taken by allowing the image of the galvanometer lamp to fall on a strip of bromide paper wrapped around the drum of a recording camera which was driven by a spring clock. The drum had a circumference of 50cm and rotated at one revolution every 2 hours giving a paper speed a trifle over 4 mm per minute. As the period of the galvanometer was about 10 sec., this speed was sufficient to reveal all detail in the galvanometer deflections. The width of the paper was usually about 12 cm, and the necessarily long records were taken by setting the rate of drift at a convenient rate (usually ca. 1cm/hr.) so that as the drum turned, the galvanometer spot moved continuously across, tracing a helical path on the drum.

The rate of drift was regulated by making use of the balancing characteristic illustrated in Fig. 2. It is seen from this figure that the galvanometer deflection is a function of the filament current. As the filament current declines at a very constant rate due to the discharge of the storage battery, it is easily seen that the drift can be given any desired value by setting the filament current at the requisite point. In general the filament current is set at a point a little less than the equilibrium value. The galvanometer spot then drifts across the paper at a speed which is roughly parabolic with distance from the balance point. This is well illustrated by the sample record of Fig. 3 which is a copy of a twenty hour run with the grid resistance of 5.3×10^{11} ohms in the circuit.

Ionization Chambers.

Two types of ionization chambers were used. The first one followed the design of Hoffmann[33] and was constructed of a light framework of wire in the form of a vertical cylinder 2 cm high and 6 cm in diameter, which was rigidly supported by a brass ring on which the sample whose activity was to be examined could be so placed as to form the top of the chamber. This inner chamber was surrounded by a brass housing whose walls were far enough

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[33] G. Hoffmann, Ann.der Phys. 62, 738, (1920).

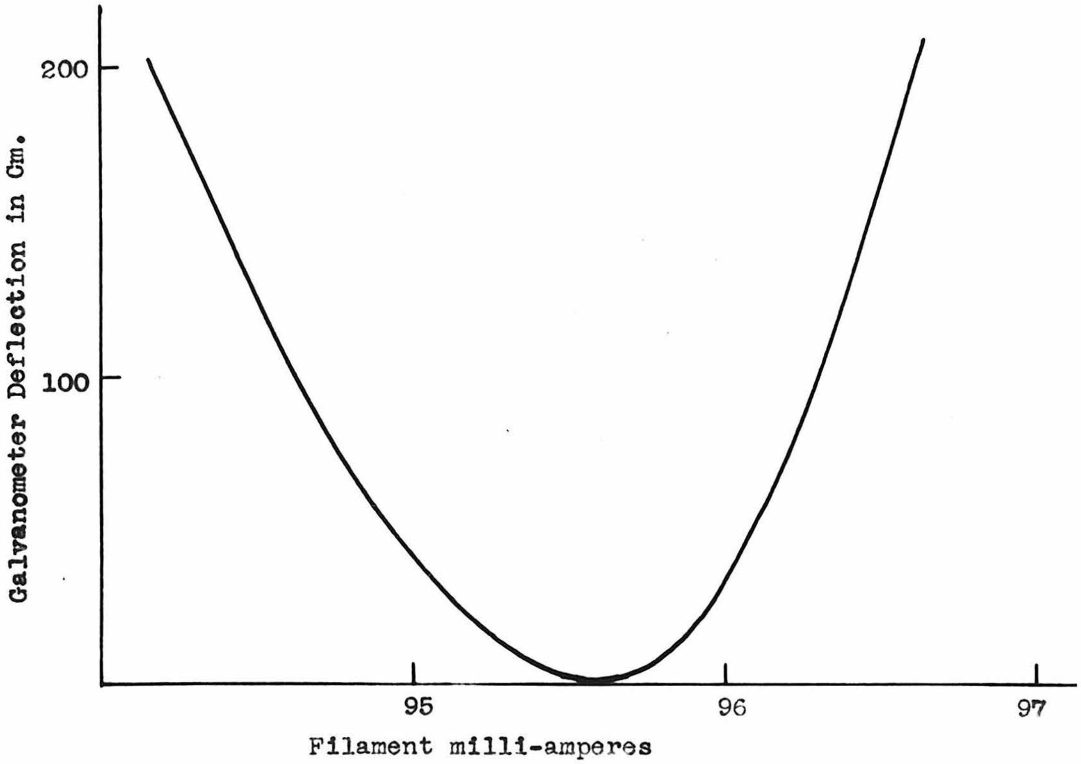

FIG. 2. BALANCING CHARACTERISTIC

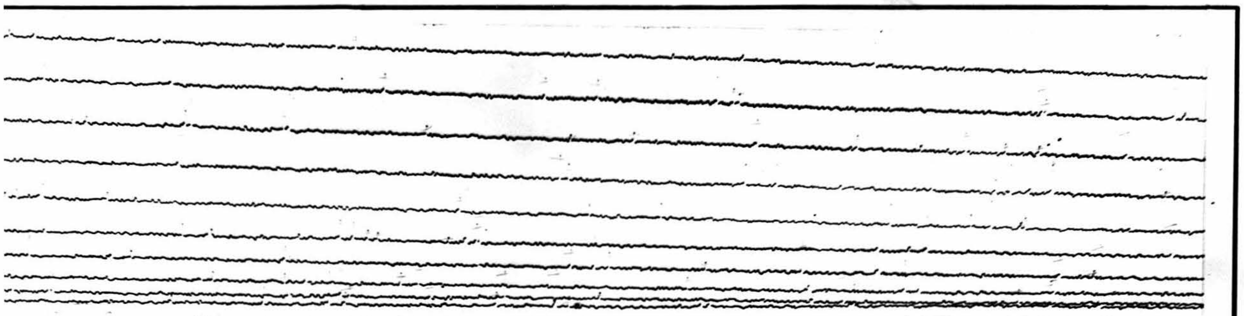

FIG. 3. BACKGROUND WITH GRID LEAK.

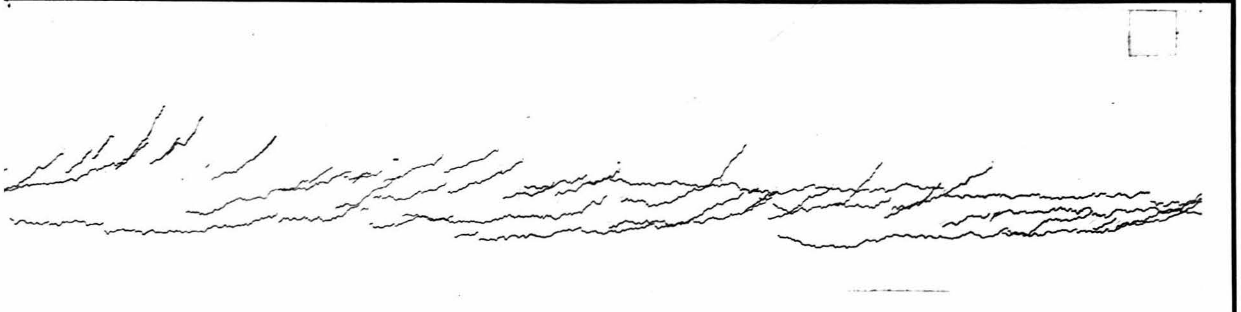

FIG. 4. BACKGROUND WITH GRID FLOATING.

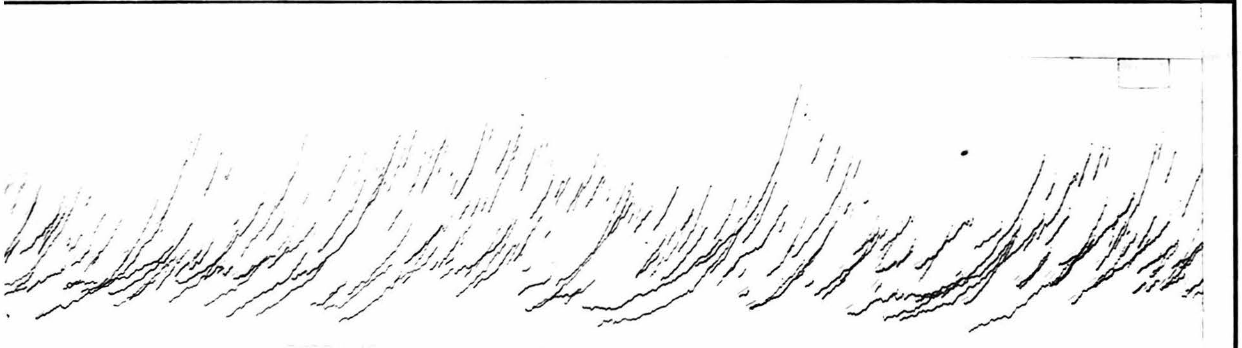

FIG. 5. ROCK RUN WITH GRID FLOATING.

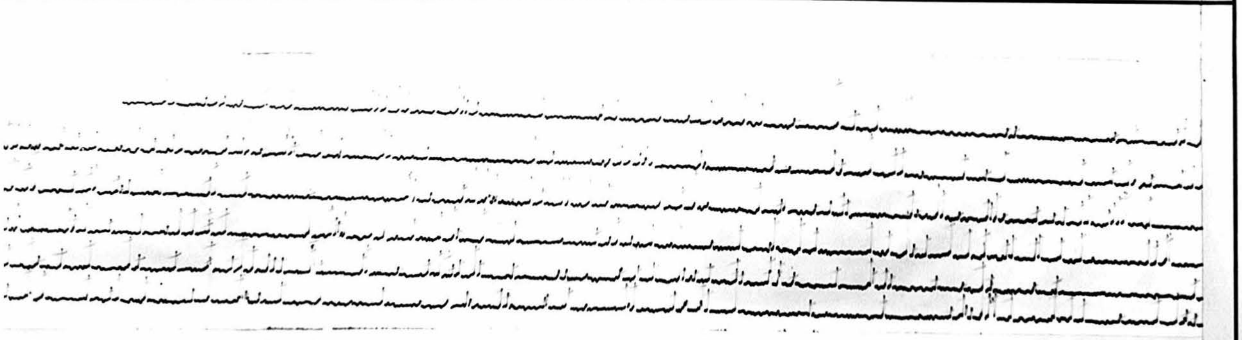

FIG. 6. ROCK RUN WITH GRID LEAK.

away to prevent all but a few of the longest alpha particles from reaching it. This outer chamber was gas tight and filled with emanation-free CO_2 which possessed the additional advantage of increasing the specific ionization of alpha particles by 50%.

The whole assembly was waxed directly to the top of the 2-inch brass tube which housed the FP-54 and which was at all times kept evacuated to a pressure of less than 2 mm of mercury in order to avoid the recording of as much contamination as possible. The inner collecting electrode communicated with the grid of the FP-54 by means of an insulating amber bushing. All records with this chamber were taken with the grid at "floating" potential, i.e. the equilibrium potential assumed by the grid when the total current to the grid is zero, and whose value was about 2 volts for the tube used by the writer. While the net grid current is zero the fact that this current is formed of two opposing currents of equal magnitude of the order of 10^{-15} amperes and formed of discrete charges, single electrons for the internal grid current, causes the grid potential to execute random fluctuations around the floating potential. The magnitude of these fluctuations has been calculated by Hafstad, whose theory correctly predicts the observations. The tube used by the writer gave somewhat greater fluctuations than those reported by Hafstad but no greater than might be expected from normal differences in grid currents of different tubes.

Figures 4 and 5 illustrate records of typical runs at floating potential. Fig. 4 is a background run taken with a clean brass plate covering the top of the ionization chamber and Fig. 5 is the record of a run taken when a thin film of a finely ground rock was depostied on the under side of the brass plate, exposed to the ionization chamber.

Figures 3 and 6 of the same photograph are runs made with the grid resistance of 5.3×10^{11} ohms connected, grid bias still maintained at floating potential. The input period is reduced from about 5 minutes to about 5 seconds, thereby causing the grid voltage to remain close to the equilibrium value instead of wandering rather widely as is the case with floating grid. The greatly improved ease of counting, especially for high activities, is apparent. The charge sensitivity is reduced 50% but this is not an important disadvantage.

In order to illustrate that the fluctuations observed are genuine grid fluctuations and not due to a defective circuit we can compare sections of two records photographed at natural size. Fig. 7 is a 16-hour record taken with grid and electrode floating, with a pressure of

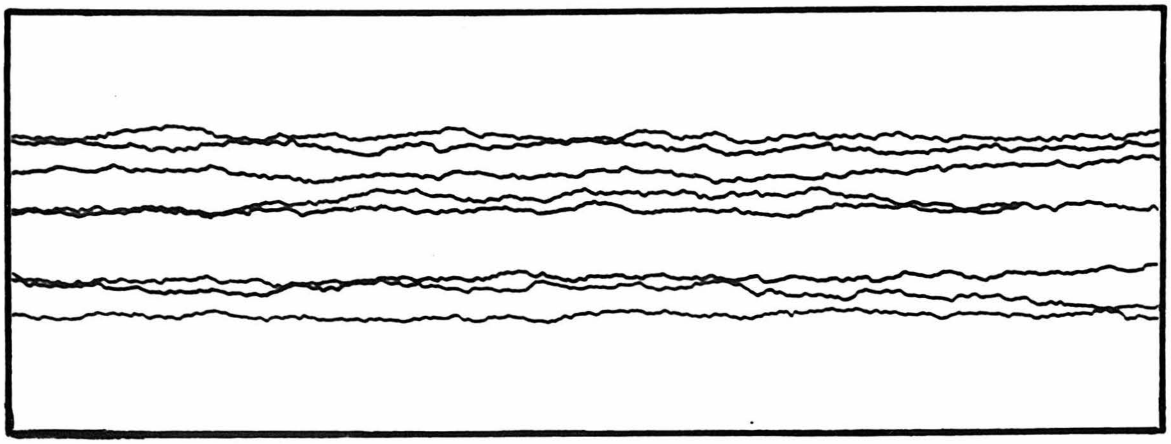

FIG. 7. GRID FLOATING IN VACUUM

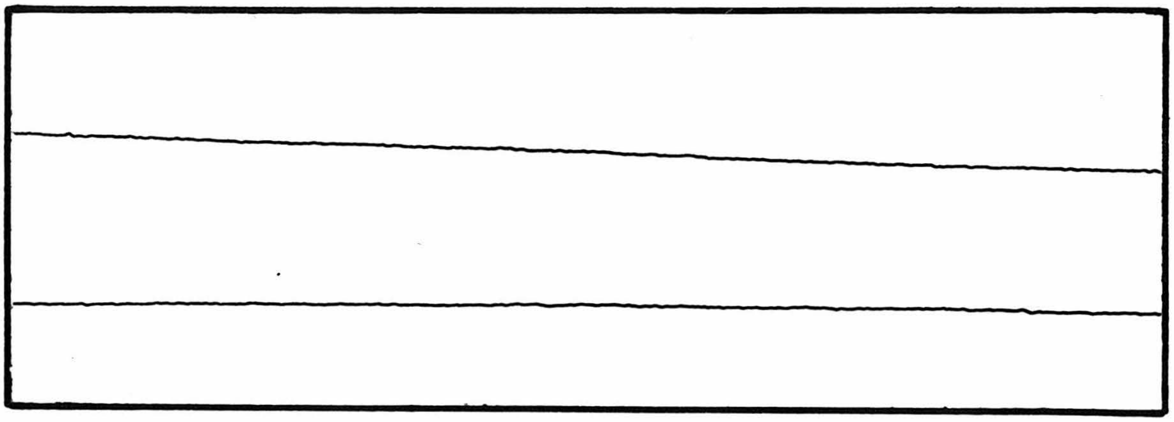

FIG. 8. GRID GROUNDED

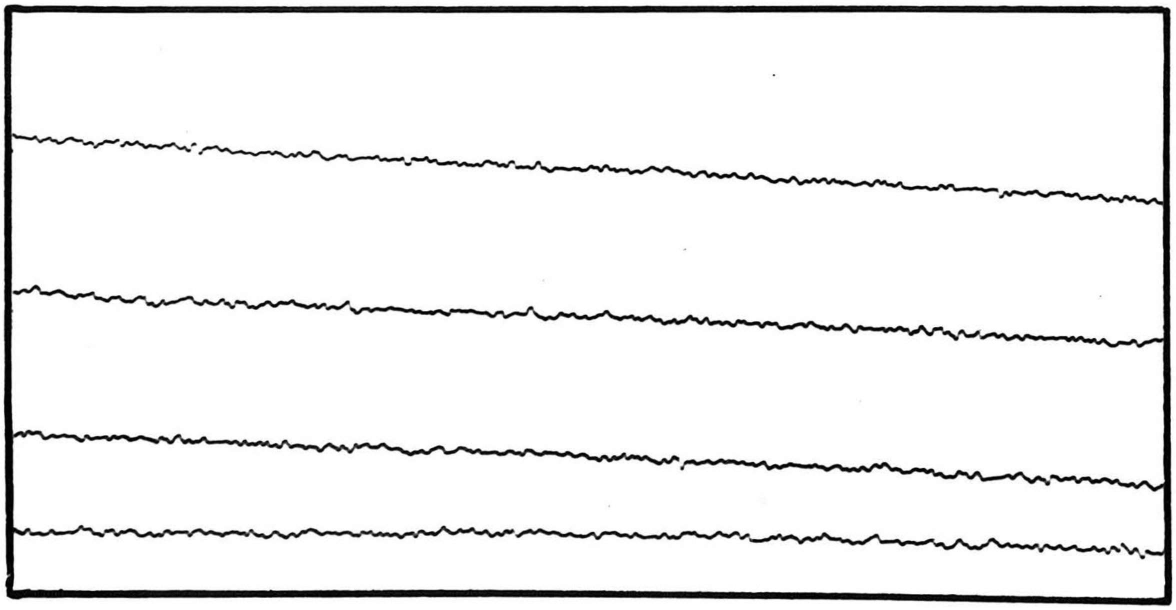

FIG. 9. GRID LEAK CONNECTED; CHAMBER EVACUATED

2 mm in the ionization chamber and a collecting potential of 315 volts. The voltage sensitivity is 14,600 mm per volt. The fluctuations observed are the same as those obtained with zero collecting potential showing that the batteries are adequately quiet in operation. Fig. 8 is taken at the same sensitivity as Fig. 7 but with the grounding key closed. It is seen that the very small fluctuations of Fig. 8 are insignificant compared to those at floating potentials.

When the grid leak is put in the circuit the long period fluctuations are suppressed but the short ones are about the same in magnitude. This is illustrated in Figures 3 and 6, and in more detail in the natural size record of Fig. 9 which was recorded with the ionization chamber at 2 mm Hg pressure and 315 volts collecting potential.

In the second type of ionization chamber, with which most of the quantitative measurements of alpha activities were made, the walls of the inner chamber were made of solid brass instead of a light net-work of wire. The outer housing was turned out of a solid steel rod, 6 inches in diameter, cut to give a small clearance on all sides between the inner and outer chambers and allowing at least $1\frac{1}{2}$ inches thick of iron all around to act as an absorption shield for the gamma rays emitted from the radioactive contaminations of the walls of the laboratory.

This thickness of iron reduces the intensity of background gamma radiation to a value less than that of the cosmic radiation so that the resultant ionization fluctuation is due chiefly to cosmic radiation and cannot therefore be further reduced without the use of awkwardly large thicknesses of shield.

The ionization space proper is in the shape of a cylinder 1.84 cm in depth and 5.08 cm in diameter, the upper lid of which is removable . The collecting electrode is a steel drill rod 1 mm in diameter, extends to within about 2 mm of the top cover, and leads to the FP-54 below, through an amber insulator. The top cover of the outer housing gives a gas-tight fit by means of a tongue-and-groove joint with a rubber gasket and is bolted down with 6 quarter-inch cap screws.

Fig. 10 gives a close-up view of the ionization chamber with outer and inner covers removed. Fig. 11 is a picture of the complete apparatus with the ionization chamber supported by the FP-54 housing occupying the right central portion of the picture. At the rear is seen the galvanometer shield and to the left we see successively the control box (containing rheostat and resistances), thermally insulated storage battery box and CO_2 cylinder. To the extreme right is a box containing the Ayrton shunt, II

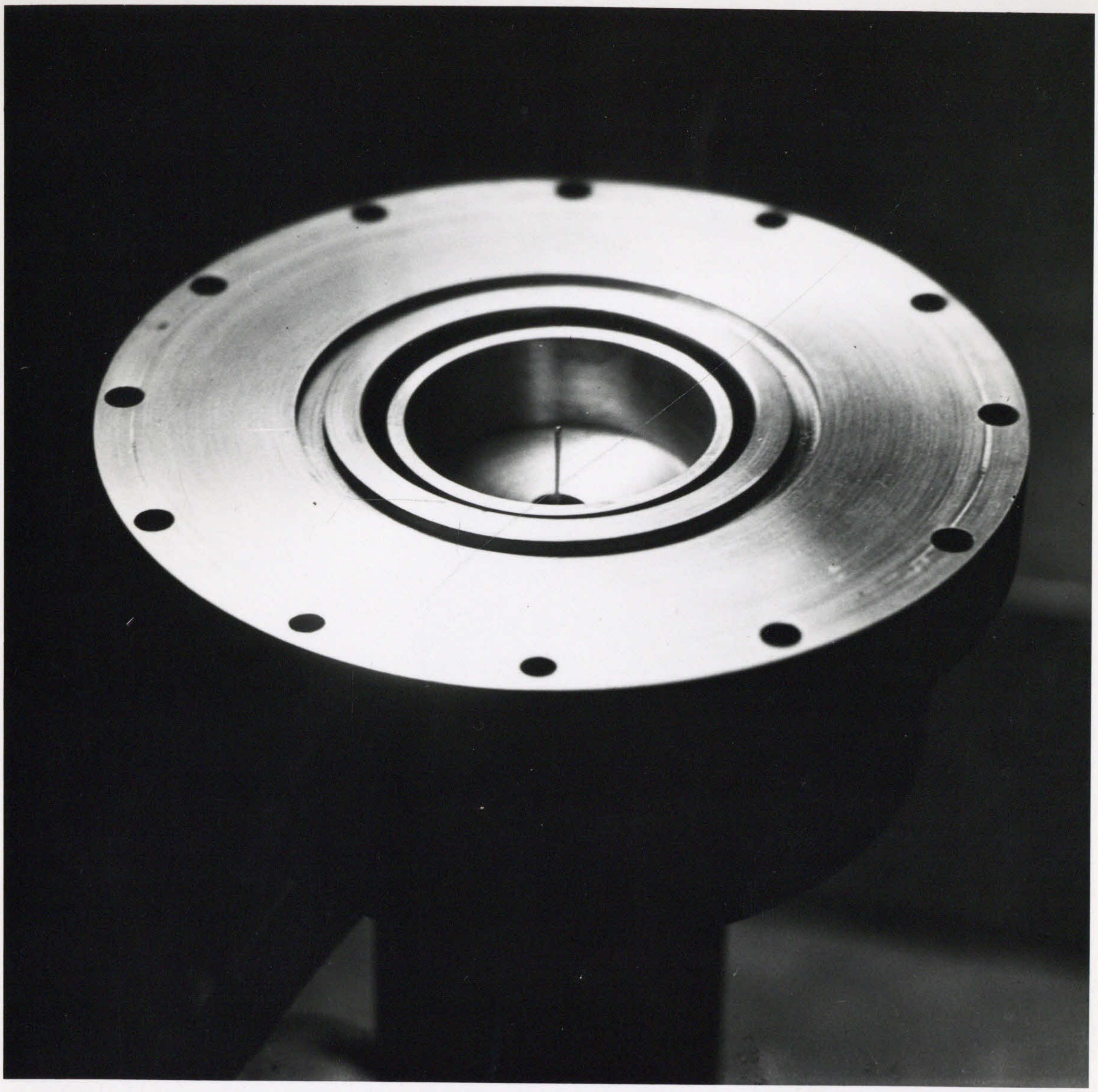

FIG. 10. IONIZATION CHAMBER CLOSE-UP

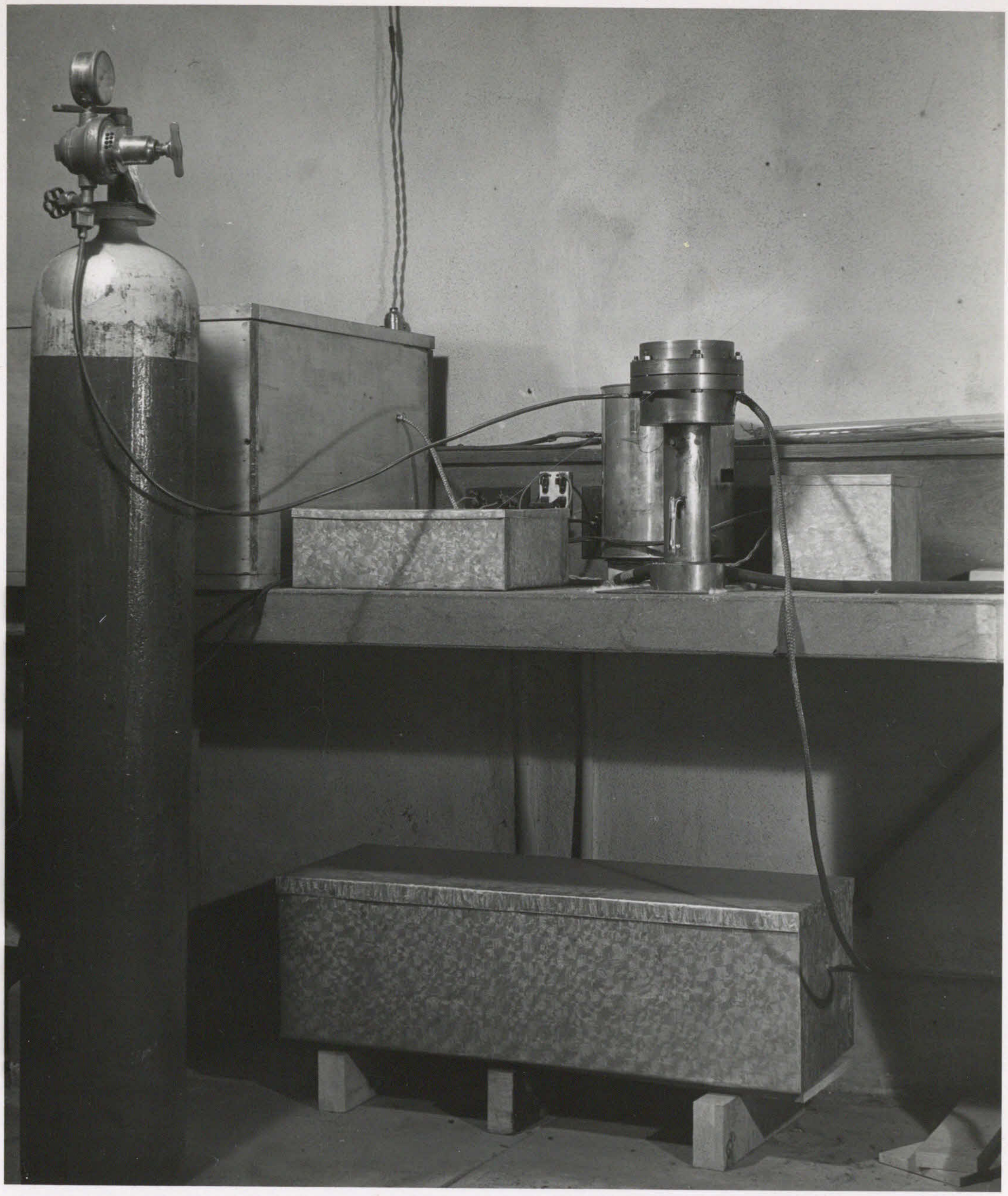

FIG. 11. APPARATUS; COMPLETE ASSEMBLY

and the long box below the bench conceals the block of "B" batteries. It is to be observed that all parts of the circuit are completely shielded. The shielding boxes are constructed of galvanized iron, brass or copper, and all connecting wires are encased in Roebling copper-braided shielding which is in all cases securely soldered to the shielding terminating the ends of the braids. Fig. 12 gives a closer view, in which the ionization chamber unit is shown in more detail.

While the major work of this investigation was done with the second ionization chamber, we shall first discuss the use of both and then give a more detailed descriptions of the operations with the second.

Measurement of Charge Sensitivity.

The effective charge sensitivity in terms of the actual ionization produced in the ionization chamber was measured by placing a small drop of dilute uranyl nitrate solution on the brass plate which formed the top of the chamber. The sizes of the kicks recorded were then measured and the distribution curves of the kicks thus obtained revealed the two groups of alpha particles from UI and UII.

In Fig. 13 are plotted the results of such a distribution curve taken with the first chamber and with

FIG. 12. APPARATUS; CLOSER VIEW

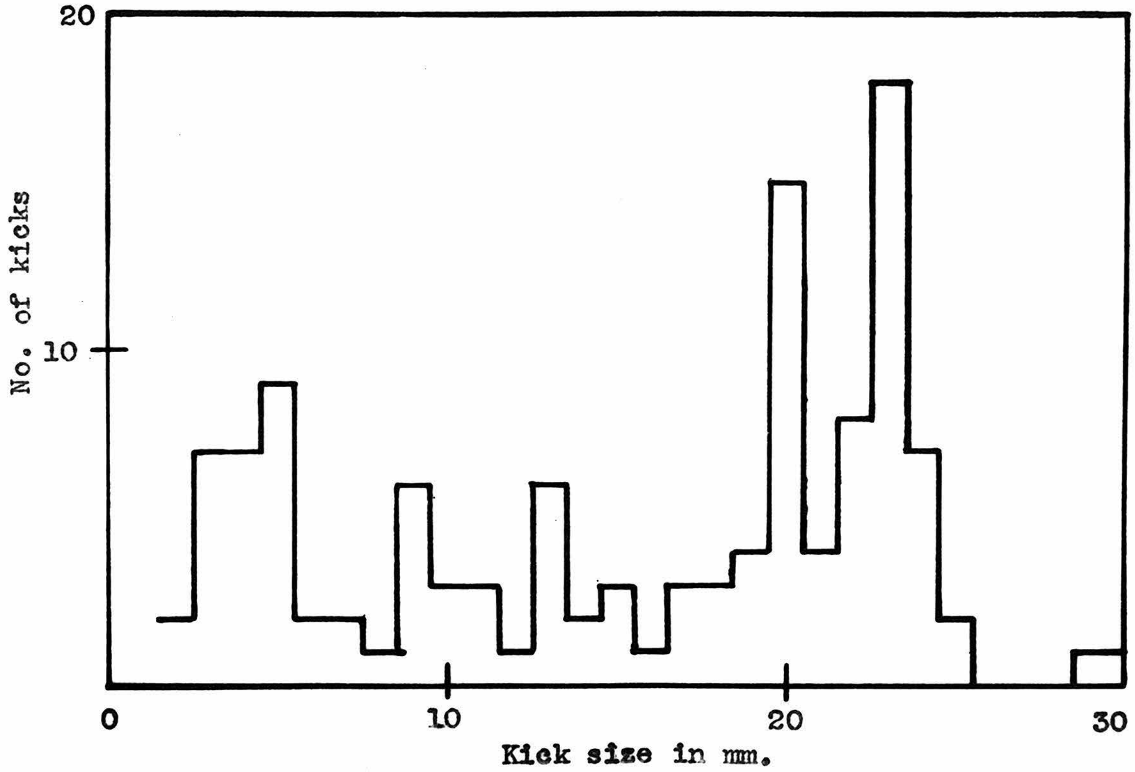

FIG.13. KICK DISTRIBUTION FROM URANIUM; 315 VOLTS.

grid floating at 315 volts collecting voltage and one atmosphere pressure. To illustrate the effect of collecting voltage on the collection of ions, we include Fig. 14 which is similar to Fig. 13 with the exception that the data were taken at 135 volts and one atmosphere. The kick sizes for the two groups are reduced, as is also their resolution.

Ziegert's[34] very careful determination of a total ion production of alpha particles from UI and UII gives the values 1.16×10^5 and 1.39×10^5 ions respectively. Taking the two groups of kicks at 20 mm and 23 mm of Fig. 13 to represent these ionization values we calculate the effective charge sensitivity in the actual recording of alpha particles at 1 atm. and 315 volts to be 5800 and 5600 ions per mm, respectively with a mean value of 5700 ions per mm. The voltage sensitivity was 12,7000 mm per volt. All subsequent measurements of alpha activity were made at the standard conditions of 1 atm. pressure of CO_2 and 315 volts collecting voltage.

This sensitivity is applicable only to the first chamber used with grid floating. In all runs made with the first chamber, this condition was satisfied.

Not only is the capacity of the second chamber different from the first, but it was never used with grid floating, only with grid resistance connected. Accordingly a

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[34] H.Ziegert, Zeit. f. Phys. 46: 668, (1927).

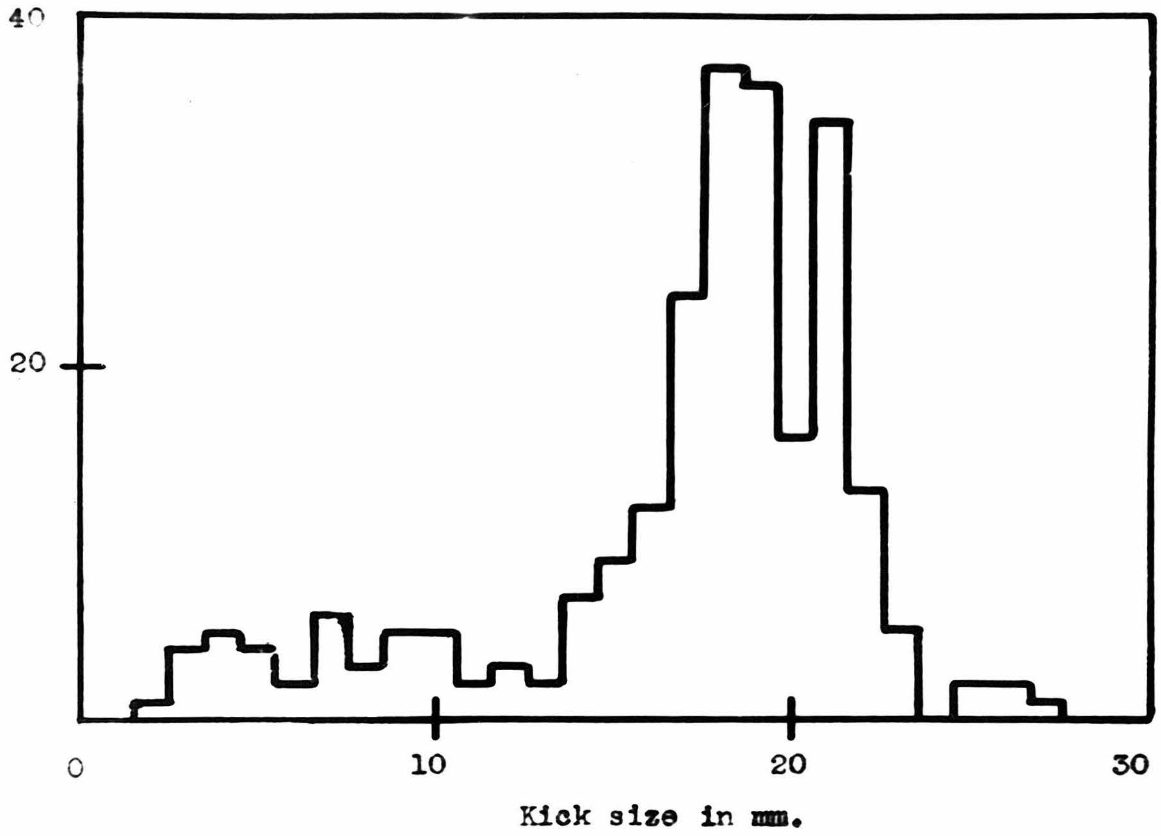

FIG.14. KICK DISTRIBUTION FROM URANIUM; 135 VOLTS

determination of the charge sensitivity of the second chamber was made with grid leak connected and a portion of the record obtained in this run is pictured in Fig. 18. Due to the reduced sensitivity, kicks were tabulated at 0.5mm intervals instead of 1 mm intervals as before. The two UI and UII groups gave kicks of 9.00 and 10.0 mm and we obtain an average charge sensitivity of 12,900 ion pairs per mm. The voltage sensitivity in this case was 14,600 mm per volt. This change in the voltage sensitivity was caused by a re-leveling of the galvanometer necessitated by shifting it to another position and is not due to a change in the tube characteristics. No change in the sensitivity could be detected which was not due to a disturbance of the galvanometer.

V METHOD OF MEASUREMENT

It is apparent that the above apparatus provides a simple, easy and reliable method of determining the rate at which alpha particles enter the ionization chamber. To do this one need merely set the galvanometer at the requisite point, load the camera, start the record, and the data will be recorded with no attention whatever from the operator for periods of 8 to 12 hours or more or any length of time needed to secure sufficient kicks to give accurate results. The recorded kicks may be easily and quickly counted.

Furthermore there is a permanent record of the measurement and infallible evidence of the behavior of the apparatus during the measurements.

Thick Sources and Thin Sources.

Measurement of alpha particle activities may be conducted on a source whose thickness exceeds the range of the alpha particles in the material composing it or it may be made on a source thinner than the range of alpha particles. For rocks, the former may be a smoothly polished surface of the rock or a deep layer of finely ground rock. The latter may be a thin layer of the rock very finely powdered and deposited on a backing of zero or very low alpha activity.

Disadvantages of Thick Sources.

If one can assume complete homogeneity of the rock, the alpha activity in alpha particles emitted per cc per unit time. can be easily calculated from the measured rate of emission of alpha particles by a smoothly polished face of the rock, provided that the stopping power of the rock for the alpha particles is known. If both the uranium and thorium series are present, the result will be a function of the Th/U ratio, since the mean ranges of the two series are different. It will not be a sensitive function, however, and for most purposes the error introduced by neglecting to make a separate determination of radium will not be important.

Nevertheless in spite of its apparent advantages this method was abandoned after tests on only two smoothly polished rock specimens. It is necessary to mention only a few of the chief reasons. In the first place, since the observed alpha particles come from many different depths within the rock they possess all ranges from zero to the maximum and the counting error is directly proportional to the accuracy with which we determine the limiting size of alpha particle to be counted. Also, it is obvious from inspection that the average rock can by no means be considered homogeneous for distances in the rock of the order of alpha particle ranges. Furthermore, the calculated result is directly proportional to the stopping power of the rock, the average value of which may be calculated with fair accuracy for the rock as a whole, but into which considerable uncertainty may be introduced if the radioactivity has a tendency to be concentrated into inclusions of a stopping power appreciably different from the average.

Probably the biggest objection is that a considerable fraction of the material to be studied can not be suitably ground in the form of a smoothly polished surface. Much of the cosmic ray material is of this type. At best, the polished surfaces are obtained only after long and arduous grinding. The chance of picking up contamination during this process are very great.

These objections give the chief reasons why this method was abandoned in favor of the use of a thin deposit of the rock which has been ground to a powder of a fineness such that the largest grains have a smaller maximum diameter than the range of the shortest alpha particle.

Kovarik and Adams[35] have demonstrated that uranium oxide may be ground in an agate mortar to a particle size sufficiently small that all alpha particles emitted in the forward direction from a deposit of the order of 1 mg per cm^2 may be quantitatively counted. This is a very straight test, since uranium emits the shortest of known natural radioactive alpha particles with the exception of samarium whose activity and concentration are so low as to have no significance in ordinary rocks. To test the efficacy of the agate mortar in reducing the particle size of a rock to a value smaller than the shortest alpha particle range, ground specimens were examined under a microscope equipped with a micrometer eye-piece and were observed to have a grain size considerably below the maximum allowable limit. Figure 15 shows photomicrographs of 3 typical views of a sample of granite ground in an agate mortar and dispersed on a slide by the method of Green[36]. For purposes of measurement there is also shown a small section of a micrometer scale ruled on glass at 100 lines per mm, photographed at

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[35] A.F. Kovarik and N.I. Adams, Jr., Phys. Rev. 40: ~~178~~ 718, (1932).

[36] H.Green, Journ. Frank. Inst. 192: 637, (1927).

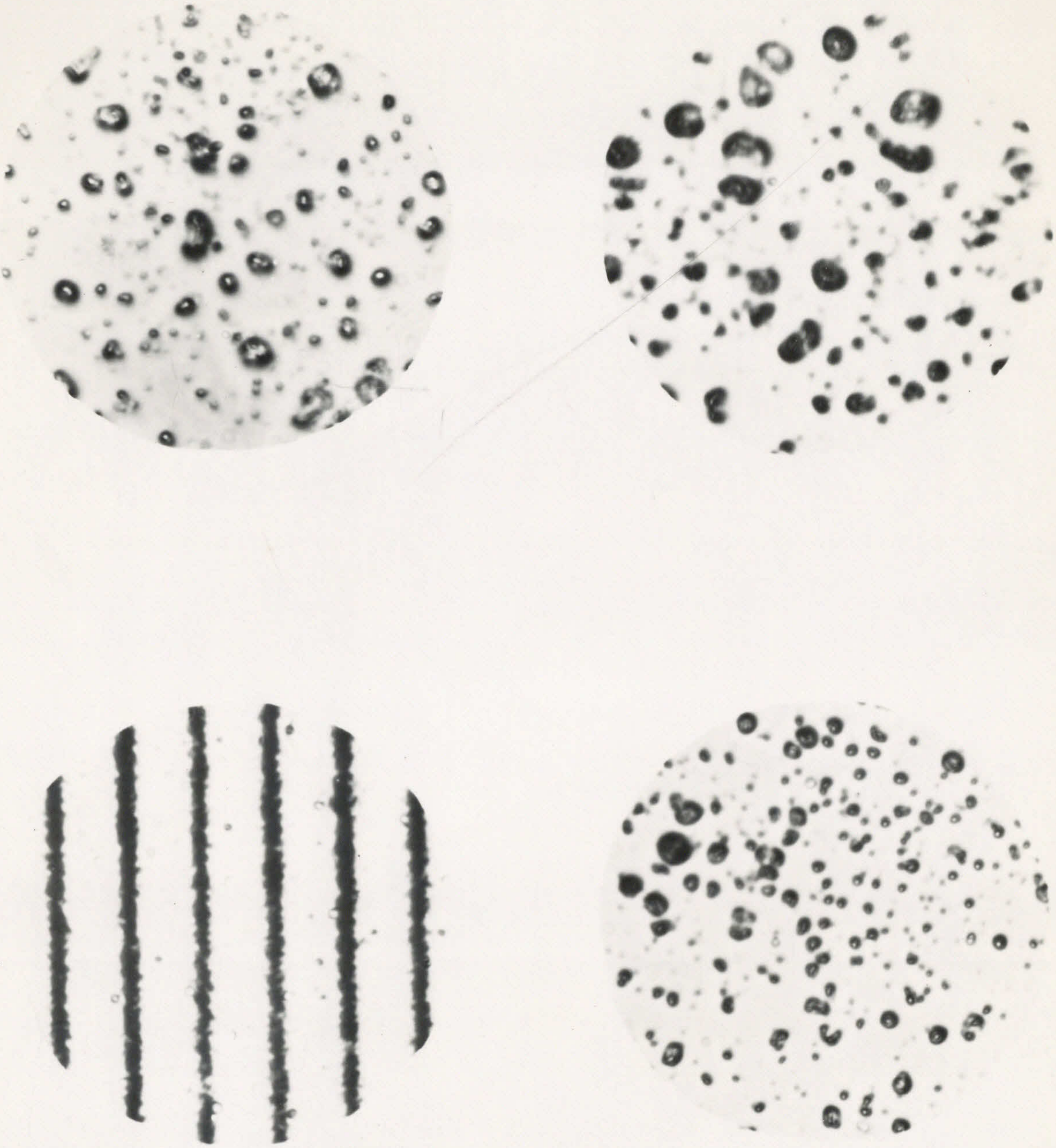

FIG. 15. PHOTOMICROGRAPHS OF POWDERED ROCK

the same setting of the photomicrograph camera. It is clear from these pictures that the largest grains are smaller than the range, 0.0126 mm, of the shortest alpha particle in biotite[37] a typical rock mineral,and the vast majority of the grains are a small fraction of this range.

With these results as a basis, the rest of the investigation was devoted to the determination of the alpha activities of a number of the cosmic ray rocks by the use of thin sources.

Experimental Procedure.

Into the brass lid of the second ionization chamber the one illustrated in Fig. 10, a recess was cut, 48.0 mm in diameter and 0.8 mm deep. After careful cleaning and weighing, this disc was placed in an accurately level position, a 1.5 cc suspension of the finely ground specimen in amyl acetate was poured into the depression in the lid and the material in the suspension settled slowly and uniformly over the surface. After the amyl acetate had evaporated, this process being accelerated by means of radiation from a polar cub heater, a thin, even coat of powder remained adhering strongly to the disc.

The appearance of a typical source thus prepared is illustrated in Fig. 16.

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[37] "Age of the Earth." Nat. Res. Council Bull. 80, (1931).

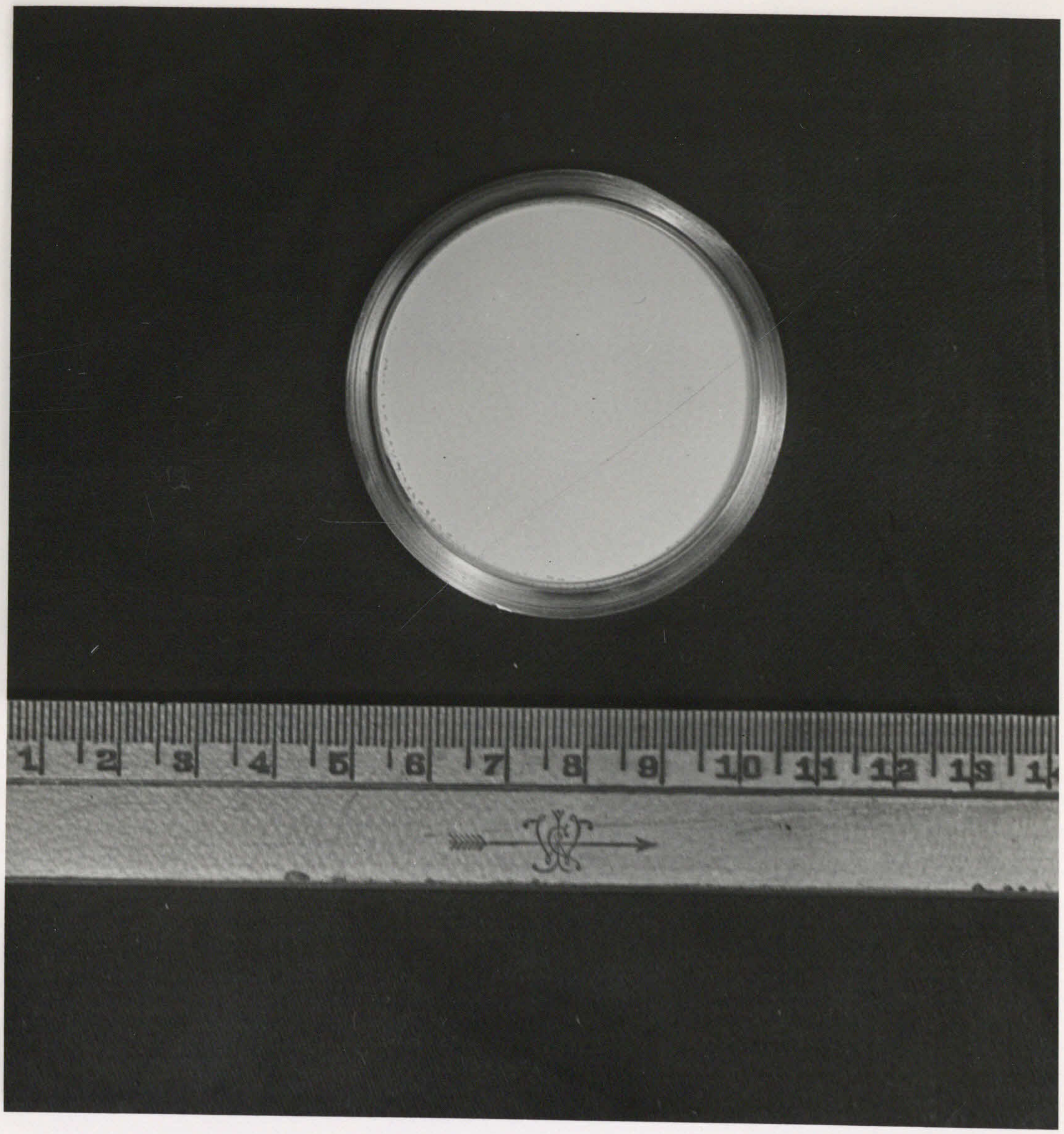

FIG. 16 SOURCE DISC WITH ROCK POWDER DEPOSITED

When the amyl acetate had thoroughly dried, the disc was carefully reweighed and the increase in weight gave the weight of the deposited sample. This weight was obtained accurately to 0.1 mg.

The grinding of the sample was done in an agate mortar and was continued until the rock had been reduced to as fine a size as could be accomplished. It was then thoroughly dispersed by grinding in a few drops of amyl acetate in the mortar, after which 1.5 cc of amyl acetate were added and, after agitating to insure complete suspension, the entire 1.5 cc was picked up in a medicine dropper and deposited on the disc.

After drying and weighing, the disc was replaced on top of the ionization chamber, to which it fitted closely, the chamber was thoroughly flushed with CO_2 and sealed. The potential of 315 volts was then applied, the amplifier was balanced and the galvanometer spot set correctly, the camera loaded and a record started. Some 10 or 12 hours later, during which time the apparatus needed no attention whatsoever, the writer returned, and developed the record. If considerable time elapsed before the return to the apparatus could be made, the galvanometer spot simply drifted off the paper and no harm was done.

All of the runs made with the apparatus just described were made with the grid resistance in the circuit. Typical records are illustrated in their entirety at 1/3 natural size in Figs. 3 and 6 while Fig. 17 illustrates a portion of a run on a typical rock, photographed at natural size. Fig. 18 of the same plate gives, for purposes of comparison a natural size portion of a record obtained from a very small uranium infection on the ionization chamber lid.

After the record had dried, the alpha particle kicks were counted with a hand counter and the results tabulated in terms of alpha particles per hour. A kick size of 1.5 mm was chosen as the limit above which all kicks were counted and below which none were counted. By making runs at low pressure, it was demonstrated that no spurious kicks greater than 1.5 mm were produced by instrumental disturbances in the amplifier. This size kick represents an alpha particle of 20,000 ions.

At frequent intervals, between rock runs, background determinations are made in which the preparation of the source is in every way identical to the case of a regular run, with the single exception that the 1.5 cc of amyl acetate evaporated on the disc contains no suspension. Besides giving the true background this method reveals the extent to which contamination may be picked up in the process of deposition.

The subtraction of the background rate from the rate recorded with the rock sample gives the rate due to the rock alone.

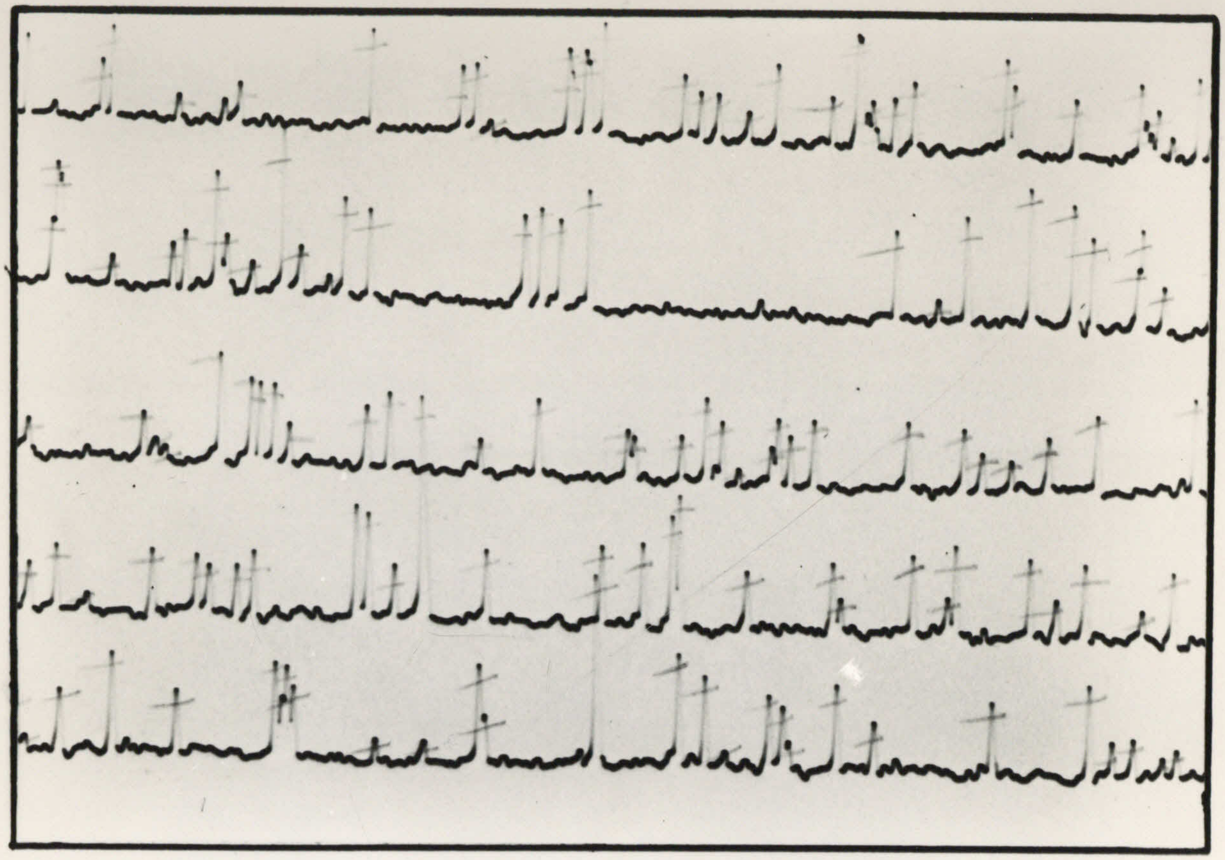

FIG. 17 ALPHA PARTICLES FROM ROCK

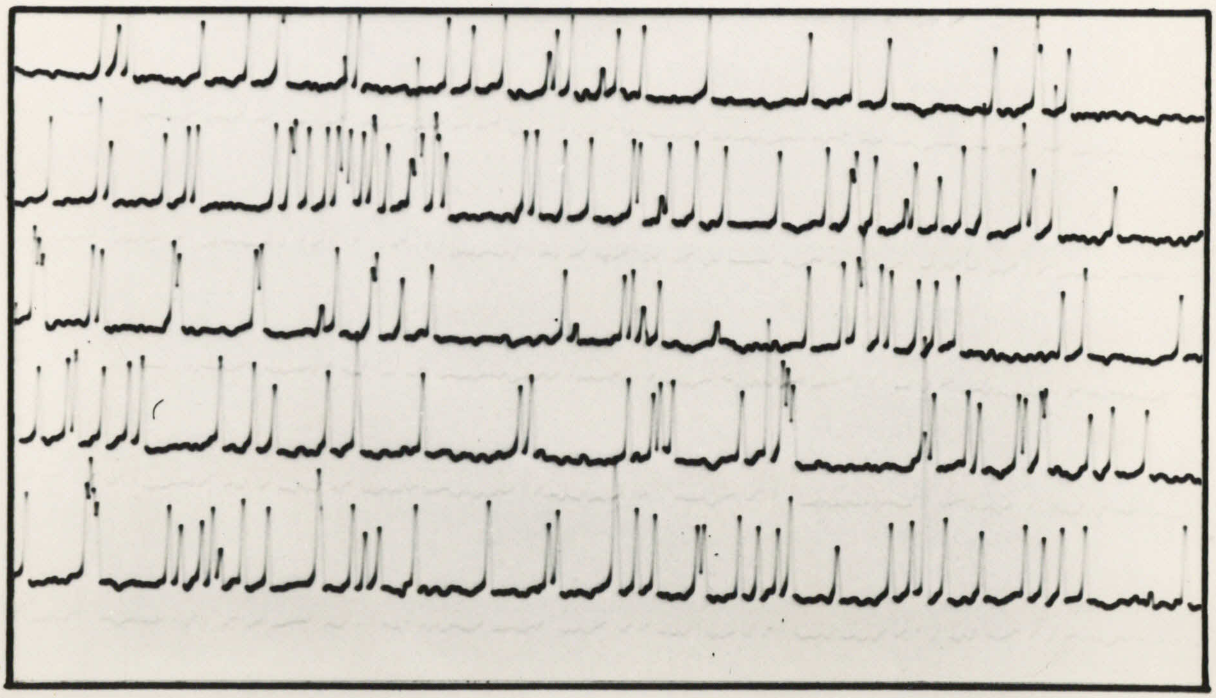

FIG. 18. ALPHA PARTICLES FROM URANIUM

Counting Errors.

The emission of alpha particles is a random process and hence the mean deviation[38] of an observed number, n, of alpha particles is equal to $\sqrt{n}$. The probable error[39] of the observation will be $0.67\sqrt{n}$, in which it is assumed that the deviations of Poisson's law[38] from a Gaussian error curve, in the case of the numbers of particles counted in the average runs, is unimportant. With each observation of counting rate, the probable error calculated in this manner is also tabulated.

Corrections.

As the ratio of the thickness of the source to the ranges of alpha particles in the rock is made to approach zero, the ratio of the alpha particles projected into the ionization chamber to the weight of the sample will accurately represent the rate of alpha particle emission per unit weight. In practise, this ideal situation cannot be attained and some corrections must be applied to the measured rate to obtain the true activity.

(1) Chance coincidences.

Due to the random nature of alpha particle emission there is a finite probability that two or more alpha particles will enter the ionization chamber in an interval

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[38] H. Bateman, Phil. Mag. 20: 704, (1910).
[39] R.D. Evans and H.V. Neher, Phys. Rev. 45: 144, (1934).

smaller than the galvanometer period even though the average interval between particles is much greater than this. In order to make the correction for this effect quantitatively calculable, those particles were counted which entered the chamber after the galvanometer swing caused by the preceding particle had reached maximum amplitude. The time taken to reach this amplitude, as measured with a stop-watch on a number of alpha particle kicks, is 5.7 seconds. If x is the average rate of appearance of particles in this interval then the probability[38], Pn, of n particles appearing, is

$$P_n = \frac{x^n e^{-x}}{n!} \tag{1}$$

Since any group occurring in this interval will be observed as one particle, the observed rate, y, will be

$$y = \sum_{n=1}^{\infty} \frac{x^n e^{-x}}{n!} = e^{-x}(e^x - 1) = 1 - e^{-x}$$

Solving for the true rate, x we get

$$x = -\log(1-y) = y + \frac{y^2}{2} + \frac{y^3}{3} + \cdots \tag{2}$$

Let Z' be the measured rate of emission of alpha particles; then $y = .00158Z'$. Since Z' rarely exceeds a value of 100 particles per hour we may neglect terms of higher order than y^2 and we see that the error of chance coincidences is eliminated by multiplying the measured rate Z' by the factor

$$(1+0.00158Z'/2) \qquad (3)$$

(2) Short lived alpha emitters.

The alpha particle emitted by an element whose mean life is small compared to the time of the galvanometer swing will be counted only half of the time if its immediate parent is an alpha emitter. The actinium series possesses two of such substances and the thorium series possesses one. Hence we may calculate the real rate of emission of the actinium and thorium series from the measured ones by multiplying the observed rates with the factors 7/6 and 12/11, respectively. Rigorously, this correction necessitates the a knowledge of the Th/U ratio, but for many purposes the error introduced by assuming equal activities of the thorium and uranium series will not be important. In this case, the correction factor is 28/27 or 1.04.

(3) Absorption in the source.

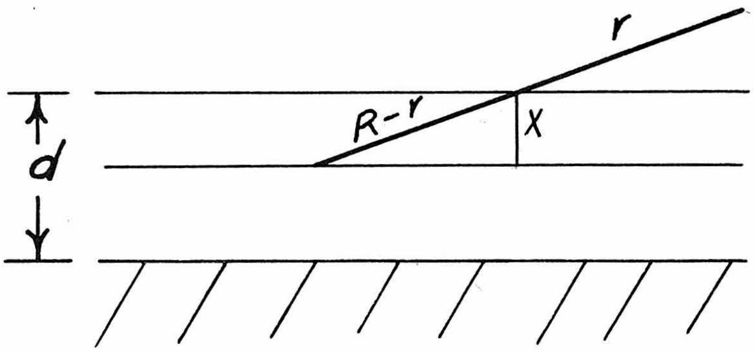

Fig. 19

Consider the sample to be uniformly spread over the source in a layer of a depth whose air equivalent for the stopping of alpha particles is d. Assuming unit intensity of emission of alpha particles of range R per unit volume, the total number of particles of range greater than r, emitted into the ionization chamber from an element of unit area and depth dx is seen from the figure to be equal to

$$\frac{1}{2}\left(1-\frac{x}{R-r}\right)dx$$

The number emitted with ranges between r and r + dr is the differential of this quantity,

$$\frac{1}{2}\,\frac{x\,dx\,dr}{(R-r)^2}$$

The distribution function f(r)dr which gives the rate of emission per cm^2 particles of range between r and r dr for the whole layer is obtained by integrating the above expression over x, noting that x cannot exceed R-r. The function has two parts:

1. $0 < r < R - d$

$$f(r)dr = \frac{dr}{2}\int_0^d \frac{x\,dx}{(R-r)^2} = \frac{d^2}{4(R-r)^2}\,dr \tag{4}$$

2. $R - d < r < R$

$$f(r)dr = \frac{dr}{2}\int_0^{R-r} \frac{x\,dx}{(R-r)^2} = \frac{dr}{4} \tag{5}$$

If we know the limiting range, S, which corresponds to an alpha particle of 20,000 ions, chosen as the lower limit of ionization pulse to be counted as an alpha particle, we can integrate over the distribution function of path lengths between the limits of S and R and obtain the actual number of alpha particles emitted into the chamber. Before using this result to calculate the total activity of the rock from the observed activity we must take into account another fact, the limitation of range by the geometry of the chamber.

(4) Geometrical Limitation of Ranges.

Some of the alpha particles, emitted from materials deposited on the lid closer to the walls than the critical range, S, will lose most of their range in the wall and the ionization pulse produced will not be counted.

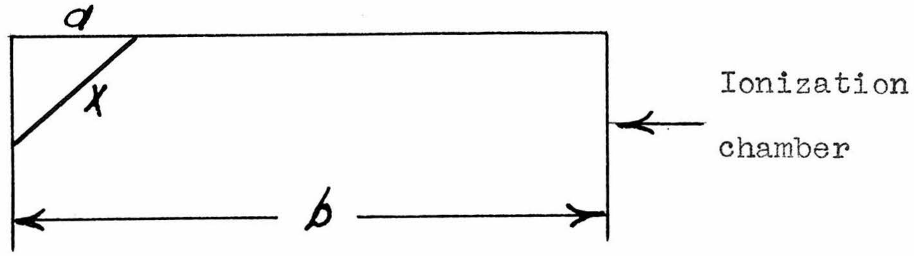

Fig. 20

The geometrical situation is illustrated in Fig. 20. An alpha particle situated a distance, a, from the cylindrical wall of the ionization chamber whose diameter is b, possesses the shortened range, x, due to impact with the wall. As the critical range, S, is small compared to b and we are interested in determining the number of alpha particles with path lengths in the chamber less than S, we may assume with sufficient accuracy for the purpose of this calculation, that the angle between the top and wall of the chamber can be considered a dihedral angle.

Consider that the top of the chamber is covered uniformly with an alpha emitter of homogeneous range, r, with unit activity per unit solid angle and cm^2. The total number of particles of limited range less than x emitted from a strip of width da is $\pi^2 bda(1 - a/x)$, in which x must be greater than a. The activity of particles whose range is between x and x + dx is the differential of this quantity

$$\pi^2 \frac{b\,a\,da\,dx}{x^2}$$

The total activity of those particles whose range is limited by the wall is the integral

$$\int_0^r \int_0^x \frac{\pi^2 b\,a\,da\,dx}{x^2} = \frac{\pi^2 b r}{2}$$

The total number emitted is $\pi^2 b^2/2$ and the number whose range is unlimited is hence

$$\frac{\pi^2 b^2}{2} - \frac{\pi^2 b r}{2} = \frac{\pi^2 b^2}{2}\left(1-\frac{r}{b}\right)$$

The distribution function f(x)dx of those whose range is limited is

$$\frac{\pi^2 b\,dx}{x^2}\int_0^x a\,da = \frac{\pi^2 b}{2}\,dx = \frac{\pi^2 b^2}{2}\,\frac{dx}{b}$$

From these results it is seen that if the distribution-in-range of the particles emitted from the top of the chamber is given by f(r)dr, the resultant distribution produced in the ionization chamber (for values of r small compared to b) will be

$$\left(1-\frac{r}{b}\right)f(r)dr + \frac{dr}{b}\int_r^R f(r)dr \tag{6}$$

where f(r)dr is given by equations (4) and (5). The integral of (6) becomes

$$\int_r^R f(r)dr = \frac{d^2}{4}\int_r^{R-d}\frac{dr}{(R-r)^2} + \frac{1}{4}\int_{R-d}^R dr$$

$$= \frac{d}{2}\left(1-\frac{d}{2(R-r)}\right) \tag{7}$$

Hence for $0<r<R-d$ we obtain the distribution function

$$\frac{d^2}{4}\left(1-\frac{r}{b}\right)\frac{dr}{(R-r)^2} + \frac{d}{2b}\left(1-\frac{d}{2(R-r)}\right)dr \tag{8}$$

We are interested in the number of track lengths, r, less than the critical length, S. To obtain this we integrate the function (8) from 0 to S.

$$\frac{d^2}{4}\int_0^S \left\{ \frac{1}{(R-r)^2} - \frac{r}{b(R-r)^2} + \frac{2}{db} - \frac{1}{b(R-r)} \right\} dr$$

$$= \frac{d^2}{4}\left[\left(1-\frac{R}{b}\right)\frac{1}{R-r} + \frac{2r}{db} \right]_0^S$$

$$= \frac{d^2}{4}\left\{ \frac{1}{R-S} - \frac{1}{R} + \frac{2S}{db}\left[1-\frac{d}{2(R-S)}\right] \right\}$$

As the material deposited on the ionization (9) chamber lid was assumed to have unit activity per unit volume, and the distribution function of (4) and (5) was expressed as rate of emission per cm^2, the true rate of emission per cm^2 of alpha particles having a downward velocity component, is d/2. The number of those absorbed in the source is obtained by letting r = 0 in equation (4) and subtracting this expression from d/2. The result is d^2/ 4R. Adding this quantity to equation (9) we obtain

$$\frac{d^2}{4}\left\{ \frac{1}{R-S} + \frac{2S}{db}\left[1-\frac{d}{2(R-S)}\right] \right\} \qquad (10)$$

which is the quantity by which the measured emission differs from the true one.

The measured emission is

$$\frac{d}{2}\left\{1-\frac{d}{2(R-S)}-\frac{S}{b}\left[1-\frac{d}{2(R-S)}\right]\right\} \tag{11}$$

In order to make corrections small and therefore give a more accurate result, d was always made small in comparison to the average alpha particle range. Hence d/2(R-S) can be neglected in the bracket of the last term of (11). We therefore see, that in order to correct for absorption in the source and geometrical limitation of range we must divide the measured result by the factor:-

$$\left(1-\frac{d}{2(R-S)}-\frac{S}{b}\right) \tag{12}$$

The value of S is difficult to estimate. Geiger's[40] rule relating ion production with range falls down completely for small ranges. Evans[41] has estimated the range-ionization relation for short particles by graphical integration of the empirical ionization curves of alpha particles. From his result, the range of a 20,000 ion alpha particle is 5 mm in air or about 3 mm in CO_2.

For the purposes of this investigation it seemed advisable to make a direct determination of the value of S/b both to induce more confidence in the calculation of the correction and as a check on the above figure.

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40. H. Geiger, Proc. Roy. Soc. <u>83A</u>: 505, (1910).
41. R.D. Evans, Phys. Rev. <u>45</u>: 29, (1934).

Accordingly a distribution curve of kick size at intervals of 1 mm was made for alpha particles from a source formed of 10.5 mg of powdered rock C7, a granite. The source area was 18.1 cm^2 and the density of deposit was 0.580 mg per cm^2.

This distribution curve is plotted in Fig. 21. It is the result of 48.7 hours of observation and 1482 measured kick lengths. The distribution curve of 645 background kicks taken over a period of 102.1 hours is illustrated in Fig. 22 and the difference of the two, the distribution due to alpha particles from the rock alone is plotted in Fig. 23.

If this curve is extrapolated to zero kick size, the area under the curve included between the axis and the ordinate at 1.5 mm represents the activity of particles of range less than the critical range, S. The ratio of this number to the total activity observed gives the value of S/b.

The observed activities were 1.77 alpha particles per hour for particles of kick length less than 1.5 mm and 24.1 alpha particles per hour for the total activity. The resultant value of S/b and estimated error is (0.07 ± .01). Since b is 50.8 mm, the value of S obtained from this result is 3.5 mm. We therefore assume that the limiting kick size, 20,000 ions represents an alpha particle with a range

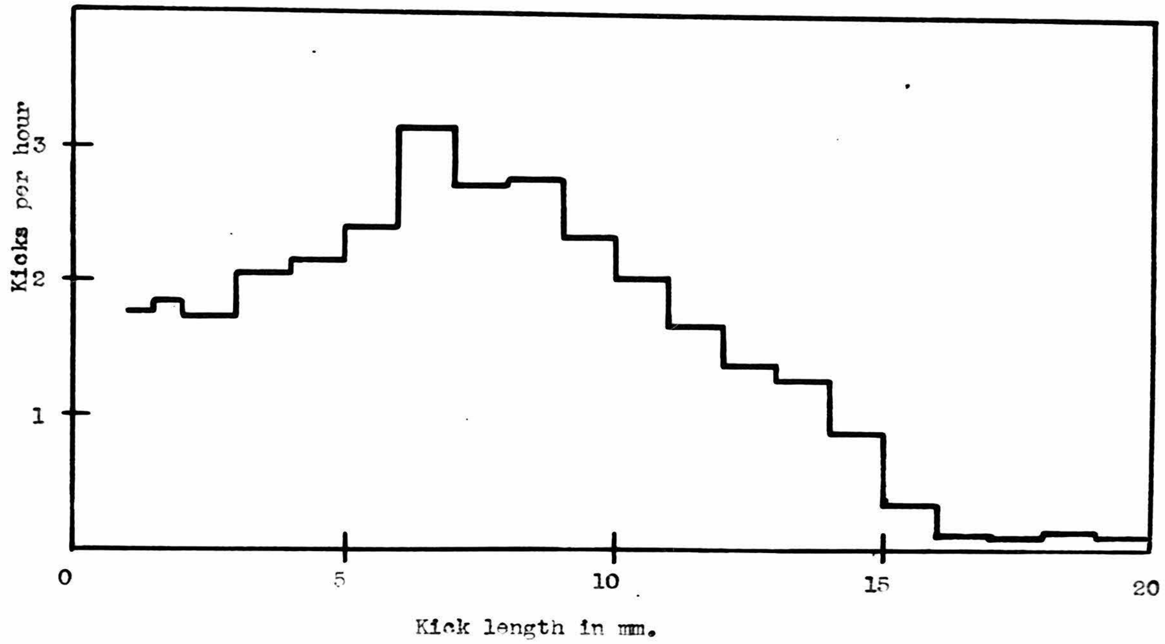

FIG.21. KICK DISTRIBUTION FROM ROCK AND BACKGROUND

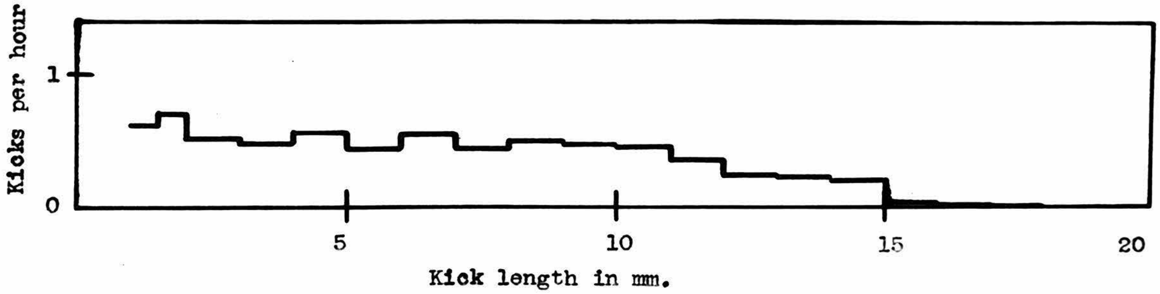

FIG.22. KICK DISTRIBUTION FROM BACKGROUND ALONE.

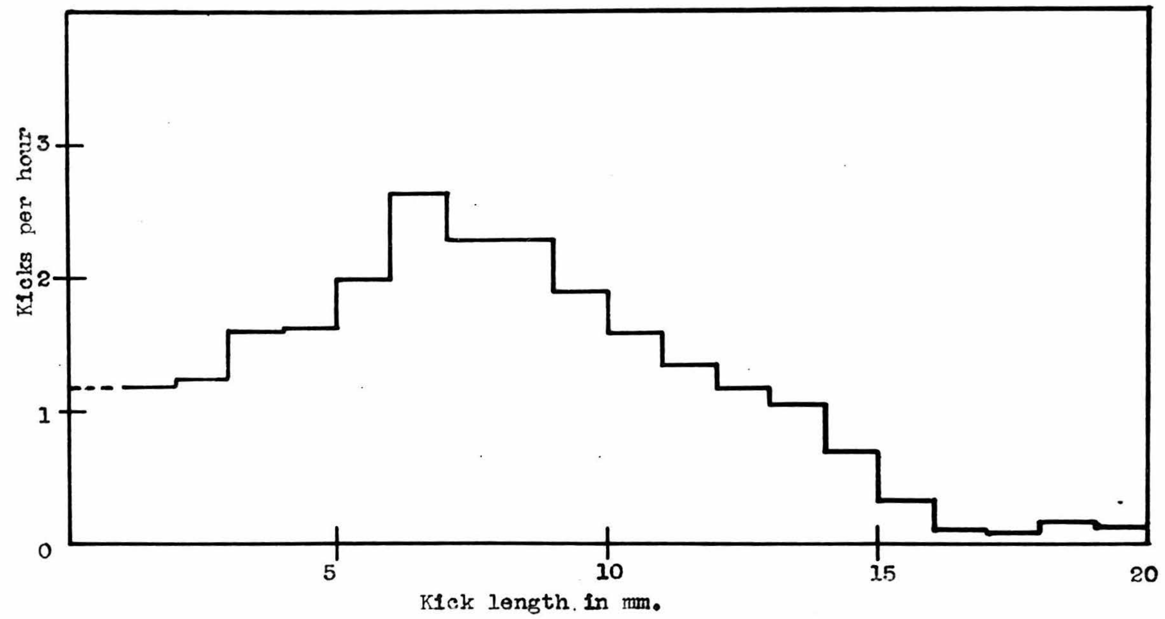

FIG.23. KICK DISTRIBUTION FROM ROCK ALONE

of 5 mm in air and the term $d/2(R-S)$ of the correction factor is calculated on this basis.

Stopping Power of Rocks.

As d and (R-S) must be expressed in equivalent stopping powers, air at 0° C and 760 mm Hg was chosen as the standard substances. The mean value of $1/R-S$ for the alpha particles of the uranium series[42] in air at these standard conditions, is 0.330, for the thorium series, 0.268, and for the actinium series 0.264. The mean value of all series, chosen for the calculation of the correction factor is 0.30.

The equivalent thickness, d, is calculable from the surface density of the rock source, by the use of the Bragg and Kleeman rule[43] that the atomic stopping power is proportional to the square root of the atomic weight.

The factors, F, by which the surface density of the source layer in mg per cm^2 must be multiplied to obtain its equivalent thickness in cms of air at 0° C. and 760 mm Hg, were calculated for a number of chemical substances of common occurrence and are tabulated in Table III.

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[42] Rutherford, Chadwick and Ellis, "Radiations from Radioactive Substances", p. 86.

[43] loc. cit. p. 99.

Table III. Factors for conversion of mg per cm^2 to cm air, for various substances.

Substance	F
$CaCO_3$	.645
SiO_2	.656
Al_2O_3	.647
Fe_2O_3	.499
FeO	.473
MgO	.654
CaO	.539
Na_2O	.648
K_2O	.518
H_2O	.990
TiO_2	.551
$KAlSi_3O_8$ (orthoclase	.630
$Al_2Mg_2KHSi_3O_8$ (biotite)	.633
$FeMgSi_2O_3$ (pyroxene)	.606

It is seen that the most common rock constituents do not vary widely in the value of F.

Using the values of Table III, the factors for a number of igneous rock types ranging from high to low activity were calculated from the rock compositions as given by Clarke[44]. They are tabulated in Table IV along with the conversion factor of a composite sample of a large number of igneous rocks[45].

Table IV. Conversion factors for igneous rocks.

Rock	F
Granite	.643
Diorite	.654
Peridotite	.668
Hornblende picrite	.622
Gneiss	.643
Igneous composite	.638

The average is 0.64 and the extreme variations of different rock types introduces negligible error into the corrected rock activity. Hence, if ρ designates the surface density of the source in mg per cm^2, we may write, using the above values for 1/(R-S) and F:-

$$\underline{d/2(R-S) = 0.096\rho} \qquad (13)$$

The source used had an area of 18.1 cm^2.

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[44] F. W. Clarke, Data of Geochemistry. U.S.G.S. Bull. 770, (1924).

[45] G. v. Hevesy, Chemical Analysis by X-Rays, p. 258.

If m designates the total weight in mg of rock deposited on the chamber lid, then

$$\underline{d/2(R-S) = 0.0053m} \qquad (14)$$

The application of this result is not correct if the equivalent thickness, d, exceeds the value of (R-S) for the shortest alpha particle, 2.1 cm for that of UI. The upper limit for surface density set by this range is 2.1/.64 or 3.28 mg per cm^2, a value which was never exceeded in practise.

Sources of Error.

(1) Emanating power.

For hard and firm rocks of large grain size, the loss of emanations by diffusion through the surface is negligible[14]. However, for rock thicknesses less than alpha particle ranges, the effect must be considered. The short half-life of thoron and actinon inhibit their diffusion from all but the most porous material and we need only consider the effect of radon for the present problem.

The first ionization chamber allowed this radon to escape into the large volume of the outer chamber. In the second one this outer volume was considerably reduced so that any radon emitted from the source would be distributed between the ionization space proper and the outer chamber in the ratio of their volumes. The result is not only to reduce the effect of emanating power but to make possible its detection by a change in the recorded activity.

If one notes that the radon in the gas emits alpha particles in all directions instead of half the total solid angle, then the emanating power effect can be completely cancelled by making the ionization space equal to 3/4 of the total free volume. In the actual chamber used, this volume was 48% of the total. For the extreme case of a source formed completely of the uranium series and having 100% emanating power, the result on the recorded equilibrium count will be (1.5 - 2 x 0.48)/4 or 13.5% less than the true effect for zero emanating power. For the more common case of equal activities of the uranium and thorium series, the effect is 7%.

For the rock samples treated by the writer, special attempts to detect this effect were made by continuing the records of single samples in the sealed ionization chamber over periods of one week or more. No change in the counting rate greater than the probable error of counting could be detected indicating that it is safe to neglect this effect for most cases.

(2) Contamination.

The counting rates involved in this work are so small that the introduction of extremely minute quantities of radioactive impurities can make the results erratic and meaningless. However, by continuous care and cleanliness this effect can be avoided or at least reduced to a point where it is not serious.

Table V gives the results of all background determinations on the disc on which most of the rocks were deposited for measurement.

Table V. Background counting rates.

Time of run in hours	Alpha particles per hour.
19	6.2 ± 0.4
35	4.9 ± 0.3
38	6.2 ± 0.3
19	5.9 ± 0.4
19	7.3 ± 0.4
15	7.0 ± 0.5
23	5.6 ± 0.3
12	6.4 ± 0.5
20	6.2 ± 0.4
38	5.8 ± 0.3
41	7.1 ± 0.3
22	6.9 ± 0.4
37	5.6 ± 0.3
57	6.2 ± 0.2

The data are presented in chronological order and were taken over a period encompassing 8 months. Each reading represents a separate background reading, prior to which the disc was used for a rock determination. The designated probable errors of each run were calculated from the total number of counts in each case. The average deviation of these readings from their mean value is 0.49 alpha particles per hour and the probable error of each reading is 0.85 x $0.4\overset{9}{4}$ or 0.42. The average value of the probable errors of individual counts is 0.36.

The small difference of these two values makes it impossible to prove from them that contamination produces an appreciable effect. It seems safe to conclude that the probable error induced by fluctuation in the background contamination is less than 0.3 alpha particles per hour, a quantity which therefore sets the lower limit for detection of activity. This value is the observed activity of uranium and decomposition products in equilibrium with 6 x 10^{-16} gms of radium.

VI EXPERIMENTAL RESULTS

The alpha activities of a number of the cosmic ray rocks were determined by the above method and the results are tabulated in Tables VI and VII.

The first column of Table VI lists the rock designation, the second the weight of sample in mg,

Table VI. Alpha activities of cosmic ray rocks.

Rock number	Sample weight in mg	Counting rate	Activity in alpha particles per mg-hour	Mean
C5	24.5	13.2 ± 1.0	1.22 ± 0.09	
	35.5	16.7 ± 0.9	1.14 ± 0.06	
	16.3	9.1 ± 0.7	1.23 ± 0.09	1.20
C6	29.4	9.5 ± 0.6	0.84 ± 0.05	
	29.3	11.7 ± 0.6	1.04 ± 0.05	0.94
C7	10.7	27.8 ± 0.6	5.96 ± 0.13	
	23.3	47.1 ± 0.7	4.94 ± 0.07	
	21.9	47.6 ± 0.9	5.36 ± 0.10	5.42
C8	14.0	1.8 ± 0.7	.30 ± 0.12	
	30.4	5.9 ± 0.4	.50 ± 0.03	
	16.7	3.9 ± 0.5	.54 ± 0.07	0.46
C9	27.9	30.6 ± 0.8	2.81 ± 0.07	
	46.6	44.9 ± 1.1	2.83 ± 0.07	
	17.9	23.2 ± 0.5	3.15 ± 0.07	
	23.8	31.9 ± 1.2	3.08 ± 0.12	2.97
C10	25.8	8.7 ± 0.5	0.85 ± 0.05	
	28.8	8.2 ± 0.5	0.73 ± 0.04	0.79
C11	26.2	8.4 ± 0.5	0.81 ± 0.05	
	26.8	9.3 ± 0.5	0.88 ± 0.05	0.85
C12	25.2	7.8 ± 0.5	0.78 ± 0.05	
	29.6	8.6 ± 0.5	0.75 ± 0.04	0.76
C13	12.7	21.8 ± 0.5	3.99 ± 0.09	
	28.1	45.0 ± 0.9	4.15 ± 0.08	4.07
C16	20.4	1.2 ± 0.5	0.13 ± 0.05	0.13
C17	36.5	2.9 ± 0.4	0.21 ± 0.03	0.21
C18	28.6	12.3 ± 0.6	1.10 ± 0.05	
	27.8	12.9 ± 0.6	1.19 ± 0.06	1.14
C19	20.5	42.0 ± 1.0	5.16 ± 0.12	
	9.5	23.4 ± 0.9	5.60 ± 0.21	5.38
C20	18.7	19.6 ± 0.7	2.38 ± 0.09	
	32.7	33.6 ± 0.6	2.71 ± 0.05	2.55

the third observed counting rate, and the fourth the calculated activity in alpha particles per mg-hour. The fifth column gives the mean values of check runs upon samples of the same rock. The listed probable errors are calculated from the total number of counts observed in each run and include the probable error of the background, 0.3 counts per hour.

Columns two, three and four of Table VII list the alpha activities in alpha particles per mg-hour of the uranium series, the thorium series and the total activity, respectively. The uranium series activities include the actinium series and are calculated from the radium determinations by the use of the accepted[46] Ra/U ratio of 3.40×10^{-7}, by the experimental determination of Kovarik and Adams[35], that one gm of uranium emits $\underline{24.8} \times 10^{3}$ alpha particles per second, and by the actinium-uranium activity ratio of 0.04 determined by v. Grosse[8].

Using these three data we calculate the alpha activity in alpha particles per mg-hour of uranium and equilibrium products by multiplying the radium concentration in units of 10^{-12} gm per gm of rock with the factor 1.06. OK

Activities of the thorium series are obtained by subtracting the figures of column two, Table VII, from those in column five, Table VI and by multiplying the dif-

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[46] Lind and Roberts, Jour. Am. Chem. Soc. <u>42</u>: 1170, (1920).

Table VII. Results of radioactivity measurements

Rock	Uranium activity	Thorium activity	Total	Local radiation	Th/U activity	Th/U concentration
C5	0.55	0.71	1.26	98	1.72	4.53
C6	0.24	0.76	1.00	12	4.22	11.13
C7	1.61	4.16	5.77	97	3.45	9.12
C8	0.20	0.28	0.48	45	1.87	4.94
C9	1.42	1.69	3.11	97	1.59	4.20
C10	0.51	0.31	0.82	32	0.81	2.14
C11	0.64	0.23	0.87	34	0.48	1.27
C12	0.34	0.35	0.69	25	1.37	3.62
C13	1.94	2.32	4.26	135	1.59	3.14
C16	0.12	0.01	0.13	6	0.11	0.29
C17	0.34	—	0.34	15	—	—
C18	1.01	0.14	1.15	24	0.19	0.50
C19	1.85	3.85	5.70	148	2.78	7.34
C20	2.52	0.03	2.55	60	0.02	0.05

ference by the factor 12/11 to correct for the fact that half of the time the short lived product ThA, is emitted into the chamber with the Tn alpha particle and is hence not counted as a separate entity.

The total activities of column four are the sums of the figures of columns two and three.

In the fifth column of Table VII are listed the readings of local ionization in ions per cm^3- sec determined by cosmic ray electroscopes placed over the material from which the specimens were taken.

The sixth and seventh column list the ratios of the elements uranium and thorium both in terms of alpha activity and in terms of concentration. The first ratio is 4/3 of the ratio of the activities of the two series in equilibrium while the latter is calculated from the former by multiplying it with the ratio with which the same quantities of uranium and thorium emit alpha particles, determined by Kovarik and Adams[35] and Fesefeldt[47] respectively.

VII CONCLUSION

In figures 24, 25, and 26 are plotted the results of the radioactivity determinations as ordinates and the local radiation measurements as abscissae. All rocks of

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47. H. Fesefeldt, Zeit. f. Phys. 86: 605, (1933).

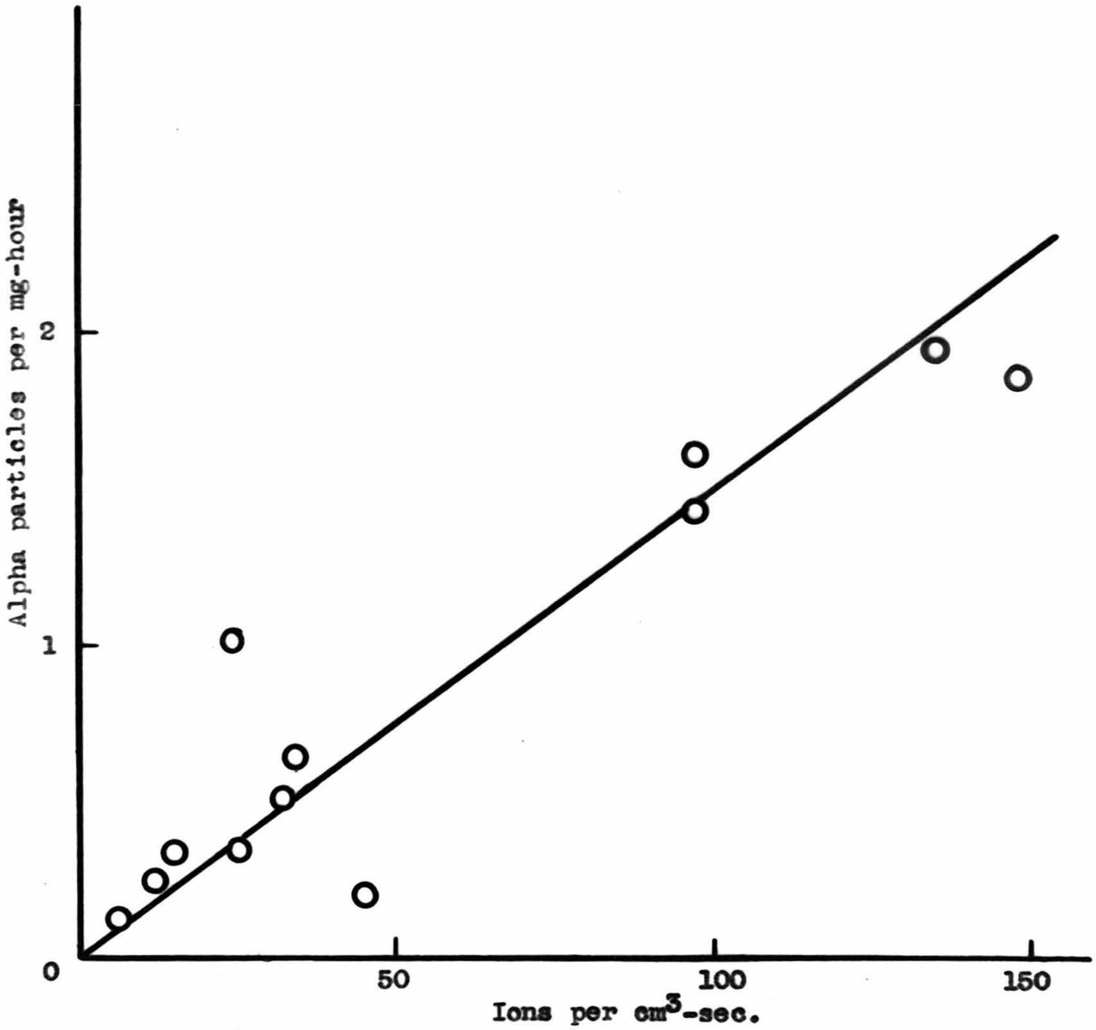

FIG.24. LOCAL RADIATION VERSUS URANIUM ACTIVITY

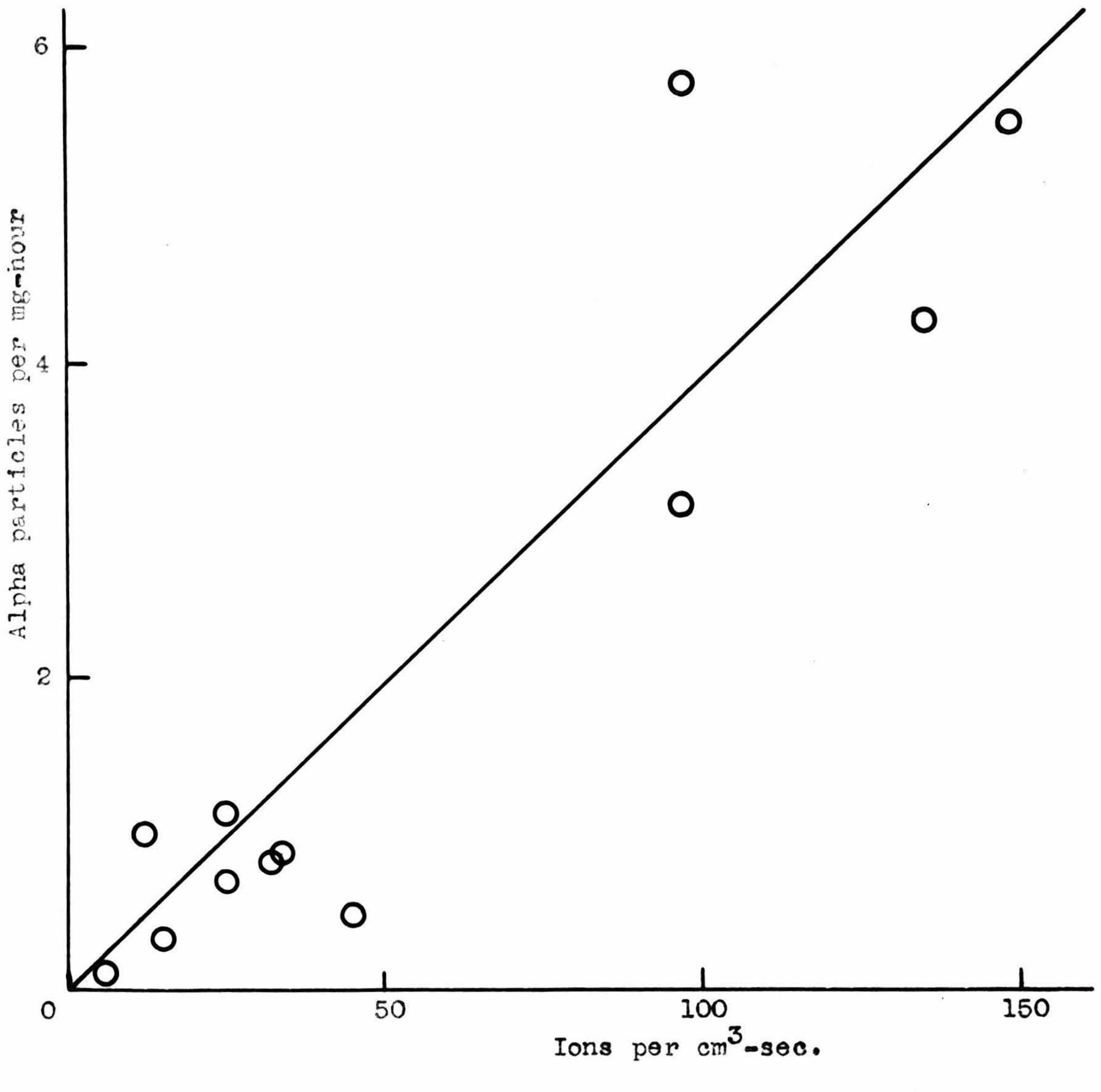

FIG.25. LOCAL RADIATION VERSUS TOTAL ALPHA ACTIVITY

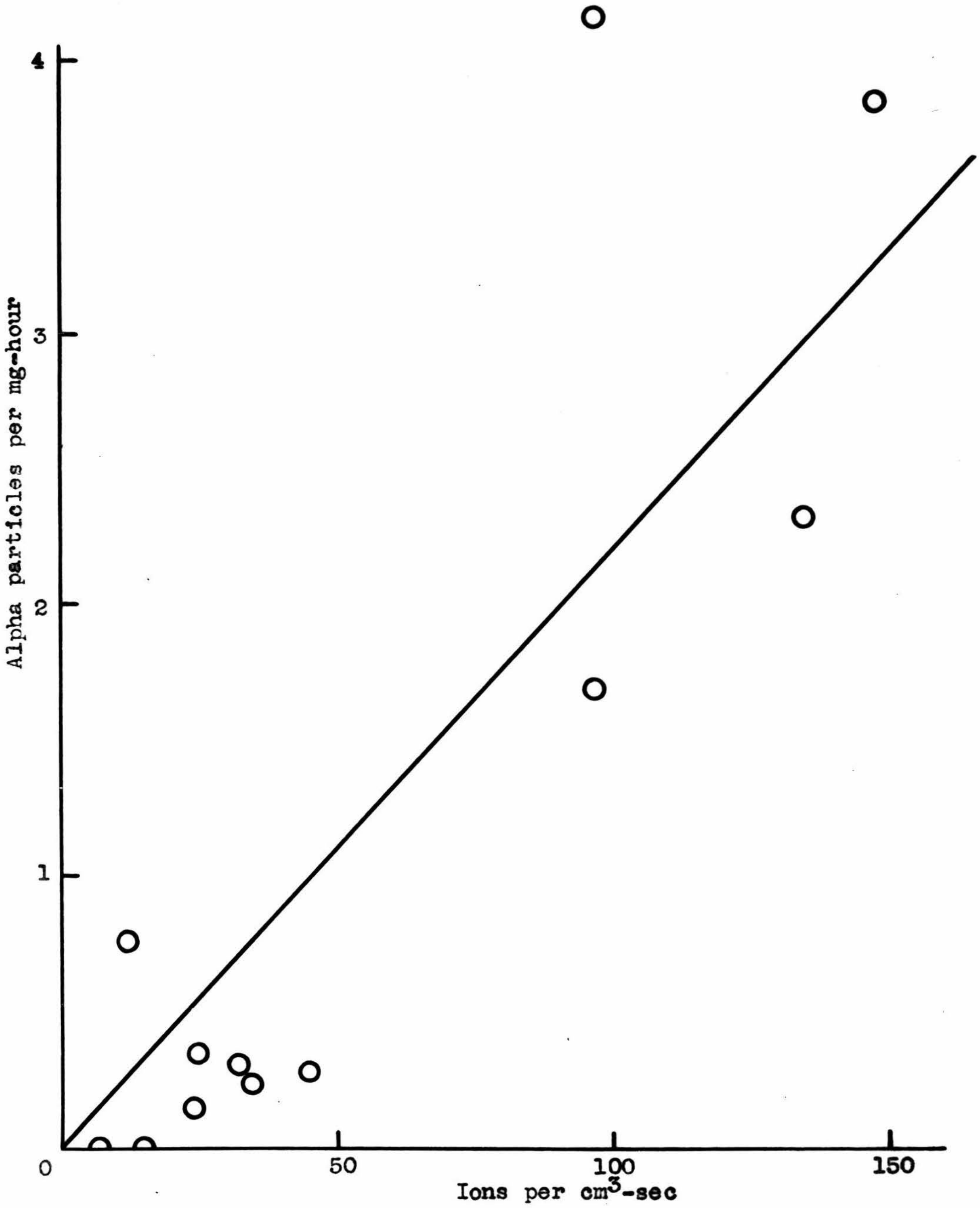

FIG.26. LOCAL RADIATION VERSUS THORIUM ACTIVITY

Tables VI and VII are plotted, with the exceptions of C5, which is a road gravel transported from a distance and not representative of the terrain underlying the electroscope,and C20 which is one type of two rock types of widely different radium concentrations selected from the electroscope site.

It is seen from these figures that a rough correlation exists and that in a general way the alpha activities are proportional to the local radiation.

If the local ionization is produced entirely by the radioactivity of the underlying terrain the data should fit an equation of the form

$$I = K_1(U) + K_2(Th)$$

where I represents the local ionization, (U) and (Th) represent concentrations of uranium and thorium respectively and K_1 and K_2 are constants, dependent on the gamma ray production of the series.

The scattering of the points is such that it seems futile to attempt an accurate fit and thus determine the relative influence of the uranium and thorium concentrations on the local radiation measurements. However, the tendency of the points to cluster more or less closely about a line passing through the origin is good evidence that by far the greater part of the local radiation arises from the radioactive elements in the rocks surrounding the electroscope.

In interpreting the above data recognition should be made of the fact that the electroscope collects radiation from a large amount of material, of which only a small part is available for direct radioactive determination. Sampling variations may be expected to play a major role.

Another interesting result of the data of Table VII is obtained on examination of the Th/U ratios. Four of the rocks are limestones, C11, C16, C17, and C18 and all of these show very low ratios. The fifth rock with an anomalously low ratio is a chert, a type of rock which is formed by chemical deposition in a manner very similar to the deposition of limestone. All other rocks of the suite investigated are igneous or formed from igneous rocks by ordinary physical processes of erosion and deposition.

The average Th/U concentration ratio of these rocks is 5.6, while that of the chemically deposited rocks is 0.42.

Furthermore it is to be observed that the figure 5.6 is more than twice the value usually accepted as reresentative of rocks as a whole. There are not enough determinations in this collection to determine whether or not the former estimates are too low. Nevertheless the predominantly large part played by the thorium series in the total activity of these rocks, and the wide fluctuations in the Th/U ratios observed, prove the necessity of giving major consideration to thorium in geological problems involving the presence of radioactivity in the earth's crust.

Acknowledgements.

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