Gamma-Radiation from Granite

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(Received December 5, 1947)

It has been reported that measurements of the ionization produced by the gamma-rays from Quincy granite are in excess of the ionization computed from the measured uranium, thorium, and potassium content of the rock by a factor of over two. It was concluded that either the measured radioactive contents were much too low or that a hitherto unknown penetrating radiation was given off by the granite.

Measurements of the gamma-radiation from Quincy granite have now been made in such a way that it is unnecessary to *compute* the ionization arising from the reported radioactive contents in order to make the comparison. After the measurements of the ionization produced by the Quincy granite were made, uranium, thorium, and potassium were added in the form of pitchblende, monazite, and potassium carbonate in amounts which would exactly double the reported radioactive contents. The ratio of the measured ionization in the latter case to that in the former was 2.06 ± 0.13. The gamma-ray ionization from the Quincy granite thus seems to be fully accounted for by the measured uranium, thorium, and potassium content.

Now, when the results of the previously reported measurements of the ionization from the Quincy granite and the present measurements are reduced to comparable conditions, the two sets of measurements are in good agreement; therefore, it seems likely that the discrepancy which had indicated a large excess of penetrating radiation, or a large error in the measurement of the radioactive content, may have arisen in the computation of the expected ionization. This conclusion is fully supported by very recent work on the gamma-radiation from potassium which was reported after the preparation of this paper.

1. INTRODUCTION

THE ionization produced by the gamma-rays from Quincy granite according to the measurements made by V. F. Hess[1] exceeded the expected ionization computed from the measurements of the uranium, thorium, and potassium content by a factor of about two. Hess came to the conclusion that either the measured uranium and thorium contents were much too low or that one has to assume that granite, and perhaps most of the other rocks, emits a penetrating radiation of unknown origin. Either conclusion is sufficiently intriguing to warrant further investigation.

It is possible to avoid any computation of the expected ionization in making a comparison of the gamma-ray ionization produced by the granite with that produced by amounts of uranium, thorium, and potassium equal to the reported contents in the rock by adding to the granite amounts of these elements equal to the reported contents. The ionization should be exactly doubled if there exists no unknown radiation from the granite and if the reported contents are correct. An increase in the ionization by only 40 to 50 percent would confirm the result obtained by Hess.

The added uranium and thorium must have all their daughter elements present in equilibrium. Uranium was added in the form of pitchblende from Great Bear Lake. The sample was the remainder of a powder analyzed for uranium. Thorium was added in the form of monazite from Mars Hill, North Carolina, and a part of the same specimen has been analyzed by Marble.[2] Potassium was added as the carbonate.

The principal, if not the only, source of ionization from U and Th in the chamber is the hard gamma-ray from RaC and ThC″, both of which are below radon and thoron in the series. In order to be sure that radon and thoron were not lost from these powdered samples, the materials were bottled and sealed in square-type quart Mason jars. As a further precaution, these jars were stored for 35 days following addition of the internal standards.

A question arises as to whether the background ionization with air surrounding the chamber, which was subtracted in the experiments of Hess from that measured with the granite, might not be lower than the required background be-

[1] V. F. Hess, Trans. Am. Geophys. Union **27**, 670 (1946); Phys. Rev. **72**, 609 (1947).

[2] J. P. Marble, Am. Mineralogist, **21** 456 (1936).

cause of the cosmic-ray transition resulting from the mere presence of the granite. In order to settle this point, several hundred pounds of the ultrabasic rock dunite[3] was collected and sorted at Addie, North Carolina. This rock is practically free of activity. Measurements of the radium content by G. L. Davis at this Laboratory showed 0.007×10^{-12} g Ra per g, which is equivalent to 0.7 percent of the uranium content of the Quincy granite. The potassium content is very small (less than 0.0005 percent), and the thorium content is presumably not very different from that given by the universal ratio of Th/U equal to 3 in rocks. Thus, all of the three elements are present to about the extent of 0.5 to 0.7 percent of the content in Quincy granite. The ionization from the dunite is, therefore, quite negligible. Hess[4] showed with this dunite that any increase in background resulting from cosmic-ray transition in a solid mass surrounding the chamber was quite negligible.

In the experiments of Hess the rock was surrounded by sufficient iron to shield out the gamma-radiation from the walls of the room. In the present experiments, installation of such a shield was not feasible; the true background, therefore, was obtained with a duplicate set of Mason jars filled with dunite.

2. EXPERIMENTAL

The apparatus, primarily designed for the determination of small quantities of radium, has been described.[5] Deflections and hourly calibrations of the Lutz-Edelmann electrometer are recorded on microfile film. As used to determine radium, the apparatus contains a dual system of balancing chambers. In order to measure the gamma-ionization from a source such as granite outside the chamber, the lower chamber was grounded and the upper chamber was surrounded by a circular table, the table top being level with the bottom of the cylindrical chamber.

Without rebuilding the apparatus, it was possible to surround the chamber for an arc of only 194 degrees and for a distance of 45 cm from the axis of the chamber. The upper level of the rock in the Mason jars was approximately level with the top of the chamber.

The most satisfactory arrangement of the 41 bottles on the table to form a closely packed, more or less semicircular mass of rock was established, and positions and bottle numbers were mapped on the table. This "geometry" was rigidly maintained for all the measurements.

For these experiments Dr. Hess kindly lent me 200 lb. of the same Quincy granite that he had used in his measurements. The granite was approximately $\frac{1}{2}$-inch mesh in size. The dunite was crushed and sieved to about the same size. The interstices in this coarse packing of both the granite and the dunite were filled with the same weight of fine dunite (through 32 on 65 mesh). This served the double purpose of increasing the stopping power of the granite-filled bottles to approximately that of the dunite-filled bottles, and of providing a carrier for the internal standards.

Alternate determinations of the ionization with dunite-filled bottles and with the granite-filled bottles were first made. The internal standards were then added to the granite-filled bottles, which were sealed and set aside for 35 days, after which the alternating measurements were repeated. Each determination lasted 16 to 22 hours, the totals being 162 hours for dunite, 66 hours for the granite, and 58 hours for the granite with the internal standards.

All measurements were made with the original filling of nitrogen in the ionization chamber, the density being such that the factor for reduction to 0°C and 760 mm was 1.108. In these experiments the procedure is purely one of comparison, and, therefore, the results were not reduced to N.T.P.

3. RADIOACTIVE CONTENT OF QUINCY GRANITE

Keevil[6] has measured the radium and thorium content of Quincy granite from several quarries —and at several depths in the Swingle quarry, the one from which the material was taken for Hess. Evans and Goodman[7] report a radium and thorium value for the 150-ft. level. The average

[3] The average atomic number of dunite is 10.1, that of Quincy granite 10.2; therefore, there should be no difference in the cosmic-ray transition in the two rocks.

[4] V. F. Hess, Phys. Rev. **72**, 609 (1947).

[5] Wm. D. Urry and C. S. Piggot, Am. J. Sci. **239**, 633 (1941).

[6] N. B. Keevil, Am. J. Sci. **36**, 406 (1938).

[7] R. D. Evans and C. Goodman, Bull. Geol. Soc. Am. **52**, 459 (1941).

TABLE I. The gamma-ray ionization, I, produced by Quincy granite, and Quincy granite to which internal standards of uranium, thorium, and potassium were added to double the reported radioactive content.

Date	Dunite (background)	Quincy granite	Quincy granite with internal standards
June 1947	5.18 ± 0.04[a]	6.22 ± 0.13	
	5.16 0.05	6.02 0.11	
	5.49 0.17	5.96 0.11	
	5.01 0.10	6.29 0.07	
	5.11 0.12		
	5.45 0.04		
Aug. 1947	5.43 0.08		7.28 ± 0.09
	5.07 0.10		7.09 0.06
	5.26 0.08		7.06 0.08
Weighted mean	5.264 ± 0.021	6.165 ± 0.048	7.124 ± 0.042
Weighted (June	5.26 ± 0.02)		
mean (Aug.	5.28 ± 0.05)		
Quincy granite with internal standards less background			1.86 ± 0.05
Quincy granite less background			0.90 ± 0.05
Ratio of Quincy granite with internal stds/ Quincy granite			2.06 ± 0.13

[a] The root mean square probable error, σ, used to weight the individual determinations by $1/\sigma^2$.

of the results of both investigators for the levels of 12, 120, 150, and 350 feet yields a radium content of 0.99×10^{-12} g per g, with variations from 0.91 to 1.14. The mean thorium content is 8.95×10^{-6} g per g, with variations from 7.0 to 12.4. Dr. L. F. Curtiss of the National Bureau of Standards determined the radium content of the Hess sample to be 1.02×10^{-12} g per g. The radioactive content of the Quincy granite is apparently remarkably uniform. For our purposes we shall take 1.00×10^{-12} g Ra per g and 8.95×10^{-6} g Th per g.

Dr. E. G. Zies of this Laboratory kindly determined the potash content of a quartered sample of the Hess Quincy granite. It contains 4.64 percent K_2O, equivalent to 3.85 percent potassium. This result is in excellent agreement with analyses by Warren[8] on other samples.

4. INTERNAL STANDARDS

The total weight of granite was 51,560 grams, which required the addition of 3508 grams of potassium carbonate to double the potassium content of 3.85 percent. By analysis at this Laboratory, the potassium carbonate contained 0.55×10^{-12} g Ra, equivalent to 0.0055 g U in the total weight to be added.

[8] C. H. Warren, Proc. Am. Acad. **49**, 227 (1913).

The monazite contained 5.56 percent Th, which required the addition of 8.299 grams of monazite to double the thorium content, but the monazite also contained 0.0178 percent U, equivalent to 0.00147 g U in the total weight to be added.

The radium content of the granite is equivalent to 2.87×10^{-6} g U per g, which will be doubled by the addition of 0.148 g U, of which 0.007 g will be contributed by the impurities calculated above in the potassium carbonate and monazite, leaving 0.141 g U to be provided by 0.266 g pitchblende containing 53.00 percent U.

In order to insure uniform distribution, the above weights required to double the reported contents were divided by the number of bottles, and the corresponding amounts were added to each bottle. Each bottle contained the same weight of granite. The amount of fine dunite filling the interstices between the granite, with which the internal standards were mixed, was reduced by the weight of the potassium carbonate at this stage.

5. RESULTS

The results are reported in Table I in ion pairs per cc per sec. (designated by I). The accuracy of the absolute values of I is of no importance in this comparison method, but as shown below, the electrostatic capacity has been accurately determined.

Within the narrow limits of the probable error, the ionization produced by the gamma-rays from Quincy granite is just doubled (2.06 ± 0.13) *by adding the amounts of uranium, thorium, and potassium equal to the reported contents.* There is no reason to suspect the reliability of the modern techniques used in the determinations of the small amounts of radium and thorium in rocks, nor to doubt the accuracy of the standards employed in these techniques.

Had the experiment confirmed the result reported by Hess, the measured ratio of the ionizations in the above experiment (if we use his figures[9]) should have been $(9.50+4.12)/9.50=1.43$, or, stated another way, a value of only $1.29I$ should have been obtained with the internal standards. Hess employed radium, thorium, and

[9] V. F. Hess, Trans. Am. Geophys. Union **27**, 670 (1946). See Table 2.

potassium contents of the Quincy granite, which were somewhat lower than those based here on all the observations in the literature, on a more recent determination by Dr. Curtiss on the sample used by both of us, and on the determination of the potash content at this Laboratory. The effect of using the newer data is to raise the ratio slightly—from 1.43 to 1.49.

At one time it was thought that components of the gamma-radiation softer than those arising from RaC and ThC″ might have caused ionization through the thin wall used by Hess. The softer gamma-rays were neglected in the calculation. Hess[10] has recently repeated the experiments with sufficient lead between the chamber and the granite to absorb any gamma-radiation softer than that arising from RaC, ThC″, and K. The result was essentially the same and equal to 1.55 in terms of the above ratio.

There is, of course, the possibility that an unknown hard gamma-radiation is emitted from the uranium or thorium series or from potassium. Calculation of the expected ionization requires a knowledge of Eve's constant, the number of ion pairs per cc per sec. per gram of uranium (thorium or potassium) at unit distance from the center of the ionization chamber. The only likely source of error, involving an unreported hard gamma-ray among the familiar radioactive elements, is in the value of Eve's constant for potassium. This constant has not been measured. Calculating for the first method employed by Hess, namely, from the three photons emitted for every 100 disintegration electrons, we find that the discrepancy between the computed ionization and the measurements over a flat surface of the Quincy granite would be entirely eliminated if the correct figure were about 15 photons per 100 electrons. One would doubt that Gray and Tarrant[11] could have been in error to this extent.

6. SUPPORTING EXPERIMENTS

A few experiments were performed which support the major results given in Table I.

In one series of measurements on the ionization produced by the Quincy granite with internal standards, the bottles were rearranged in the $(41-n)$ positions. A value of $7.09 \pm 0.06I$ was obtained, which, compared to $7.16 \pm 0.06I$ for the measurements in the normal positions of the numbered bottles, demonstrates uniform distribution of the internal standards.

Eve's constant for radium was measured with a 100-microgram radium standard from the National Bureau of Standards, surrounded by 1.50 cm of lead and placed at 50.0 cm from the center of the chamber. With allowance for the absorption in the 0.565-cm brass wall of the chamber (absorption coefficient $\mu = 0.370$ cm^{-1}) and in the lead ($\mu = 0.531$ cm^{-1}), the value of Eve's constant K at 0°C and 760 mm of nitrogen was found to be $5.64 \times 10^9 I$ for a ratio A/W of wall area to volume in the chamber of 0.393. This value can be compared with the plot of K against A/W given by Hess and Balling[12] by dividing by their factors of 1.062 for 20°C and 767.7 mm of nitrogen and 1.097 for the absorption in their brass wall of 0.25-cm thickness. The result is $4.85 \times 10^9 I$, which is very close to their curve of K against A/W. Such agreement lends strong support to the conclusion that the measurements are comparable in the two laboratories and indirectly substantiates the correctness of the determination of the electrostatic capacity of the ion-collecting system.

Without moving the radium standard, measurements were made with the dunite and the granite interposed, in turn, between the 100 micrograms of radium and the chamber. The ionization produced by the gamma-rays from the radium was reduced, respectively, to 0.053 and 0.051 of the ionization without absorption in the dunite or in the granite.

It was previously found that the dunite reduced the stray radiation in the room by about $1I$; accordingly, about $0.05I$ is produced by the stray radiation passing through the dunite. From the above absorption experiments, the stopping power of the dunite-filled bottles and of the granite-filled bottles cannot differ by more than 10 percent; hence, the possible error caused by different absorption of the stray radiation in the dunite and in the granite is not over $0.005I$.

From the absorption experiments, one can

[10] V. F. Hess, Phys. Rev. **72**, 609 (1947).

[11] L. H. Gray and G. T. P. Tarrant, Proc. Roy. Soc. London **A143**, 681 (1934).

[12] V. F. Hess and E. Balling, Trans. Am. Geophys. Union **26**, 237 (1945).

compute the absorption coefficients for the rock-filled bottles. With dunite $\mu = 0.0837$ cm^{-1} for a bulk density of the bottles in position on the table, $\rho = 1.935$-g cm^{-3}. With the granite $\mu = 0.0825$ cm^{-1} for $\rho = 1.84$-g cm^{-3}. The mass absorption coefficients are thus $\mu/\rho = 0.043$-cm^2 g^{-1} and 0.045-cm^2 g^{-1}, respectively, in good agreement with the accepted value of 0.045 for the gamma-rays from RaC, and also with a similar determination by Hess, which is 0.043 for the Quincy granite.

7. COMPARISON OF MEASUREMENTS

The ionization, which would have been produced in the experiments reported here if the whole chamber had been completely surrounded by an infinite thickness of the granite, can be roughly estimated and compared with the same quantity q_∞ determined by Hess.

The thickness of the wall of the chamber used by Hess was 0.25 cm (brass), whereas the thickness of the chamber wall used here was 0.565 cm (also brass). The solid angle subtended by the granite amounted to $4\pi \times 0.138$ steradians. Thus, if the present chamber had a wall thickness of 0.25 cm and granite completely surrounded the chamber, q_d would be $(0.90 \times 1.124)/0.138 = 7.33I$.

In the present experiments, the chamber was surrounded to a depth of 38 cm, but there was an air gap of 2 cm on the average between the wall of the chamber and the nearest granite. Under these conditions, and taking the absorption coefficient $\mu = 0.0825$ cm^{-1} as determined above for RaC (which will be close enough to the mean value for RaC, ThC″, and K in the granite), the value of $q_\infty = 7.33/0.804 = 9.1I$. This is in good agreement with the value of q_∞ of $9.5I$ determined by Hess, particularly in view of the accuracy of such a calculation. There is, thus, no serious disagreement between *measurements* of the ionization produced by the gamma-rays from Quincy granite in the two laboratories.

8. CONCLUSIONS

These experiments show no source of gamma-ray ionization in the Quincy granite other than the reported contents of uranium, thorium, and potassium. The measurements of the ionization produced by the gamma-rays from the Quincy granite are in good agreement with the determinations reported by Hess. Hence, there seems to be only one source of discrepancy between the absence of any extraneous gamma-radiation in the Quincy granite as established by the present experiments and the large excess, reported by Hess, of measured gamma-radiation over that computed from the uranium, thorium and potassium content. The discrepancy apparently lies in the equations for the computation or in the data entered in these equations.

Since this paper was first drafted, Gleditsch and Gráf[13] have reported a value of 3.6 gamma-quanta per second per g K. There are, therefore, 15.7 photons per 100 electrons on the basis of 23 beta-rays per second per g K, the latter figure being that used by Hess. This ratio between the photons and electrons is almost exactly that previously calculated to be necessary in order to remove the discrepancy in the experiments conducted by Hess at the Swingle Quarry (*vide supra*). The value of Eve's constant for potassium can be obtained from that for radium, combined with the value of the equivalency of potassium and radium in gamma-producing capacity determined directly by Gleditsch and Gráf to be 1.23×10^{-10} g Ra. Eve's constant for potassium $= 0.590I$ per g K for the chamber used by Hess. This yields a value of $6.34I$ arising from the potassium content (3.85 percent) of Quincy granite or $9.7I$ for U+Th+K for an infinite thickness and 4π steradians. This is in very satisfactory agreement with the measurements in both laboratories.

One may, therefore, conclude that there is no source of extraneous gamma-radiation in Quincy granite and probably not in other rocks. The experiments reported here, taken together with those performed by Hess, strongly support the latest value for the gamma emission of potassium-40 determined by Gleditsch and Gráf.

9. ACKNOWLEDGMENTS

It is a pleasure to acknowledge the continued and kindly cooperation of Dr. V. F. Hess, with whom personal contact was maintained throughout the course of these experiments and during the preparation of this manuscript. To Dr. H.

[13] E. Gleditsch and T. Gráf, Phys. Rev. **72**, 640 (1947).

Yagoda, of the National Institute of Health, the author is indebted for the remainder of a powdered sample of pitchblende that he had analyzed for uranium. The National Museum, Washington, D. C., kindly supplied the sample of monazite through the good offices of Messrs. E. P. Henderson and Wm. F. Foshag, and thanks are due Dr. J. P. Marble for his advice in the choice of a suitable thorium mineral. Dr. E. G. Zies of the Geophysical Laboratory kindly contributed to the problem through his determination of the potash content of the Quincy granite. Dr. Merle A. Tuve of the Department of Terrestrial Magnetism, Carnegie Institution of Washington, was responsible for arousing the author's interest in the problem during the course of an informal discussion concerning the possibility of obtaining for Dr. Hess a rock that would be essentially free of any radioactivity. The Harbison-Walker Refractories Company of Pittsburgh, Pennsylvania, kindly contributed several hundred pounds of such a rock in the form of dunite from their quarry at Addie, North Carolina.

PHYSICAL REVIEW VOLUME 73, NUMBER 6 MARCH 15, 1948

The Beta-Spectra of Cu^{64} as a Test of the Fermi Theory

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(Received December 10, 1947)

The momentum spectra of the negatrons and positrons emitted by Cu^{64} have been measured with high resolution under such conditions that the usual distortions resulting from source thickness and backing, scattering, and finite counter window are completely negligible for almost the entire range of energies. Whereas the Fermi theory predicts the correct distribution for the high energy region, it is found that there are more negatrons and positrons than are predicted by the theory at low energies. The shapes of the spectra and the fact that no nuclear gamma-rays are observed preclude the possibility that they are complex spectra. Deviation from the Fermi theory occurs for energies below 0.270 Mev for the positrons and for energies below 0.190 Mev for the negatrons.

Incidental to these measurements, the end-point energies have been determined as 0.657 ± 0.004 Mev for the positrons and 0.571 ± 0.002 Mev for the negatrons. The ratio of the total number of electrons to positrons is 2.0.

I. INTRODUCTION

A DETAILED investigation of the momentum distributions of the negatrons and positrons emitted by Cu^{64} has been made in order to test the validity of the Fermi theory of beta-decay.[1] When applied to reliable experimental data, the Fermi theory, using Fermi's original choice of interaction, has been remarkably successful in describing the high energy end of the beta-ray spectrum. The Kurie plot[2] of the experimental data yields a straight line for a large part of the distribution of all allowed and many forbidden transitions and is a valuable tool for accurately determining the end-point energy.

Many experimenters,[3] however, have observed that there are apparently more particles in the low energy region of the distribution than are predicted by the Fermi theory. This deviation from the theory has usually been attributed to distortion resulting from instrumental difficulties such as finite source thickness and backing, absorption in the counter window, and scattering from the walls of the spectrometer. In some cases, the excess at low energies is caused by the fact that the spectrum is complex,

[1] E. Fermi, Zeits. f. Physik **88**, 161 (1934).
[2] Franz N. D. Kurie, J. R. Richardson, and H. C. Paxton, Phys. Rev. **49**, 368 (1936).
[3] A. W. Tyler, Phys. Rev. **56**, 125 (1939). J. L. Lawson, Phys. Rev. **56**, 131 (1939). K. Siegbahn and H. Slatis, Ark. f. Mat., Ast., o Fysik **32A**, No. 9 (1946). A. A. Townsend, Proc. Roy. Soc. **A177**, 357 (1941).