MEMOIRS OF THE COLLEGE OF SCIENCE, KYOTO IMPERIAL UNIVERSITY, SERIES B,
VOL. X, NO. 1, ART. 4, 1934.

Radium Content in Granites in Nippon (Preliminary Report)

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With 3 Text-figures

(Received May, 5, 1934)

In this paper it is intended to give briefly an arrangement and technique for the determination of radium in rocks as well as the results of preliminary examination of some granites in Nippon.

Since H. MACHE, St. MEYER and E. SCHWEIDLER[1] first undertook to determine the radioactivity of rocks, various workers such as R. J. STRUTT (Lord RAYLEIGH), J. JOLY and others, have also persued the same investigation. More recently ELLSWORTH[2] and PIGGOT[3] made additional determinations of radium in rocks. Since ELLSWORTH'S work was undertaken mainly from the economic point of view, it has no direct bearing on the general distribution of radium in rocks except that it confirms the concentration of radium in pegmatite. PIGGOT's results are noteworthy as the obtained figure 0.897×10^{-12} grams of radium per gram of the sample for the average of 16 rocks (mainly granite) from eastern North America is too small compared with the average for the igneous rocks 2.5×10^{-12} grams of radium per gram of rock. This figure is adopted by HOLMES[4] as the most probable.

One of the problems of the pressent investigation is to determine the truth of the inference that acid rocks, excluding pegmatite, have as a rule a higher content of radium. A second problem is to find the relationship between the radium content and other geological or petrographical phenomena.

The methods of measuring the radioactivity of rocks may be roughly divided into three types: solution method, fusion method,

and direct method. The first two may be summarized as emanation method. The difference between them is that in the former emanation is expelled from the solution of the rock, whereas in the latter the same is done from the fused melt of the rock. The last is a method of determining the radioactivity by directly counting alpha-particles emitted from the sample with the GEIGER-MÜLLER counting tube. Since all of these methods have their own advantages, it is difficult to decide which is the best. The discussion of the methods is left for elsewhere, and here will be described the general course of determination which I have used following the solution method of STRUTT and others with several modifications.

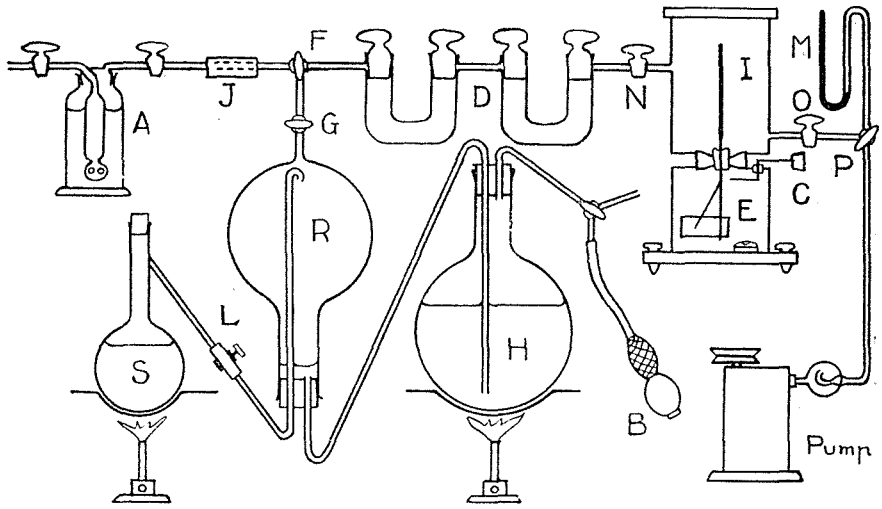
Preparation of rock solution

About 100 grams of rock are broken to pieces to 35 mesh in an iron mortar, and then pulverized in an agate mortar until all will pass 150 mesh. 2.5 grams of this sample is mixed well with eight grams of anhydrous sodium carbonate and fused in a platinum crucible. After complete decomposition, the charge is cooled and removed from the crucible to bleach with distilled water in a basin on a water-bath. When the cake is almost disintegrated, pure hydrochloric acid is added to insure complete dissolution. The fluid in the basin now contains all of the rock constituents in solution as chlorides, except the silica, most of which is in solution as soluble silicic acid and partly as insoluble particles. The mixture in the basin is then evaporated to perfect dryness in order that the silica may be rendered insoluble. When this process is completed, it is moistened with a few cubic centimeters of hydrochloric acid and then with hot water to dissolve the basic salts. Filtration removes all the silica, and the filtrate thus obtained is kept in a preserving flask of about 500 c.c. Solution corresponding to 10 grams of the sample is collected in a flask leaving a space of about 100 c.c. for subsequent boiling. If there is a suspicion that the rejected silica might be contaminated with radium salts, the former should be treated with hydrofluoric acid and the resulting residue added to the main solution after it is converted into solution with hydrochloric acid. That this precaution, however, may not be needed in an ordinary occasion was demonstrated in several sets of examinations.

Apparatus and arrangements

The whole arrangement of apparatus, essentially the same as that cited by J. JOLY,[5] B. BOLTWOOD[6] and others, is shown in Fig. 1.

Fig. 1.



Schematic diagram showing arrangement of apparatus.

In Fig. 1, S is a 500 c.c. flask containing the solution of rock to be examined. It has a side tube the end of which is closed by means of rubber tube with a good screw-pinchcock L. This is connected with the receiver R as shown in the figure. The latter has a capacity of 2500 c.c. and serves as a temporary reservoir for the air containing emanations and vapour. The thorium emanation (toron) works itself out almost completely during its suspension in R, while the radium emanation (radon) survives owing to its longer life until it is sucked into the ionization chamber I through the drying tube D. Chlor-calcium is used as the drying agent. The ionization chamber, a cylindrical brass vessel of about 2400 c.c. capacity, is attachable to the gold-leaf electroscope E. Reading is taken by means of a low power ($\times 20$) microscope with a micro-scale in its image plane, across which the gold-leaf is observed to move.

Operation

Before each determination the natural leak of the electroscope must be measured as it varies from time to time. For this purpose,

the ionization chamber is evacuated and then fresh air is introduced into it through the dryers A and D, the former being a gas washing bottle filled with concentrated hydrosulphuric acid. When atmospheric pressure is attained in the ionization chamber, the reading is started and continued at an interval of 15–20 minutes. The gold-leaf of the electroscope is always kept charged nearly to the same potential whether it is in use or not. This can be secured by good insulation of the charged system. This process is important and effective to avoid the difficulties caused by the lag in electrostatical properties of the insulator material. Several repetitions of charge and discharge just before the measurement are also recommended to make the gold-leaf work regularly. After an hour or so from the beginning, the falling rate of the leaf becomes practically constant, and the number of scale divisions crossed by the image of the leaf (precisely of a very fine fibre attached to the leaf) in unit time represents the natural leak. The jointer J is removed to make the inside of R communicate with the outer air. The reservoir R is filled with boiling water which is supplied from the large flask H by means of a hand bellow B, and the cock G is closed. The standard solution in S containing known amount of radium is boiled vigorously, the pinchcock L and three-way cock K being open. With effervescence of the solution the pressure of steam in S rapidly increases and will force the hot water back to H. On the other hand the ionization chamber, calcium tubes and their accessories that lead to the cock G are evacuated, and the cock O is closed. When the spherical part of R is nearly full of air with steam and emanations, the flame under S is quickly removed and the pinchcock L is closed. By opening G, the air with radon in R is gradually sucked into the ionization chamber followed by the rising of the water level in R which is to be stopped by closing G before the head reaches it. Again the cock L is opened and the gas flame is brought under S to continue boiling. When R is half full of vapour, the flame is again removed, and blasting of the cool air upon the upper half of R causes a falling of pressure inside. Then a dash of fresh air is brought into R through A, J, F, G. G is closed, the flame is again brought into place, and the process described above is repeated four times. During all the time the water in the flask H is kept as near to the boiling point as possible in order to keep the solubility of radon as small as possible. The repetition of the process four times to drive the radon into the ionization chamber

usually requires 20-30 minutes. After the completion of this process, a fresh dry air current run into the ionization chamber sweeps any gases remaining in the path. When the atmospheric pressure is established in the ionization chamber the stopcock N is closed. Reading is taken between the third and fourth hours after the first boiling off of radon from the solution. The rate of fall of the gold-leaf across the scale is expressed in terms of scale divisions per minute. From this must be subtracted the natural leak determined just before the run, and the figure thus obtained corresponds to the radioactivity due to the standard solution whose radium content is known. Exactly the same procedure is followed with the sample to be tested and the radium content of the sample is derived from the figure given by the standard solution.

Calibration of the electroscope

The standard solution used in the present investigation was prepared in the laboratory of Dr. S. IIMORI, at the Institute of Physical and Chemical Research in Tôkyô. The original solution contained 3.11×10^{-8} grams of radium in 84.1 c.c. To render the radium contents of the standard solution and of the rock solution to be analysed to the same order, the original solution was diluted with distilled water to the appropriate concentration. With a burette the original solution was measured out into a measuring flask of 250 c.c. capacity. Readings were taken accurately and the difference of the initial and final readings was 9.99 c.c. The taken solution was diluted with distilled water just to 250 c.c. and was noted as the normal solution A. 5 c.c. of this normal solution was transferred with a pipette into a preserving flask of 500 c.c. capacity. Then was added 5 c.c. of pure hydrochloric acid with enough distilled water to fill the flask nearly eight-tenths full, leaving room for subsequent ebullition. The solution was then boiled for about fifteen minutes, closed and set aside. Each standard solution prepared in the manner just described contains 7.39×10^{-11} grams of radium. The results of calibration are listed in the following.

Standard solution	V	VI	VII
Total leak	0.317	0.324	0.321
Natural leak	0.020	0.023	0.020
Leak due to the radon	0.297	0.301	0.301

Thus the mean leak due to the standard solution is 0.300 divisions per minute, or one division per minute fall of the electroscope leaf is equivalent to 24.66×10^{-11} grams of radium. This figure is the constant of the electroscope, which is used throughout this paper.

Results

The radium content of granites from seven different localities in Nippon, two hypabyssal rocks and allanite (orthite) from Sirakawa, Kyôto was determined, and the results are shown in Table I.

Table I

Rocks	Locality	Ra-content in 10^{-12} grm./grm.
Granite	Sirakawa, Kyôto	1.41, 1.41, 1.41, 1.36, 1.48 } 1.41
Granite	Yagi, Kyôto	1.98, 1.90, 1.88 } 1.92
Aplite	Sirakawa, Kyôto	1.53, 1.53, 1.53 } 1.53
Lamprophyre	Sirakawa, Kyôto	0.37, 0.37 } 0.37
Granite	Atehama, Kagawa	0.89, 0.79 } 0.84
Granite	Azi, Kagawa	0.79, 0.72 } 0.76
Granite	Misasa, Tottori	2.47, 2.29 } 2.38
Granite	Naegi, Gihu	2.27 } 2.27
Granite	Kamado, Gihu	1.65 } 1.65
Allanite	Sirakawa, Kyôto	48.6 } 48.6

General remarks

In biotite as a constituent mineral of granite, small circular darkened spots, often in concentric rings, may occasionally be observed under the microscope. These are known as pleochroic halos, and this colouration is explained by effects due to alpha-particles emitted from radioactive inclusion. Of the examined granites, those from Sirakawa, Yagi, Naegi and Kamado are found to be associated with halos. The representative examples of the pleochroic halos observed are shown in Figs. 2 and 3. Halos are hardly seen in

Fig. 2

Pleochroic halo developed in biotite in Sirakawa granite. (×140).

Fig. 3

Pleochroic halo developed in biotite in Yagi granite. (×140).

the biotite granites from Azi and Atehama. In granite from Misasa, halos were rarely observed. Since this granite lacks fresh biotite crystals, the halos, if existed, might have been obscured. In conclusion, there is a tendency that higher radium content of granite corresponds to the considerable presence of the pleochroic halos excepting the granite from Misasa, which is associated with unusually radioactive hot spring. This relation, however, may not always hold, as the pleochroic halos can develope in the presence of the thorium- or actinium-series as well as the uranium-radium-series.

The hyperbyssal rocks, aplite and lamprophyre, are found as dykes or veins in the granite boss in the northeastern part of Kyôto city. As they represent the salic and femic extremities differentiated from the granitic magma, they stand in the relations of "comple-

mentary rocks" in BRÖGGER's sense. With respect to the radium content, however, the simple mean of the two does not approach that of granite from the same locality as shown in the table I. Any definite conclusion from the present result will be unjustifiable owing to scantiness of the data, difference in age of formation of the rocks and other unsolved conceivable conditions, and it is desired to carry on this investigation to solve this difficulty. It must be noted that there is a conspicuous difference in the distribution of radium in these two dykes, i. e. aplite is four times as radioactive as lamprophyre. It may be said that this fact implies the law of distribution of radium in the igneous series ranging from femic rocks to salic ones.

The average radium content of seven granites is 1.60×10^{-12} grams per gram rock. The greatest is 2.38×10^{-12} for Misasa granite and the least is 0.76×10^{-12} for Azi granite.

Of the twenty-eight rocks analysed by STRUUT,[7] ten granites reveal the mean radium content of 2.80×10^{-12} grams per gram rock. Among them are granites from England, Scotland, Egypt and Cape Land ranging from 4.78×10^{-12} (Rhodesian granite) to 0.36×10^{-12} (granite from the Isle of Rum). On the other hand, JOLY's measurement on the mixture of sixty-three granites of equal mass gives 2.7×10^{-12} grams per gram of rock.[8] In 1914, HOLMES[6] cited 127 analyses of acid rocks which he regarded as representative (Table II).

Table II

	Number of specimens analysed.	Ra-content in 10^{-12} grams per gram of rock.
HOLMES and others	41	2.51
JOLY	86	3.01

JOLY's results are tabulated separately as his method of analysis seems to have given higher values than those obtained by anyone else. More recently C. S. PIGGOT (*op. cit.*) analysed 18 rocks including thirteen granites, one gneiss, two schists, and two gabbros of the eastern seaboard of the United States. With the exception of the two granites which have obvious radioactive associations, the average radium content of the rock so for examined is 0.897×10^{-12} grams of radium per gram of rock. It is to be noted that PIGGOT

uses this figure (0.9×10^{-13}) as the average of 16 granites in comparison with that of Hawaiian lavas[10], but this is not proper since among the former are included gabbros and some rocks other than granite as is cited above. WILLIS[11] also made similar mistake in discussing the distribution of radium in the earth. If we regard the granite only, the average radium content of the thirteen specimens, computed from the original data of PIGGOT (*op. cit.*), will be 1.40×10^{-12}, and if the two radioactive granites be excluded according to his suggestion, this figure will become 1.00×10^{-12}. The two excluded are granites from Stone Mountain, Georgia, and North Jay, Maine, containing respectively 3.81×10^{-12} and 3.39×10^{-12} gram of radium per gram of rock.

Several examples cited above may be contrasted with Nipponese granites, for which the mean radium content is, as already shown, 1.60×10^{-12} grams of radium per gram of rock. This figure approaches PIGGOT's average. Here it must be recalled that many radioactive halos are present in most of the Nipponese granites examined. And in particular cases, allanite, a radioactive mineral, is found in the Sirakawa granite; and the Misasa granite is taken from the Misasa hot springs, famous for their high radioactivity. According to Dr. ISHIZU[12] a litre of water from one of the springs contains 516.87×10^{-10} curies radon. Considering these conditions we can suppose the figure obtained for the mean radium content of the granites so far examined to be at least not too small for the general mean for the granites from all other parts of the Nippon Islands. Thus we might expect that the mean radium content of Nipponese granites would be somewhat smaller than the means for European granites obtained by several investigators. At present we will leave this problem only as a suggestion and wait until more data are accumulated.

In completing the first report I wish to thank Prof. M. MATUYAMA for his initial suggestion and kind guidance throughout this work.

Summary

1. An arrangement and procedure of determining radium content in rocks by solution method is described.

2. Radium content of seven granites, two hyperbyssal rocks, and allanite were determined.

3. The mean radium content of the seven granites taken from different parts in Nippon is 1.60×10^{-12} grams of radium in one gram of rock.

4. Allanite from Sirakawa in Kyôto contains 4.86×10^{-11} grams of radium per gram of mineral.

5. Granites with many pleochroic halos in biotite were generally richer in radium than those which lack pleochroic halos.

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