Radium Contents of Rocks in Nippon, I.

By

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With 4 Text-figures

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Abstract

Radium contents of granites were measured using the fusion method in which radon was expelled from the melt of rock substance by heating in an electric furnace. The average radium content of twenty granites was determined to be 1.46×10^{-12} grams per gram. The principle, apparatus and technique of the measurement were described as well as the critical discussion of the method. Separation of silica was also carried out and it was observed that radioactivity has the general tendency to increase with silica-percentage. Suggestion was made that radium contents vary antipathetically with alkali-lime indices of the rock series.

1. Introduction

Radioactivity of rocks from various parts of the world has been investigated by many, but so far the Nippon Islands have been almost, if not entirely, devoid of such investigation. It is, therefore, desirable to have data of Nippon that may afford some contribution to the problem on the distribution of radioactive elements throughout the crust of the earth. The present paper is a report of a step in the work aiming at such a contribution, in which are paid special attentions from geological, petrological and geochemical points of view.

In the preliminary report[1] were described some tests on granites by the so-called "solution method." In this paper the radium determination by the "fusion method" is treated instead.

Further investigations are now on their ways and the author is greatly indebted to Prof. M. MATUYAMA for his kind suggestions and encouragement.

2. The principle of the method

A quantity of pulverized rock is fused with alkaline carbonate and the resulted cake is kept closed in a vessel until equilibrium is established between the contained radium and the radon generated from it. The radon initially existed in the rock is driven off during the fusion. Radium in the solidified melt, however, continuously produces radon at a constant rate of n particles per second, while radon decays itself at a rate proportional to the present number of atoms N. Therefore, at any time t, the rate of net increase of radon is ;

$$dN/dt = n - \lambda N \qquad \dots\dots\dots\dots\dots\dots\dots\dots\dots\dots\dots(1)$$

where λ is the transformation constant of radon.

The solution of this differential equation is ;

$$N = ae^{-\lambda t} + n/\lambda \qquad \dots\dots\dots\dots\dots\dots\dots\dots\dots\dots\dots(2)$$

where a is an integration constant. Since radon is absent at the end of the fusion, we have $N = 0$ at $t = 0$, whence we have $n/\lambda = -a$. As the time elapses and the number of radon atoms increases, so does the number which is transforming per second; and finally an equilibrium state will be approached after a sufficiently long time. Let N_e denote the number of atoms of radon at this equilibrium state. Putting $t = \infty$ in the equation (2), we get $N_e = n/\lambda (= -a)$. The equation (2), therefore, becomes :

$$N = N_e(1 - e^{-\lambda t}) \qquad \dots\dots\dots\dots\dots\dots\dots\dots\dots\dots\dots(3)$$

The transformation constant λ of radon is $2.097 \times 10^{-6} \text{sec.}^{-1}$ after RUTHERFORD. In Fig. 1 the ratio of accumulated radon N to the equilibrium amount N_e is plotted against time in days.
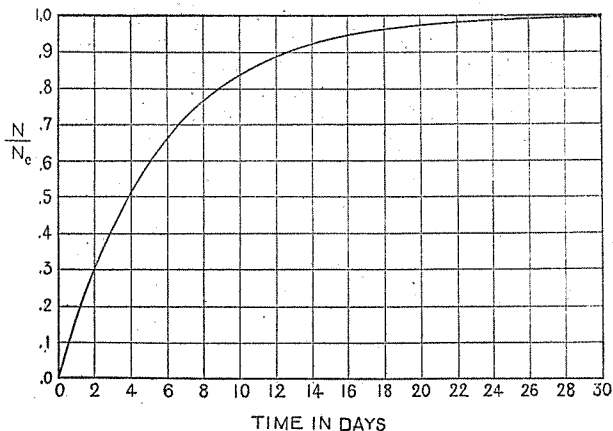

TIME IN DAYS

Fig. 1. The Growth Curve of Radon.

Measurement may be conducted at any time after the considerable amount of radon accumulated, preferably when the gradient of the growth curve of radon becomes low, or when the equilibrium is practically attained. If the measurement is effected in short way of equilibrium, the equilibrium amount of radon may easily be calculated from the equation (3), the time of accumulation of it being known.

In the measurement, the preserved cake of rock substance is heated to re-fusion in a closed vessel in the electric furnace, in order to expel radon from the melt. The reduced pressure over the melt secures the expulsion of radon. Thence the generated gases including radon are sucked into an ionization chamber connected to an electrometer. Any amount of moisture makes the insulation bad in the ionization chamber so that on the way of the transference of radon, the moisture should be removed from the gases by drying agent such as calcium chloride or phosphorus pentoxide. The admitted radon in the ionization chamber transforms unceasingly into radium A which transforms again into radium B and so on. The radioactive rays emitted by these disintegration products of radium ionize the air in the chamber, the effect being measured by means of the electrometer. Here α-ray plays a principal rôle in ionization, β- and γ-rays having relatively weak ionization power. The total ionization due to these three kinds of rays from radon and its disintegration products increases rapidly at first and slower later reaching maximum practically after three hours from the introduction of radon, when the values of ionization current are to be measured.

3. Apparatus and operation

The essential parts of the arrangement are ionization chamber, electrometer and furnace with a combustion tube in which the rock material is to be fused. The whole arrangement is shown in Fig. 2.

The ionization chamber made of brass is of cylindrical form, 14 cm in inside diameter and 19 cm in internal height. The central electrode, a brass rod 3 mm in diameter and 15 cm long, is screwed in the connecting rod which penetrates through the insulating amber plug into the leaf chamber. In this chamber another rod of semicircular section is screwed to the former, with a gold leaf hanging from it. Holding of the leaf is effected by a strip of steel spring instead of pasting against the flat surface of the rod. The gold leaf has a pointer of fine glass fibre attached to an end. This makes
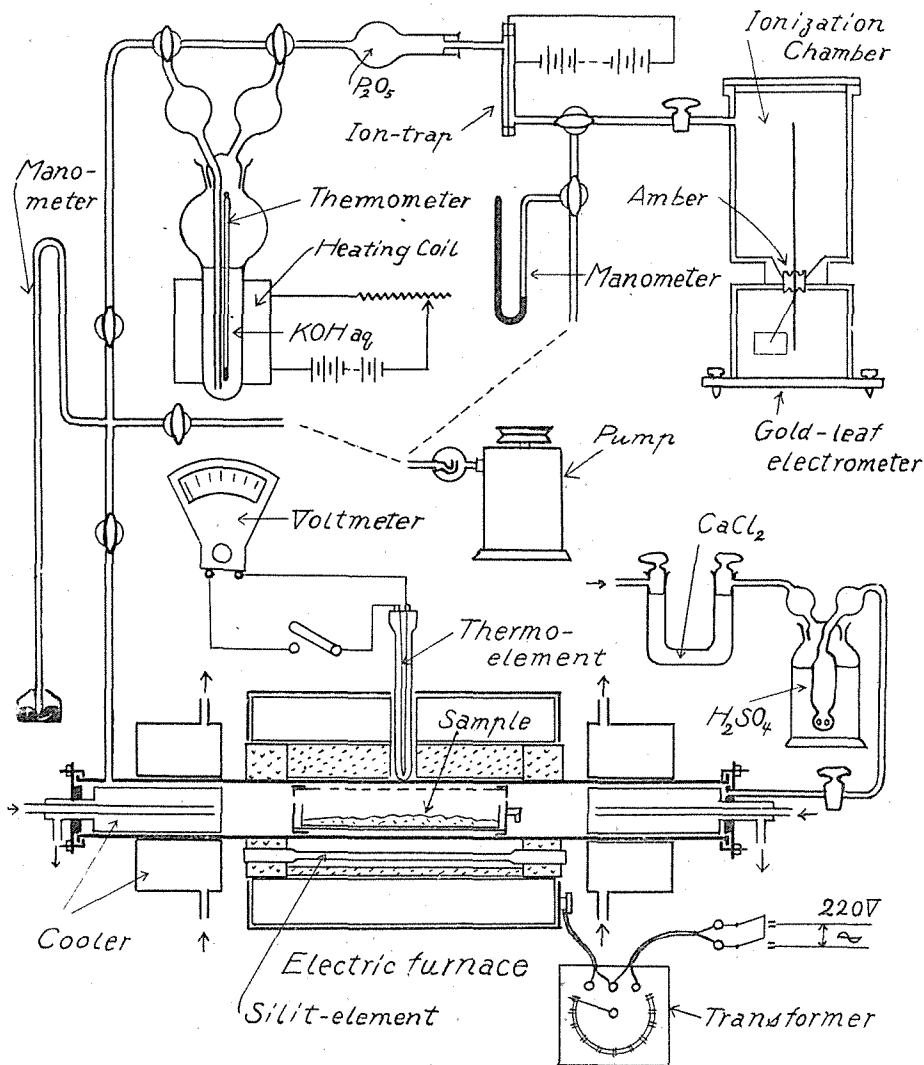
Fig. 2. Arrangement for Fusion Method.

possible the accurate observation to a tenth of a scale division. The insulator is of transparent amber with well polished surface cut in-to a special form for the purpose of minimizing the surface leak. Inside the electrometer there is a small glass vessel containing phosphorus pentoxide which keeps the interior dry.

The fusion of rock material is carried out in an iron boat with a holder of the same material enclosed in a combustion tube of steel instead of such a precious metal as platinum used by JOLY.[2] The

holder not only affords a clue for pulling out the charge from the tube but protects it against the contamination caused by the accidental splash or frothing over of the melt. The combustion tube is 4.5 cm in inside diameter, 1 m long and about 3 mm thick. The use of an iron combustion tube is a suggestion of F. BěHOUNEK[1] and though the oxidation of the outer surface of it near the part heated to a high temperature as far as 1100°C. is somewhat serious, yet it can stand about ten times use. With the intention to diminish oxidation, electro-plating with nickel was attempted, but the result was undiscernible. The either end of the tube is provided with lid having rubber packing. To insure air-tight condition within the tube, its two ends are cooled both inside and outside with water coolers. The function of these arrangements was so satisfactory that even the temperature in the furnace was kept at 1100°C. for a long time, the lids remained cool in spite of slow circulation of water. Near one of the ends is fitted a small side tube through which the escaping gases pass towards the ionization chamber. The lid against the opposite end has also a fine tube which serves as an inlet for the wash-out air current when the main introduction of radon into the ionization chamber is completed. The electric furnace is of a tube type with two "*Silit*" elements of effective length 30 cm heated by the alternating current of 110–220 volts and its maximum capacity is 5 K.W. The characteristic of this furnace is that it can endure the rapid increase of temperature.

On the way of transference to the ionization chamber, the gases including radon are passed through a scrubber, drying tube and ion trap. The scrubber is half filled with concentrated KOH solution which is heated to above 90°C. from outside with heating coil of nichrome wire by electric current. The temperature of the solution is read on the thermometer inside of the scrubbing bottle. The KOH solution absorbs CO_2 and other acidic gases. It must be kept hot during the passage of gases, though if slightly diminishes the absorbing power for CO_2, because the cool aqueous solution appreciably absorbs radon itself. In usual determinations, the generation of CO_2 out of the fused melt is neglisible, if not null, so that the scrubber is put outside of the passage of gases by the by-path; and it is in use when rock powder, instead of cake, is directly fused with flux in the furnace. The running gases are then dried by phosphorus pentoxide before entering the ionization chamber as the moisture seriously damages the insulation. The ion trap serves as an arrester for thermoions and decay products of thoron, if any, which may affect the observation.

The cake of rock material to be tested is prepared more than a month before the fusion. The preparation is as follows: A sample not less than 100 grams is finely pulverized to pass entirely the mesh of which opening is 0.1 mm. 15 grams of the powder is fused in a platinum crucible with 50 grams of natrium carbonate. The resulted melt is cooled and hermetically preserved in a closed vessel. In the re-fusion, air in the combustion tube is pumped out to a pressure of a few millimeters and the current for the furnace is turned on. The fusion of the cake seems to begin at a temperature near 650°C. which was once determined using a special device for ascertaining electrically whether the cake fused or not. The fusion is controlled with the regulating transformer as well as the manometric reading of which sudden fluctuation indicates the violent ejection of gases from the melt. It is sure that the generation of gases is considerably promoted by the reducing pressure. In order to attain the temperature of 1100°C., fifty minutes or thereabout is usually required; more rapid heating should cause the frothing over of the melt. As the pressure of the gas approaches the atmospheric, the cock leading to the ionization chamber is opened and the escaping gases are admitted into the exhausted chamber. When the pressure equilibrium is approached between the ionization chamber and combustion tube, wash-out air is introduced through the combustion tube to sweep away the remaining radon in it into the ionization chamber until the atmospheric pressure is attained in it. During these processes, the gold leaf is kept charged to a proper potential and the reading is taken when the rate of falling of the leaf becomes maximum which is attained after three hours from the admittance of radon. For the considerations on the use of the electrometer the previous paper (*loc. cit.*) is to be referred to.

4. Discussion of the method

The critical study upon the method is described in this paragraph.

(1) *On the re-fusion:* The rock powder is not directly fused with alkali-carbonate which is the usual way followed by others. One of the reasons is that the capacity of the boat in which charge is fused does not allow direct fusion of sufficient amount of rock material for the present sensibility of the electrometer. The direct fusion without flux devised by EVANS[1] is preferable, but it requires a furnace of a special type. The rock powder and alkali-carbonate of three times in weight occupy larger space than the cake resulted

from the same amount of materials, and frothing over, which is probable, accompanied by the generation of CO_2 is quite serious. Another reason is that by this process CO_2-absorber can be eliminated which otherwise affords the serious risk of absorbing radon. From a different view-point the violent frothing attending direct fusion with flux facilitates radon to be set free from the melt. This, however, may be required when fusion is done in the crucible as used by others. As in the present case when it is done in a boat in which melt spreads in a thin layer, this is not always necessary. So far as granite is concerned, the melt has extremely low viscosity and this, as well as the relatively large area exposed to space compared to the volume, favours the escape of radon. It is also conjectured from the traces frequently observed on the boat-holder after the run is over that frothing must have occurred. Thus insufficient expulsion of radon is highly improbable.

(2) *On the effect of fusing flux:* According to JOLY,[5] carbonates used as fusing flux show feeble radioactivity and the correction may amount to as far as ca. 0.13×10^{-12} grams of radium per gram of mixed carbonates (anhydrous carbonates of sodium and potassium taken in equal proportions). In later paper,[6] however, in company with POOLE, he reported: " Although many of them gave null results, yet some seemed to indicate that they might contain small sporadic quantities of radium,....... ." And they dealt with the carbonates de-emanated immediately before the experiment by solution in water followed by evaporation to dryness. PIGGOT's investigation[7] is interesting in which he observed that sodium- and potassium-carbonates possess radioactivity in the ratio of about 1 to 3.3 and resulting mixed flux (in equal parts) gives an electrometer reading corresponding to 0.132×10^{-12} grams radium per gram.

In the present investigation, 100 grams of anhydrous sodium carbonate was fused and resulting cake was tested for radium after proper preservation. The result showed that in some cases there was a slight increase of leak and in another cases a small negative effect. In all cases the effect were very small and if it be real, it may hardly affect the last figure shown in the list.

(3) *On the natural leak:* The charged system of the electrometer reveals the phenomenon of natural leak, gradually losing its charge even when radon is not in the ionization chamber. This phenomenon may be considered to be due to the overlapped effect of back-ground ionization and leakage through the insulator. What-

ever may be the cause, the variation of natural leak obscures the accuracy of determination.   Therefore it is desirous to keep the laboratory in as nearly a constant state as possible.   In the present measurement the natural leak was measured just before each run for at least two hours at an interval of ten minutes and the mean value for the last an hour, when the leak is practically constant, were adopted as the value during the measurement of the effect of radon.   The followings are the results of experiments specially made for the purpose of observing the constancy of the natural leak :

No.	Date.	Leak (L)	$V=(\bar{I}\sim L)$	V^2
1	March 29	0.026 div./min.	0.0007	49×10^{-8}
2	,,	.027	.0017	289 ,,
3	March 30	.025	.0003	9 ,,
4	,,	.025	.0003	9 ,,
5	April 1	.025	.0003	9 ,,
6	,,	.024	.0013	169 ,,
7	April 2	.025	.0003	9 ,,
8	,,	.026	.0007	49 ,.
9	April 8	.024	.0013	169 ,
10	April 9	.026	.0007	49 ,,
	Mean $(\bar{I})=0.0253$			$\Sigma V^2=810\times10^{-8}$

The probable error for one set of observation, computed from $\varepsilon = 0.6745\sqrt{\dfrac{\Sigma V^2}{n-1}}$, is 0.00064 div./min. which corresponds to 0.16×10^{-12} grams of radium.   As 15 grams of rock powder were used in each radium determination, the uncertainty for it is 0.01×10^{-12} grams of radium per gram of rock.

(4)   *On the transference of radon :*   The generated gases including radon are received in the ionization chamber which is evacuated in advance and as the equilibrium of pressure is nearly established between the chamber and combustion tube, outer air is introduced to sweep the remaining radon in the latter into the former.   Let us estimate how much gases are transferred before the pressure-equilibrium is attained in the operation.

Let the conditions in the combustion tube just before the transference be : V_b, P_1, T_1 and those in the ionization chamber at the time of equilibrium be : V_a, P_2, T_2, in which V is the volume, P the pressure and T the temperature of the gases.   If the volume of gases transferred before the equilibrium is established is V_t calculated for the pressure P_t, we have the following relations ;

$$P_1 V_1 = P_2 V_a \{1 + \beta(T_1 - T_2)\} \qquad \dots\dots\dots\dots\dots(1)$$

$$P_1(V_b - V_1) = P_2 V_b \qquad \dots\dots\dots\dots\dots\dots(2)$$

where β is the pressure coefficient for gases and is equal to 0.37×10^{-2} for air, oxygen, H_2O vapour, CO_2 etc. Eliminating P_2,

$$\frac{V_b}{V_1} = \frac{V_b}{V_a} \cdot \frac{1}{1 + \beta(T_1 - T_2)} + 1.$$

In this equation V_a is 2950 c.c., the capacity of the ionization chamber and V_b is the effective volume within the combustion tube and cal-culated to be 793 c.c. For simplicity take $T_1 - T_2 = 1000°C.$, which was nearly the actual case, we have

$$\frac{V_b}{V_1} = \frac{793}{2950} \cdot \frac{1}{1 + 0.37 \times 10^{-2} \times 1000} + 1$$

or $\qquad \dfrac{V_1}{V_b} = 0.946 \qquad \dots\dots\dots\dots\dots\dots(3)$

Thus 95% of the gases enter the ionization chamber before the equilibrium of pressure is reached. If the distribution of radon in the combustion tube is uniform, this proportion of radon is conveyed into the ionization chamber leaving the remainder for the transference by the sweeping air. This assumption seems to contain some diffi-culties, yet it is obvious that the intervening of CO_2-absorber and drying agent much raises the efficiency of transference of radon.

(5) *On the influence of thorium series:* One gram of average granite is so far reported to contain 9.0×10^{-6} grams of uranium (viz. 3.1×10^{-12} grams of radium) and 20.0×10^{-5} grams of thorium.[8] This value of uranium content is too high compared to my results obtained for rocks in Nippon, but as only the ratio of uranium to thorium comes into the present interpretation, the above figures will correctly be adopted. Denote the number of radon and thoron atoms contained in equilibrium in 1 gram of granite by N_{Rn} and N_{Tn} re-spectively. Their values will be given by

$$N_{Rn} = \frac{6.06 \times 10^{23}}{226} \cdot \frac{\lambda_{Ra}}{\lambda_{Rn}} \times 3.1 \times 10^{-12},$$

$$N_{Tn} = \frac{6.06 \times 10^{23}}{232} \cdot \frac{\lambda_{Th}}{\lambda_{Tn}} \times 20.0 \times 10^{-5},$$

where $\qquad 6.06 \times 10^{23}$: AVOGADRO's number,
$\qquad$ 226 and 232: atomic weight of radium and thorium,
$\qquad \lambda_{Ra} = 1.373 \times 10^{-11} \text{sec.}^{-1}, \qquad \lambda_{Th} = 1.33 \times 10^{-18} \text{sec.}^{-1},$
$\qquad \lambda_{Rn} = 2.097 \times 10^{-6} \text{sec.}^{-1}, \qquad \lambda_{Tn} = 1.27 \times 10^{-2} \text{sec.}^{-1},$

Most of thoron expelled from the melt will die out during the heating as the life of thoron is so short that in each 54.5 sec. half of the existing number of atoms disintegrates. The rate of generation of thoron, however, is rather rapid and in consequence a part of thoron which escape disintegration during the transference may enter the ionization chamber. Though we can not estimate how much thoron atoms reach there, let us assume for safety that the equilibrium quantity of thoron is introduced. The initial intensity of ionization due to α-particles from radon and thoron will be, respectively,

$$I_0^{Rn} = \lambda_{Rn} N_{Rn} k_{Rn},$$
$$I_0^{Tn} = \lambda_{Tn} N_{Tn} k_{Tn},$$

where the k's are the number of pairs of ions produced by one α-particle, and have the values

$$k_{Rn} = 1.55 \times 10^5,$$
$$k_{Tn} = 1.78 \times 10^5.$$

The ionizing intensity of radon and its disintegration products after θ hours from the time of entrance of radon is,

$$I_\theta^{Rn} = I_0^{Rn} / f_\theta$$

in which f_θ is a reduction factor and is 0.460 for $\theta = 3$ hours.[9]

To obtain the intensity of ionization due to active deposits from thoron, suppose that the matter initially considered is thoron only and then it disintegrates into ThA, ThB, ThC etc. successively, of which number of particles after any time t are $P, Q, R, S, \ldots\ldots$ respectively. Then if the number of particles of thoron initially present be P_0, by the theory of successive transformations we have;

$$P = P_0 e^{-\lambda_1 t} \quad \ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots(1)$$

$$Q = \frac{P_0 \lambda_1}{\lambda_2 - \lambda_1} (e^{-\lambda_1 t} - e^{-\lambda_2 t}) \quad \ldots\ldots\ldots\ldots\ldots\ldots(2)$$

$$R = P_0 (a e^{-\lambda_1 t} + b e^{-\lambda_2 t} + c e^{-\lambda_3 t}) \quad \ldots\ldots\ldots\ldots(3)$$

where $\quad a = \dfrac{\lambda_1 \lambda_2}{(\lambda_2 - \lambda_1)(\lambda_3 - \lambda_1)}, \quad b = \dfrac{\lambda_1 \lambda_2}{(\lambda_1 - \lambda_2)(\lambda_3 - \lambda_2)}, \quad c = \dfrac{\lambda_1 \lambda_2}{(\lambda_1 - \lambda_3)(\lambda_2 - \lambda_3)}.$

$$S = P_0 (a e^{-\lambda_1 t} + b e^{-\lambda_2 t} + c e^{-\lambda_3 t} + d e^{-\lambda_4 t}) \quad \ldots\ldots\ldots(4)$$

where $\quad a = \dfrac{\lambda_1 \lambda_2 \lambda_3}{(\lambda_2 - \lambda_1)(\lambda_3 - \lambda_1)(\lambda_4 - \lambda_1)}, \quad b = \dfrac{\lambda_1 \lambda_2 \lambda_3}{(\lambda_1 - \lambda_2)(\lambda_3 - \lambda_2)(\lambda_4 - \lambda_2)},$

$$c = \frac{\lambda_1 \lambda_2 \lambda_3}{(\lambda_1 - \lambda_3)(\lambda_2 - \lambda_3)(\lambda_4 - \lambda_3)}, \quad d = \frac{\lambda_1 \lambda_2 \lambda_3}{(\lambda_1 - \lambda_4)(\lambda_2 - \lambda_4)(\lambda_3 - \lambda_4)}.$$
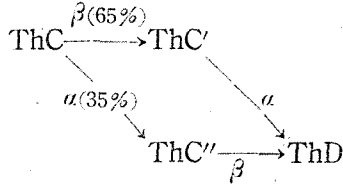
Now,

$$P(\text{Tn}): \quad \lambda_1 = 1.27 \times 10^{-2} \quad \lambda_1 t = 1.37 \times 10^2 \quad e^{-\lambda_1 t} \doteqdot 0$$
$$Q(\text{ThA}): \lambda_2 = 4.78 \quad \lambda_2 t = 5.16 \times 10^4 \quad e^{-\lambda_2 t} \doteqdot 0$$
$$R(\text{ThB}): \lambda_3 = 1.82 \times 10^{-5} \quad \lambda_3 t = 1.97 \times 10^{-1} \quad e^{-\lambda_3 t} = 0.822$$
$$S(\text{ThC}): \lambda_4 = 1.91 \times 10^{-4} \quad \lambda_4 t = 2.06 \quad e^{-\lambda_4 t} = 0.144$$

$$\dots(5)$$

where $t = 3$ hours or 10800 sec.
Putting these values into (1), (2), (3) and (4), we have

Tn : $P = 0$

ThA : $Q = 0$

ThB : $R = 0.824 P_0$

ThC : $S = 0.071 P_0$

$$\text{ThC} \xrightarrow{\beta(65\%)} \text{ThC}'$$
$$\alpha(35\%) \qquad \alpha$$
$$\text{ThC}'' \xrightarrow{\beta} \text{ThD}$$

Since ThB and ThC constitutes about 90% of the whole number of atoms, the remainder will be products of ThC. ThC disintegrates in two ways as shown in the above diagram. Since ThC' has very short life (half period T=ca. 10^{-11}sec.), the changes through ThC' may be regarded for practical purpose as that 65% of ThC disintegrates directly into ThD with emission of α-particle. Taking the number of Tn atom initially present as 100 and neglecting the effect due to β-ray owing to its weakness, the ratio of initial activity of thorium series to activity after three hours may approximately be calculated as follows :

$$\frac{I_{3h}}{I_0} = \frac{7.1 \times 0.65 \times \lambda_{ThC} \times k_{ThC'} + 7.1 \times 0.35 \times \lambda_{ThC} \times k_{ThC}}{100 \times \lambda_{Tn} \times k_{Tn}}$$
$$= 1.35 \times 10^{-3}$$

where $k_{Tn} = 1.78 \times 10^5$; $k_{ThC} = 1.71 \times 10^5$; $k_{ThC'} = 2.54 \times 10^5$.
Thus we obtain,

$$I_{3h}^{Tn} \doteqdot I_0^{Tn} \times 1.35 \times 10^{-3}.$$

Hence finally

$$\frac{I_{3h}^{Tn}}{I_{3h}^{Rn}} = \frac{I_0^{Tn} \times 1.35 \times 10^{-3}}{I_0^{Rn}/f_{3h}}$$
$$= \frac{\lambda_{Tn} N_{Tn} k_{Tn} \times 1.35 \times 10^{-3}}{\lambda_{Rn} N_{Rn} k_{Rn}/f_{3h}}$$
$$= \frac{1.27 \times 10^{-2} \cdot 54.7 \cdot 1.78 \times 10^5 \cdot 1.35 \times 10^{-3}}{2.097 \times 10^{-6} \cdot 5.44 \times 10^1 \cdot 1.55 \times 10^5/0.460}$$
$$= 4.34 \times 10^{-3}.$$

Thus the effect of thorium series is neglisible in the present investigations where the experimental error is a few percents.

5. Calibration

Radium solution used as the standard was the same used in the solution method and denoted as the normal solution A in the previous paper (*loc. cit.*). This solution contains 1.48×10^{-11} grams of radium in one cubic centimeter. A known quantity of this solution was taken with the utmost care from a burette in a small glass bulb of about 2 c.c. This is slowly evaporated to dryness by immersing the bulb in a warm sand-bath, and then sealed up and left aside for more than a month. The calibration was made in two ways:

(1) When radium and radon come to an equilibrium, the standard bulb was fused with 10 grams of Na_2CO_3, and the run was carried out in the usual way. The results of three determinations were as follows:

	I	II	III
Volume of solution in c.c.	1.00	2.02	2.00
Rn-content in 10^{-11} curies	1.48	2.99	2.96
Electrometer leak in div./min.	0.036	0.149	0.144
Natural leak in div./min.	0.026	0.029	0.023
Leak due to radon in div./min.	0.060	0.120	0.121
Leak for 10^{-11} curie	0.040_5	0.040_1	0.040_0

Thus the mean leak due to 10^{-11} curie of radon is 0.0405 div./min.

(2) A standard bulb added to the mixture of granite powder and sodium carbonate was fused in a platinum crucible in a usual way. The produced cake was examined after thirty days for its radon content and the result corrected for the radioactivity of the granite gives the calibration factor. The granite selected for this purpose came from Senge Quarry in Syôdo-sima and exactly 15 grams of the sample were used in each determination. The results for granite only were:

No. of the sample	G-14-1	G-14-2	G-14-3
Total leak	0.070	0.071	0.069
Natural leak	0.023	0.025	0.024
Leak due to radon	0.047	0.046	0.045

Thus the mean leak due to radon in equilibrium with radium in 15 grams of the granite is 0.046 divisions per minute. When mixture of the granite with the standard material was examined, the results were as follows:

	I	II	III
Total leak	0.131	0.132	0.187
Natural leak	0.025	0.026	0.024
Leak due to radon	0.106	0.106	0.163
Effect of granite	0.046	0.046	0.046
Leak due to the standard	0.060	0.060	0.117
Ra-content in the standard in 10^{-11} curie	1.46	1.48	2.94
Leak for 10^{-11} curie	0.041_1	0.040_5	0.039_8

Thus 10^{-11} curie of radon causes leak of 0.0405 div./min. in average, which agrees with the result given above. This figure corresponds to 24.69×10^{-11} curies for one division per minute fall of the electrometer leaf and is tolerably close to the value obtained by the solution method. (*loc. cit.*). This figure will be used throughout this paper.

6. Results of determinations

Granites were collected from twenty different localities in southwestern part of Nippon excluding Kyûsyû. Most of them were taken at quarries where relatively fresh samples are easily obtained. So far twenty samples have been carefully examined by the fusion method above described, two or three determinations being repeated for each sample. Their radium contents thus determined are given in the 6th column in Table I, the number being expressed in 10^{-12} gram per gram of rock. The abbreviations T.L., N.L. and L.R. in the table are used to denote the total leak, the natural leak and the leak due to radon.

Table I.

Sample	T.L.	N.L.	L.R.	mean	Ra	SiO$_2$ %	Locality	Reference
G-20	0.254	0.020	0.234	0.220	3.62	79.26	Sekigane, Tottori	Apilitic granite. Biotite associates in small quantities. Sample pretty decomposed.
	0.241	0.022	0.219					
G-11	0.178	0.015	0.163	0.165	2.72	76.07	Inusima, Okayama	Biotite granite. Pleochroic halo with zircon nucleus often found.
	0.195	0.028	0.167					
G-4	0.168	0.024	0.144	0.142	2.34	77.78	Ôi, Gihu	Biotite granite of porphyritic texture. Pleochroic halo conspicuous.
	0.165	0.024	0.140					
G-7	0.145	0.020	0.125	0.124	2.04	75.32	Takeda, Hyôgo	Granite with micrographic texture, biotite altered to chlorite and magnetite.
	0.153	0.030	0.123					

Table I. (Continued)

Sample	T.L.	N.L.	L.R.	mean	Ra	SiO₂ %	Locality	Reference
G-6	0.144 0.143	0.024 0.023	0.120 0.120	0.120	1.98	70.77	Kasagi, Kyôto	Fine-grained biotite granite. Accessories are zircon, apatite and muscovite. Halo with large nucleus very common.
G-12	0.151 0.143	0.030 0.023	0.121 0.119	0.120	1.89	75.37	Siraisizima, Okayama	Biotite granite. Quartz idiomorphic. Pleochroic halo frequent though biotite rare.
G-18	0.154 0.133	0.032 0.026	0.122 0.107	0.115	1.89	73.33	Mukaisima-higasi, Hirosima	Somewhat porphyritic in texture. Fine fragments of biotite apt to gather.
G-13	0.107 0.110	0.020 0.025	0.087 0.085	0.086	1.42	72.76	Mannari, Okayama	Hornblende biotite granite with orthoclase of salmon pink colour.
G-17	0.101 0.099	0.023 0.025	0.077 0.074	0.076	1.25	73.08	Ôsima, Ehime	Fine-grained hornblende biotite granite with apatite as predominant accessory mineral.
G-10	0.093 0.104	0.020 0.028	0.073 0.076	0.075	1.23	76.76	Tangazima, Hyôgo	Hornblende biotite granite. Miarolitic structure characteristic.
G-19	0.090 0.096	0.020 0.026	0.070 0.070	0.070	1.15	73.39	Ôtuzima, Yamaguti	Hornblende biotite granite of porphyritic texture. Hornblende rare.
G-1	0.092 0.098	0.028 0.030	0.064 0.068	0.066	1.09	76.28	Midono, Nagano	Biotite granite partly porphyritic. Zircon with halo common. Other accessories titanite and magnetite.
G-16	0.087 0.087	0.024 0.025	0.063 0.062	0.063	1.04	72.85	Hirosima, Kagawa	Fine-grained biotite granite rarely containing hornblende. Pleochroic halo hardly found.
G-2	0.089 0.093	0.028 0.030	0.061 0.063	0.062	1.02	72.85	Huzioka, Aiti	Biotite granite of rather porphyritic texture. Quartz idiomorphic and has tendency to form mosaic texture.
G-15	0.078 0.073	0.028 0.023	0.050 0.050	0.050	0.82	75.25	Kose, Syôdosima, Kagawa	Biotite granite of light pink colour. Halo hardly found and titanite as accessory mineral.
G-5	0.068 0.071	0.020 0.026	0.048 0.045	0.047	0.77	69.93	Kasagi, Kyôto	Ore(pyrite?)-bearing granite rich in biotite and hornblende.
G-14	0.070 0.071 0.069	0.023 0.025 0.024	0.047 0.046 0.045	0.046	0.76	73.79	Senge, Syôdosima, Kagawa	Biotite granite, quartz idiomorphic.
G-3	0.067 0.070	0.022 0.025	0.045 0.045	0.045	0.74	69.26	Mihune, Sanage, Aiti	Hornblende biotite granite.
G-8	0.064 0.069	0.022 0.026	0.042 0.043	0.043	0.71	72.62	Sumiyosi, Hyôgo	Hornblende biotite granite with pinkish orthoclase. Magnetite abundant as inclusion in mica.
G-9	0.062 0.059	0.025 0.020	0.037 0.039	0.038	0.63	68.51	Nunobiki, Kôbe, Hyôgo	Hornblende granite.

7. Discussions

For the purpose of investigating the relation between radium and silica contents, the latter was also determined by careful analyses carried out after the usual method and precautions described by H. S. WASHINGTON.[10] The results are also given in the 7th column in Table I and are also plotted in Fig. 3 by crosses, and the curve I is drawn to represent them.
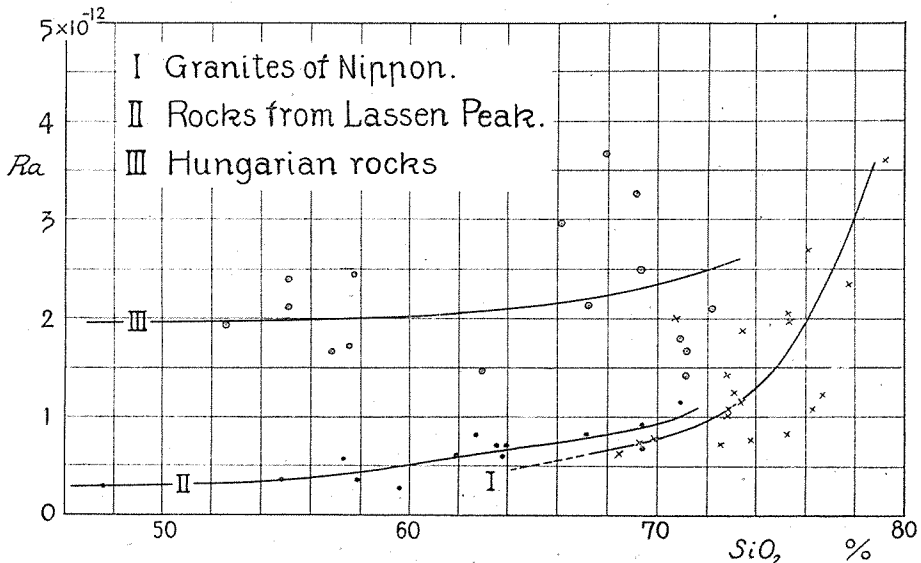

Fig. 3. Silica Percentage and Radium Content in Igneous Rocks.

Here we see the general tendency that radium content increases with increasing silica percentage. The sample from Kasagi (G-6) shows considerable deviation from the normal radium-silica relation. It is noticed that this sample is abnormally rich in conspicuous pleochroic halo with large nucleus.

For more detailed investigation, I have collected the data of radium contents and silica percentages of rocks from Lassen Peak, Cal., U. S. A.[11] and of Hungarian rocks.[12] These values are also plotted on the silica base in Fig. 3. Here we also see that the said relation roughly holds for rocks of the each province separately, but not commonly for all the rocks. Rocks from Lassen Peak show slight tendency to be richer in radium than those of Nippon but are far poorer than Hungarian rocks for the same percentage of silica.

Now when variation-curves are drawn for CaO and Na₂O + K₂O on a silica base, they always intersect each other. The value of
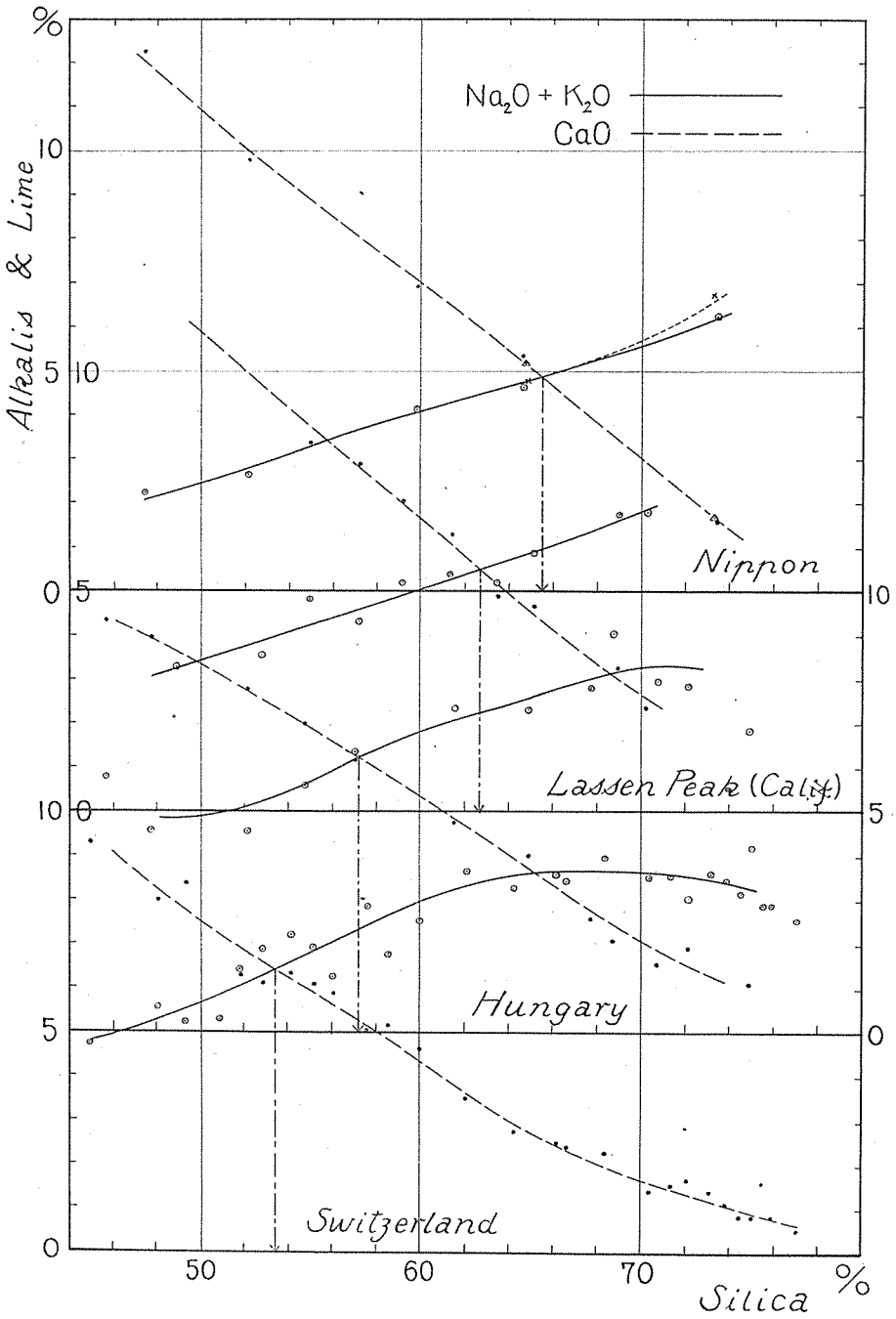
Fig. 4.   The Alkali-lime Curves.

silica percentage corresponding to this point of intersection is the
"alkali-lime index" as M. A. PEACOCK[13] calls it.   The variation-
curves for effusive rocks of Nippon were established by S. YAMADA[14]
using 136 analyses and the alkali-lime index was found to be 65.5.
Introduction of data (40 analyses) of plutonic rocks[15] slightly modifies
the form of the curve for alkali as is shown by dotted line in Fig.
4.   This, however, changes the alkali-lime index in no appreciable
amount.   For rocks in other localities, I used the data of chemical
analyses given by H. S. WASHINGTON[16] and by R. D. EVANS (*loc. cit.*)
for the Lassen Peak rocks and those of DE FINÁLY (*loc. cit.*) for the
Hungarian rocks.   From these data, the variation-curves were drawn
and the alkali-lime indices were found to be 62.6 and 57.2 respectively.
PEACOCK (*loc. cit.*) gives a figure of 62.4 as the alkali-lime index of
the former, which agrees with my figure very nearly.   Comparing
the values of the alkali-lime indices thus obtained for the rocks of
Nippon and Lassen Peak, Cal., and the Hungarian rocks, we might
notice that they are in reverse order of magnitude as the radium
content for the same silica percentage.   The data for rocks from
Switzerland are also given by WASHINGTON (*loc. cit.*), from which the
alkali-lime index was found to be the lowest of all.   On the other
hand we have the fact that rocks from St. Gotthard[17] and Aarmassif[18]
are highly radioactive.   Thus the rocks from Switzerland seem to
follow the same tendency.   In conclusion, it may be suggested that
radium contents of the rock-series in different petrographical pro-
vinces vary antipathetically with alkali-lime indices.

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