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Variations in Radioactivity and Chemical Elements across Igneous Contacts

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Abstract

The contacts resulting from the intrusion of granitic material into various rocks show large variations in chemical composition and alpha-activity along a traverse normal to the contact boundary. Studies of these variations by the analyses of closely spaced samples using semi-quantitative spectrographical method and by measurements of beta-activity has been described. The results of the analyses indicate that the major and minor rock compositions studied are mobile when moderate temperature- and concentration-gradients are present. Migration of materials is believed to have taken place late in the cooling history by diffusion along mineral boundaries.

The chemical variations found may be naturally considered to have some relations to the original compositions of the wall rock and intruding materials, and to the cooling conditions of them. The alpha-activity variations may be related to the difference in temperature of intruding material and of the wall rock. Generally smooth and continuous variations in alpha-activity have been found occurred across contacts between intrusives and wall rocks of different original compositions, which have been held at high temperature. Highly irregular variations have been found when the intrusive and wall rock were compositionally different and steep thermal gradient existed. But no remarkable variations in beta-activity distribution have been found across contacts in spite of the large variations in alpha-activity distributions.

Introduction

Several years ago Z. HATUDA projected the study of examining radioactivity distribution near the igneous contact with a view to finding diffusion features of elements afforded by intruded magma. On commencing his sutdy, he noticed that the radioactivity revealed characteristic variations within the granitic intrusive itself, which was then rather an unexpected result, while the radioactivity distribution within the wall rock showed less amplitude in variation. The writer had taken part in the former's investigation since 1955, and have executed the measurements of radioactivity on samples more than 6800 in number from 167 sites of granite contacts, by means of a sensitive radioscope specially designed, the localities of which cover Honsyû, Shikoku and Kyûsyû in Japan.

The radioactivity profiles thus obtained across igneous contacts showed a rather definite tendency of being reduciable to five types which are presumably corresponding to the conditions under which the contact phenomena took place.

As reported in the third paper, comparison of the two curves was made, one showing the alpha-activity and the other the beta-activity along a traverse as normal as possible to the plane of contact, but result was not so clear as expected, owing perhaps to insufficiency of sensitivity of the apparatus then used for measuring beta-activity.

In the present study the above difficulty has been overcome by the use of low background radiation counter, and spectrographical as well as petrographical investigations were made to yield many interesting and suggestive results which will be shown in the following.

Semi-quantitative spectrographical analyses

In order to clarify the distribution of elements across igneous contacts of which description on the sample has been made in the preceding paper[3], semi-quantitative spectrographical analyses was tried by using 3.4 m Plane Grating Spectrograph and Microphotometer manufactured by the Jarrel Ash Co. U.S.A.

(1) *Procedure*

Electrode System: The electrode is highly pure carbon rod (special electrode furnished by the Japan Carbon Co.). The lower sample-carrying electrode is about 0.6 cm in diameter and about 6 cm long, with a cup at the top 0.3 mm in diameter and 0.7 cm deep. Each cup is filled level with sample pulverized to pass 270 meshes. The upper counter electrode is pointed at the lower end. The holders of the both electrodes are water-cooled.

The arc gap width is maintained at 3 mm.

Excitation: The samples are arced in the direct current arc at an initial current of 7 amperes. After initial setting, no further adjustments of current are made. The spectrograph is taken in the following conditions.

Exposure Conditions:

Spectral region	2400–4600 Å (2 nd order)
Slit width	20 μ
Exposure period	55 sec.

Peotographic Procedure:

Development	Kodack developer D–19, 5 minutes at 20°C
Fixing	Kodack fixer

Measurement of Intensity: For this analyses, the measurement of the intensity of spectro-lines is done by comparison with standard spectro-lines of Fe, with the aid of microphotometer of the Jarrel Ash Co.

The lines used for analyses are listed in Table I.

Table I Wave lengths of analytical lines used.

Element to be determined	Wave length, in Å	Element to be determined	Wave length, in Å
Al	3028.16	P	2534.01
	3092.71		2535.65
As	2780.20		2553.28
Au	2802.19	Pb	2833.28
B	2496.78	Si	2881.58
	2497.73	Sn	3034.12
			3009.15
Da	3158.87		2863.33
	3179.33		3175.02
Co	3405.12	Th	3290.59
			3601.21
Cu	3273.96	Ti	3349.04
	3247.54		3361.21
Fe	3091.58	V	3110.71
Hf	3072.88		3118.38
	2904.41		3125.28
	2820.22		3138.48
	2641.41		3183.98
			3185.40
Mg	2852.13		
	2802.70	Zn	3282.33
			3345.02
Na	3302.32	Yt	3243.28
	3302.99		
		Yb	3289.37
K	3046.72		
	3447.70	Zr	3391.98

(2) *Samples*

For two of the typical traverses across igneous contacts which had been proved to be extremely different in radioactivity distribution, measurements were made on the samples taken at regular intervals of 10 cm as exposure allowed over the whole range of sampling. The localities of the traverses are Kasagi (Koya), Kyoto Pref. and Shishitobi, Shiga Pref.

The radioactivity measurement of these samples with the sensitive radioscope with Lauritsen-element, designed by Z. HATUDA, was already described in the third paper[3].

Another traverses across igneous contact has been also measured on the samples taken from Takamizu, Yamaguchi Pref. The way of sampling rock specimen was such that the nearer the contact boundary was, the closer were the intervals between two ajacent sites of sampling. Each specimen, about 2 grams, taken from 200 grams or more, was pulverized to the fineness to pass through 270 meshes.

(3) *Experiment*

This procedure has been applied with good success to a long list of various

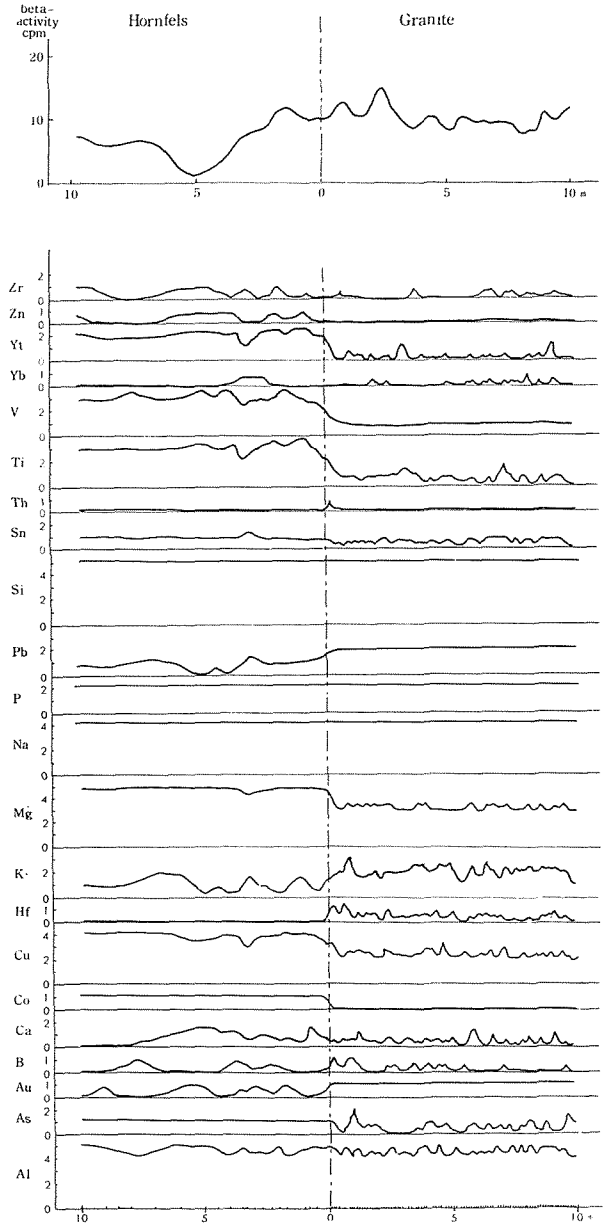

Fig. 1. Beta-radioactivity profile (the top figure) and chemical element distributions with practically continuous samples from Shishitobi, Shiga Pref. The ordinate being intensity of elements.

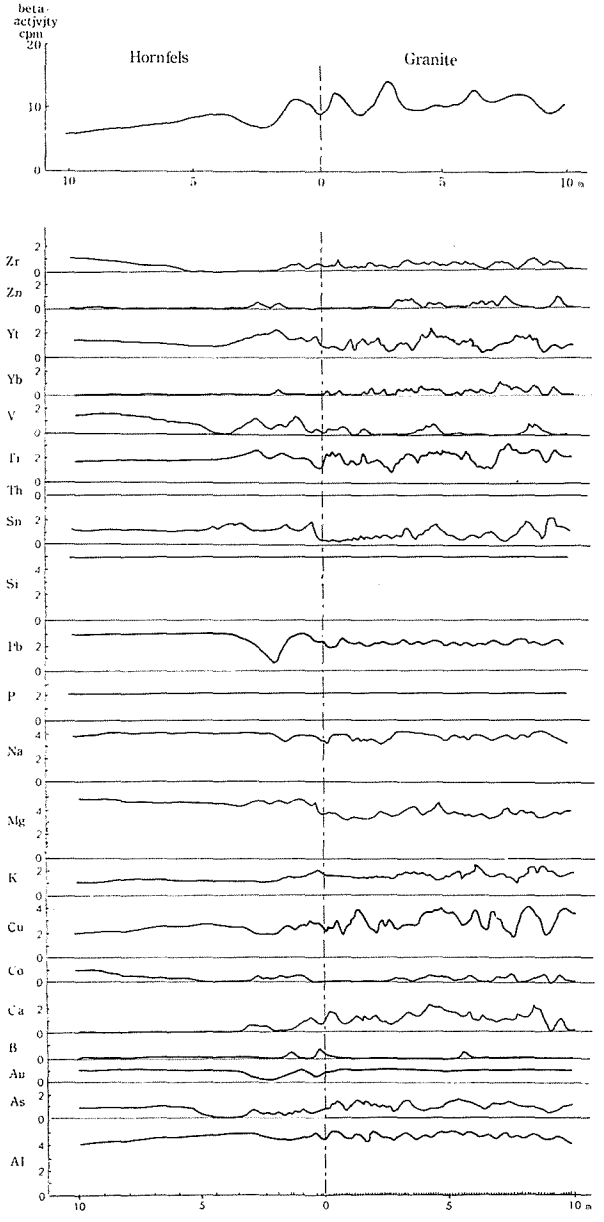

Fig. 2. Beta-radioactivity profile (top figure) and chemical element distribution with practically contineous samples from Koya, Kyoto Pref. The ordinate being intensity of elements.

materials composed of the common elements. The critical point of the procedure is the preparation of samples. Reproducibility or precision for a single measurement of a sample is of the order ±5 to ±7% of the amount present for most elements. The accuracy, however, has not been generally established because of the varieties and numbers of the samples for analyses.

Fig. 1, 2 and 3 show the results obtained in the determination. The results were shown in the following styles:

5 *Strong, strong.*
4 *Strong.*
3 *Moderate.*
2 *Weak.*
1 *Weak, weak.*
0 *Not detectable.*

(4) *Experimental Results*

The specimens from traverses chosen across the contacts in the western part of Tanakami-, Koya- and Takamizu-intrusives were submitted to spectrographical determination. Values obtained are given in Fig. 1–3. Under the above mentioned conditions, the tendencies revealed in the results are that (1) in the Tanakami traverse, the boundary are remarkable in distribution elements, but in the Takamizu and Koya traverses the boundaries are rather vague, that (2) across that contact of Tanakami granite, the increase of volatile matter within the invading rock with decreasing distance from the boundary is conspicuous, but across the contacts of Koya and Takamizu granites, the distributions of volatile matters are obscure, and that (3) notwithstanding the bad condition for analysis of Th, the spectra of Th is recognized at 20 cm from the boundary within granite at Tanakami; the position being correspond to the marked high of alpha-radioactivity.

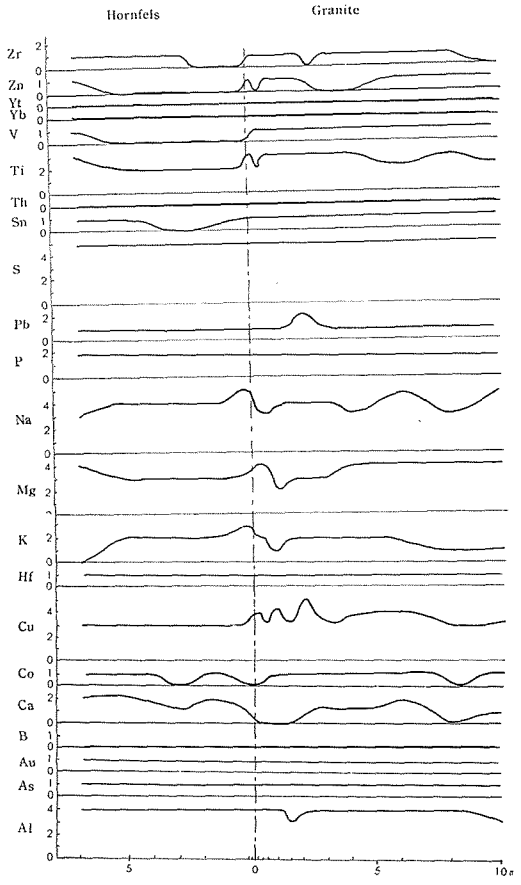

Fig. 3. Chemical element distributions with samples from Takamizu, Yamaguchi Pref. The ordinate being intensity of elements.

The variation of each element along the different traverses is as follows:

Across igneous contact of each granite, there are no marked change in the quantities of Si, Al, Zr and P. In the Tanakami traverse, the variations of quantities of As, Co, Cu, Mg, Ti, Yt, Zn and V are remarkable at the boundary, and on the other hand, in the Koya traverse, those of elements, except Sn and Mg are imperceptible. In the Koya traverse, Ca and As gradually gain in the amount 3–5 meters this side of the boundary in hornfels. The variations of quantities of Na, Ca and Cu are remarkable in the Tanakami traverse, but unremarkable in the Koya one, but as to Au, B, K and Ti, the relation is reversed. B, Th and Hf are found in abundance in the samples of high intensity of alpha-activity, and K, B and Ca in those of high beta-activily. The samples of low beta-activity reveal smaller quantity of Pb and greater one of Zn than average. Regardless of the variations of quantities of Sn, Cu, Ti and Zr, they seem to have no relation to radioactivity. The granite of each locality shows the individuality in a characteristic found in the spectrochemical-lines of B, Ca, Yb, Yt and Zr.

Measurement of beta-activity

On the samples subjected to spectrographical analyses, the measurements of beta-activity were made with a low-background gas-flow counter (**LBC**–1), manufactured by Nihon Musen Co. As an alpha-ray absorber, six sheets of aluminium foil of 1.6 mg/cm^2 were used. Gamma-rays from outside would not be counted by this counter, because in counting, the instrument was surrounded by an iron and mercury shield, as well as a ring of G–M counters surrounding the main gas-flow counter provided with an anticoincidence device[4] to remove the soft component of the cosmic rays and local gamma-ray contamination. These devices reduce the background of the counter nearly 400 counts per minute without such a device to 1 to 0.8 count per minute according to the explanatory note.

The results are shown in Figs. 1–2. Fig. 1 shows the results along a traverse across contact of Tanakami granite at Shishitobi, in Shiga Pref., Fig. 2 those along a traverse across contact of "Ryoke" granite at Watsuka, in Kyoto Pref.

Referring to these figures, in spite of the fact that the variations of alpha-activity across igneous contacts are quite different[3], those of beta-activity have much in common. This may be explained as follows:

Radioactivity families or nuclides	Contents in usual granite ppm	Decay constant sec^{-1}	Beta-ray emmission from a nuclei in its life
U^{238} family (UI)	3.0	4.86×10^{-18}	6
U^{235} family (AcU)	2.1×10^{-2}	3.11×10^{-17}	4
Th^{232} family	13	1.58×10^{-18}	4
K^{40} (beta-decay nuclei)	310	1.54×10^{-17}	1
Rb^{87}	negligible.		
Sm^{147}	negligible.		

As λ_{UI} is a fraction of *UI* transformed per second; taking UI be the amount of *UI* in granite in gm/gm and W_{UI} the atomic weight of *UI*, and assuming all members of *UI* are in equilibrium, the emissions of beta-particles per gram per second are

$$N = 6 \times 6.06 \times 10^{23} \times UI/W_{UI} = 0.22$$

Similarly, for the *AcU*, *Th* and *K*, the emissions of beta-particles per gram per sec. are 0.008, 0.21 and 0.74 respectively. As the effect of absorption of beta-ray by aluminium foil and samples in the present case is little worth consideration. Then, it may be thought that the variations of potassium content are reflected in those of beta-activity, and the variations of *U* and *Th* contents mainly in those of alpha-activity.

Petrographic modal analysis

On the same samples subjected to spectrographical analyses and measurements of beta-activity, petrographical modal analysis was made with the rock section.

In the variations of mode found in the rocks across Tanakami and Koya contacts, the tendencies revealed are that (a) of a series of samples, those which are predominant in quartz show high alpha-activity and those in orthoclase have medium and those in plagioclase have low activity, (b) the samples of which alpha-activity exceeds 1.00 div/min are rich in halos contained in biotite, and are of large grain-size in usual, but in samples with alpha-activity less than 0.25 div/min halos are hardly recognized, (c) the samples having high beta-activity are found abundant in orthoclase and especially in quartz, and that (d) across Koya contact, the chemical composition of plagioclase is richer in An-component than that across Tanakami contacts.

According to GORAI[5], granitic rocks can be classified into two groups with respect to the mode of plagioclase twinning, that is;

1. I-group that includes granitic rocks in which mode of plagioclase twinning is igneous (a fair amount of "C"-twin).

2. M-group that includes granitic rocks in which mode of plagioclase twinning is metamorphic (rare amount of "C"-twin).

And it may be permissible to infer the granites of I-group mentioned above are magmatic origin and those of M-group being the products of some metamorphic processes.

The plagioclase of Tanakami granite exhibits a fair amount of "C"-twin, while the plagioclase of Koya granite rarely shows this type of twinning. Taking GOARI's conception into consideration, Tanakami granite may be said to be magmatic rock and Koya granite recrystallized rock.

Relations between the types of radioactivity across igneous contacts and constituents of granitic rock from unaffected part

Distribution of alpha-activity along traverses across contact boundaries has been investigated by means of the radioscope, with a Lauritsen element designed specially for the pulverized samples of feeble radioactivity such as rocks and minerals[1]. By the results obtained for 167 sites of contact, the profile curves showing the alpha-activity distribution across contacts were found to be classifiable into several types[1)2)3)]. In Fig. 4, the curves representative of each type are schematically drawn, and in Table II, the localities corresponding to the types are shown.

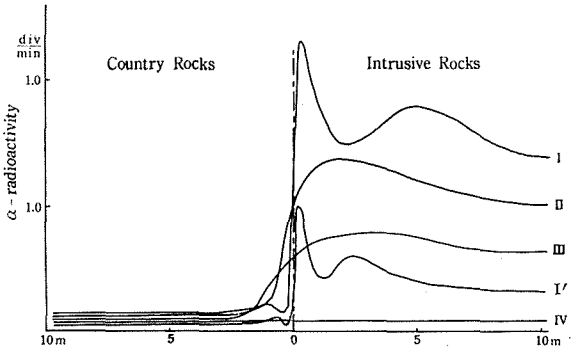

Fig. 4. Types of radioactivity distribution across contacts*.

In order to find out the relation between these types and constituent minerals of the intrusive rock concerned, the summarized data of chemical composition of granitic rocks by H. HATTORI and T. NOZAWA[6] were used.

Table II.

Radioactivity type	Localities
I	Tanakami, Shiga Pref. Hira, Shiga Pref. Tamano, Okayama Pref.
II	Tokuyama, (sourth part), Yamaguchi Pref. Itozaki, Hiroshima Pref. Kameoka, Kyoto Pref. Hiei, Kyoto Pref. Okazaki, (two mica granite), Aichi Pref.
II–III	Abukuma, Fukushima Pref. Tokuyama, (north part), Yamaguchi Pref. Matsuyama, Ehime Pref.
III	Watsuka (Koya), Kyoto Pref. Uwajima, Ehime Pref.
III–IV	Kitakami, Iwate Pref. Takamizu, Yamaguchi Pref.
IV	Yagiu, Kyoto Pref.
I′ (special)	Oike, Shiga Pref. Hita, Oita Pref.

* This classification of type is a final one.

(i) *The relation between the chemical elements and radioactivity types*: —

The relation between the chemical elements and radioactivity types were rather obscure, as observed on some examples in Fig. 5. But when K_2O–Al_2O_3 diagram was drawn (Fig. 6), it showed significant aspect of having some relation with the radioactivity types for these granitic bodies. As seen in figure, the change from Type I to IV may be considered to be corresponding to that of the composition from K_2O rich and Al_2O_3 poor to K_2O poor and Al_2O_3 rich. According to the data of S. SATO[7], these trends may be said to show the change from magmatic to metamorphic.

(ii) *The relation between normative constituents and radioactivity types*: —

(a) *Normative quartz-albite-orthoclase diagram*: —

The relation between the normative ***Q–Ab–Or*** relation and radioactivity types was obscure, but showed a tendency of compositional change from equi-portions of ***Ab***, ***Q*** and ***Or*** to ***Ab*** rich. According to BOWEN and TUTTLE[8], in the light of experimental studies in the system

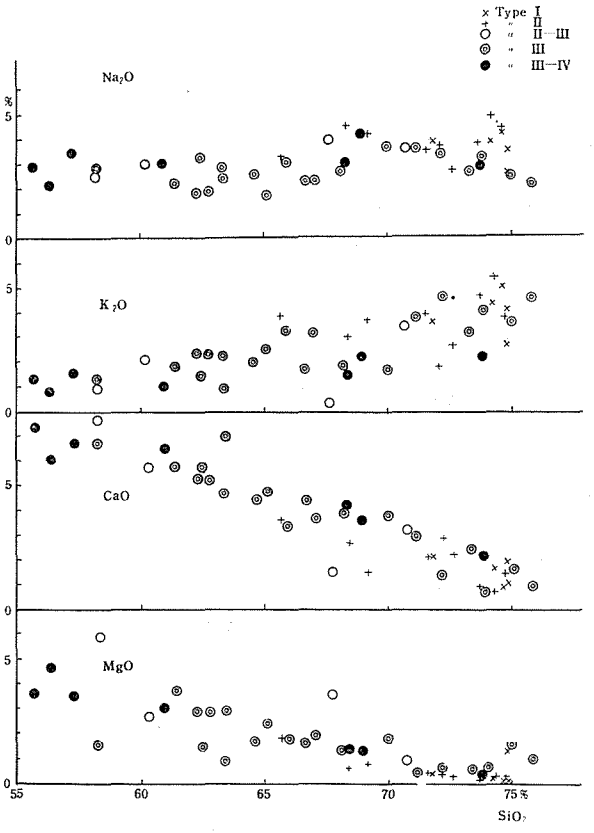

Fig. 5. Variation diagram of the granitic rocks.

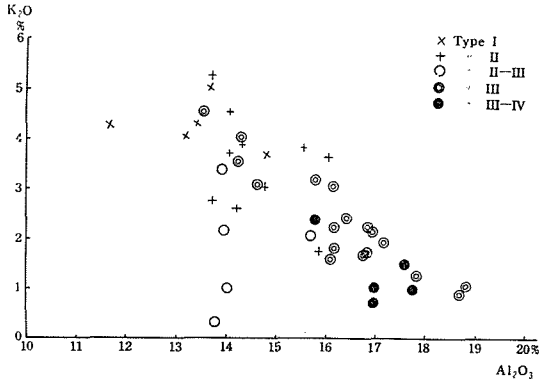

Fig. 6. Variation K_2O–Al_2O_3 diagram of the same granitic rocks as shown in Fig. 5.

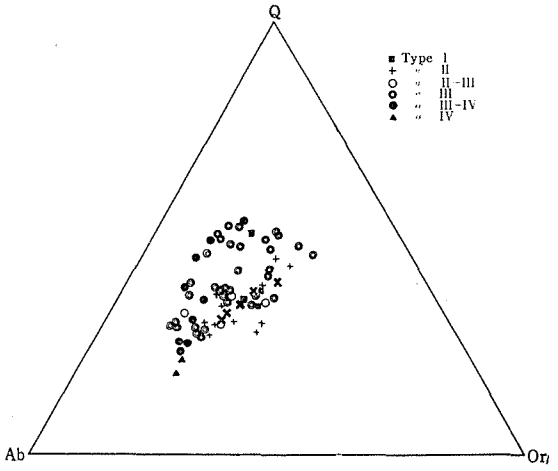

Fig. 7. Ternary diagram of the system, quartz-orthoclase-albitc relations obtained from norm calculations. Five crosses (X) represnt eutectic points under the vapour pressures of 500, 1000, 2000, 3000 and 4000 Kg/cm^2 respectively from the top.

$NaAlSi_3O_8$–$KAlSi_3O_8$–SiO_2–H_2O, most granites were formed by crystallization differentiation from magma or by remelting of the granitic crust, because their plottings of chemical analyses on the normative ***Q–Ab–Or*** diagram were remarkably concentrated near the ternary eutectic low temperature trough, and also it can be expected that the magmatic granites were formed in more shallow depth and granites or gneissose granites of metasomatic origin were formed in deeper depth. Five crosses in Fig. 7 represent eutectic points under the vapour pressures of 500, 1000, 2000, 3000 and 4000 kg/cm^2 respectively from the top cross. As seen in figure, it tends to the conclusion that Type I-granites were formed in the shallower depths and Type IV-granites were formed in the depper depths.

(b) *Normative Or–Ab–An diagram*:—

In ternary diagram, anorthite-albite-orthoclase relations obtained from the norm calculation showed significant bearings on radioactivity types for three granitic bodies, as shown in Fig. 8. For each granitic body, the trend from Type IV to Type I indicates that from metamorphic to eutectic. Relation between the representative type and the average value of feldspar obtained from norm calculation was also shown in Fig. 9. In this diagram, the granites classified as Type I fell in a limited zone, and were enveloped by those of Type II, especially in richer zone of albite. The granites of Type III-IV occupied the richer zone of albite–anorthite and those of Type III, the richer zone of orthoclase-anorthite.

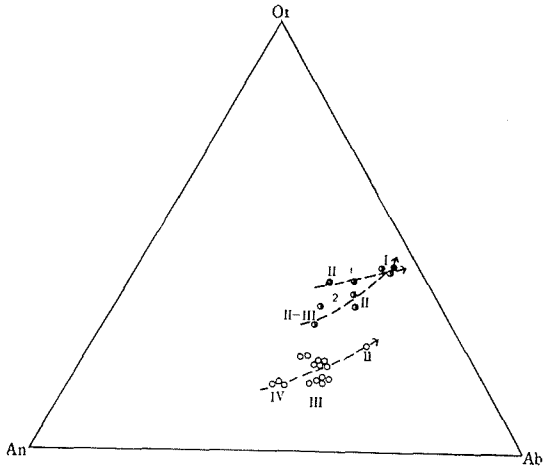

Fig. 8. Ternary diagram of the system, orthoclase-albite-anorthite relations obtained from norm calculation showing significant bearings on radioactivity types.
1: Tanakami
2: Matsuyama-Saijo-Tamano
3: Kitakami

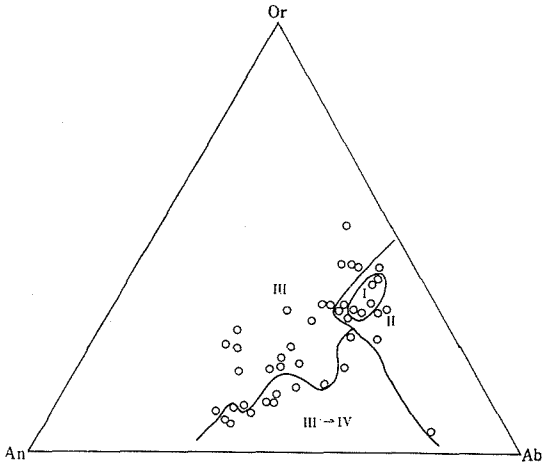

Fig. 9. Ternary diagram of the system, orthoclrase-albite-anorthite relations obtained from nom calculation showing significant bearings on radioactivity types.

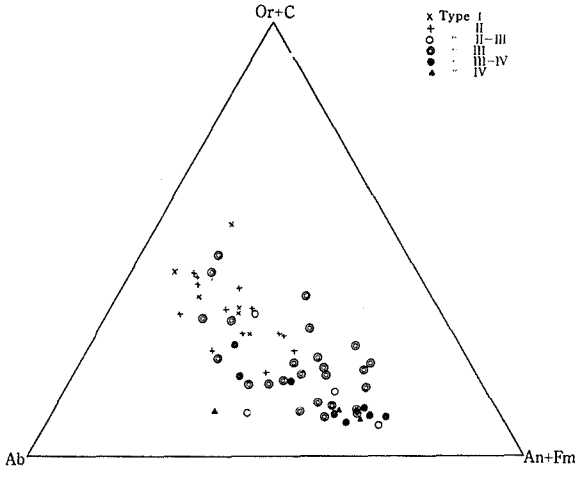

Fig. 10. Ternary diagram of the system, (orthoclase + corundum)-albite-(anorthite + femic minerals) relations obtained from norm calculations.

(c) *Normative (orthoclase+corundum)-albite-(anorthite+femic minerals) diagram*:—

As seen in Fig. 10 normative (orthoclase+corundum)-albite-(anorthite+femic minerals) diagram has been plotted for each tested granite. In this diagram, the trend from Type I to Type IV shows one corresponding to the change from (orthoclase+corundum) and *Ab* rich zone to (anothite+femic minerals) rich zone.

A factor controlling the distribution of radioactive elements in granitic rocks

There are a lot of problems unsolved concerning the origin of granite, but as a result of wide experience, long studies, and many discussions, the writer has come to agree with the conception that granites are formed by cooling of granite magma or migma[9)10)11)]. The physical states of the latters may be compared, as it were, to those of the *broth, mash* and *bouile* as has been done by READ[12)]. A mash may, for example, be consisting of magma, and minerals already crystallized from this magma, and the complete consolidation of this mash would give rise to an igneous rock. If the product had the specified mineral composition and texture, it would be called granite. Other kinds of mashes might be conceivably have the different origins. For example, a mash might be produced by the persistence of relict solid pieces in base which had become fluid, or by the breaking-up and strewing-about of solid rock by intruded fluid. Such mash would appear to be what M. REINHARD (1935) has called migma. When mash of the sort consolidates, it will not give rise to igneous rock in its strict sense, but in most cases, to mixed rock or migmatite. The migma, if the amount of its liquid portion become great enough, would acquire mobility to flow and could intrude itself into its surroundings in typical eruptive or intrusive fashion. The products of its consolidation, however, would still not be igneous rocks. But when the relict portions or the solid rock portions so completely disappear that the migma becomes entirely liquid. The resulted liquid would not differ much from the magma. With such considerations, one can naturally come to a conclusion that, whether as migma or as magma, when granitic materials move into the various higher levels in the crust, producing there the discordant granite bodies with aureoles of thermal metamorphism, there may arise a whole series of types (I to IV) of radioactive distribution at the contact depending upon the mobility of the granite materials and the difference of pressure and temperature and quantities of elements between intrusive and country rocks. The writer is now, to some extent, acquainted with the properties of such systems under varying conditions of temperature and pressure, and possesses in the available data the key to clarify the distribution of radioactive elements in the intruded rocks arising from magmatic or migmatic processes.

To begin with, we may examine the behavior of a melt or a partial melt

under a pressure and temperature corresponding to those at a depth in the eath's crust. According to N. KAWAI[13], in the case when a few of different Curie points are found in plutonic rock, the lowest Curie point is to be used for the depth estimations. From this stand point, granitic rocks from Mt. Rokkô and Ryoke zone in Kinki district were examined with thermo-magnetic analyses for depth estimation, giving the depth of less than 4 km and 18 km respectively. The temperature prevailing at the depths 4 km and 18 km are about 135°C and 600°C respectively[14]. At the depth 2 km, generally the pressure will be sufficient to prevent the escape of the volatile substances, no matter what the temperature may be[15]. But the difference of temperature between the intruded body and country rocks controls the mobility of volatile elements. The temperature of intruded body is estimated in various cases as about 550–600°C (wet melt) and 800–900°C (dry melt) (RAMBERG, 1952)[16], 1000°C (NIGGLI, 1929)[15], 1000°C (BOWEN, 1956)[17] 1100~1800°C (ITO, 1956)[18], 1487°C (KUMAGAI and ITO, 1959)[19], and also cited as 800°C (WATANABE, 1956)[20] and 1000°C (TAKIMOTO, 1952)[21].

At a shallower level where large difference of temperature prevails between invading melt and country rock, interior of intruding body is cooled slowly, while thin solid film may be produced outside of it comparatively in short time, but at a deeper level with small temperature difference, the cooling within the intruding body will be pretty more slow.

Now, in order to treat theoretically the cooling of an intrusive body, the following consideration has been made. Let the semi-infinite solid, consisting of country rock, be bounded by the plane $x=0$ and extend to infinity in the direction of x negative, the initial temperature being given by T', and assuming that at time $t=0$, a melt of a temperature T intrudes into the region x positive, the right side part of this plane. Suppose that the solidification of magma begins at a temperature T_2, and ends at a temperature T_1. The latent heat L of magma is liberated constantly over this range of the temperature. Put the specific heat of liquid magma be c_2', then an amount of heat c_2 defined by

$$c_2 = c_2' + L/(T_2 - T_1) \qquad \cdots\cdots(1)$$

will be given up by each gram of liquid when its temperature falls by 1°C in the range of temperature from T_2 to T_1. The melt is assumed to be of density d_2, thermal conductivity K_2, and diffusivity $h_2 = K_2/d_2 \cdot c_2$, and the country rocks to be of density d_1, thermal conductivity K_1 and diffusivity h_1. It was found by SCHWARTZ (1933) that the corresponding quantities for country rock and solidified magma have different thermal properties, but this was neglected here since it has been desired to keep the number of parameters to be considered as few as possible. For this reason, it is assumed the magma is intruded at the temperature T.

It can be shown following the analyses given by CARSLAW and JAEGER (1947)[22], that, at a time t after intrusion, the position X of the plane at which

solidification is just complete and so that the temperature is T_1, is given by

$$X=2\lambda(h_1t)^{1/2} \qquad \cdots\cdots(2)$$

where λ is a numerical parameter calculated as the root of the following equation,

$$\left.\begin{aligned} &\frac{1-erf\, b\cdot\lambda}{1+erf\,\lambda}\exp\left[(b^2-1)\lambda^2\right]=\frac{a(T_2-T_1)}{T_1} \\ \text{where}\quad &a=\frac{K_2h_1^{1/2}}{K_1h_2^{1/2}}=(K_2d_2c_2/K_1d_1c_1)^{1/2},\quad b=(h_1/h_2)^{1/2} \\ \text{and}\quad &erf\,\lambda=\frac{2}{\sqrt{\pi}}\int_0^{\lambda}e^{-\beta^2}\alpha\beta \end{aligned}\right\} \qquad \cdots\cdots(3)$$

is the error function usually tabulated. The temperature T_s in the solidified magma and country rock is given by

$$T_s-T'=\frac{T_1}{(1+erf\,\lambda)}\cdot\left\{1+erf\frac{x}{2(h_1t)^{1/2}}\right\} \qquad \cdots\cdots(4)$$

The temperature T_l in the liquid is

$$T_l-T'=(T_1-T_2\,erf\,b\lambda)/(1-erf\,b\lambda)+\frac{T_2-T_1}{1-erf\,b\lambda}\,erf\left(\frac{x}{2(h_2x)^{1/2}}\right) \qquad \cdots\cdots(5)$$

For simplicity, it is supposed that a platy sheet of melt with a width D, intruded into the region $-\frac{1}{2}D<x<\frac{1}{2}D$ between country rocks, x being the direction normal to the slab, the region $-\frac{1}{2}D>x$ and $\frac{1}{2}D<x$ consist of country rock. Fig. 11 is represented the result of an example calculated, assuming $T_2-T'=700$, $T_1-T'=500$, $L=80$, $K_1=K_2=0.005$, $c_1=c_2=0.28$, $d_1=d_2=2.7$ which may be reasonable for the case of granite intrusion. In each case, the temperature are plotted as a function of the time in years, at the points distant from the center by 0, 0.1D, 0.2D, 0.3D and 0.4D (in the igneous material), 0.5D (at the contact), and 0.6D, 0.7D, 0.8D, 0.9D and D (in the country rock), D being the width of the igneous materials in meters.

On the other hand, we must not leave the pressure problem out of account. It is well known that magma generally contains various volatile constituents. The influence of any such substances upon the properties of a silicate melt is unquestionably greater than that of a corresponding quantities of any of the ordinary rock-forming oxides. In this connection, many binary systems, which consist of certain compound with water, have been completely studied by BOWEN[17] throughout the range of compositions for pure compound.

NIGGLI[23] and SCHNEIDERHÖHEN[24] also discussed this problem. In Fig. 12 and Fig. 13, the behavior of a melt under conditions so defined may be followed. "a" represents volatile substance and "b" non-volatile substance. According NIGGLI, it has been shown that the complete process of solidification of a melt

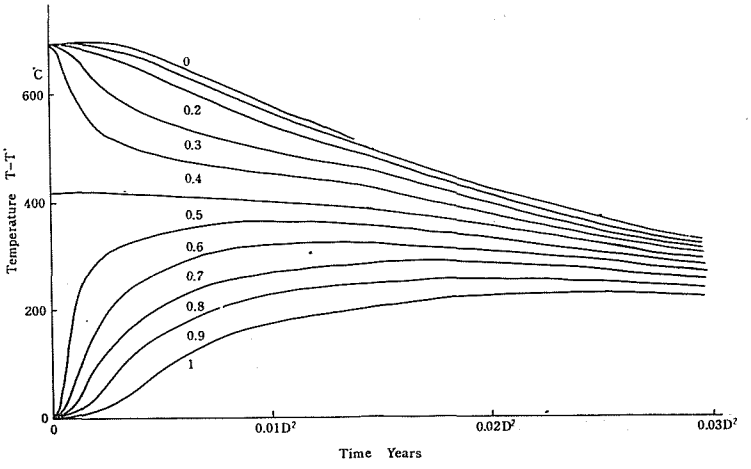

Fig. 11. Temperature $T-T'$ at various points in an intrusive sheet of thickess D meters for which $T_2-T'=700°C$, $T_1-T'=500°C$, $L=80$. The numbers on the curves are distance from the center measured as fractions of the thicknss.

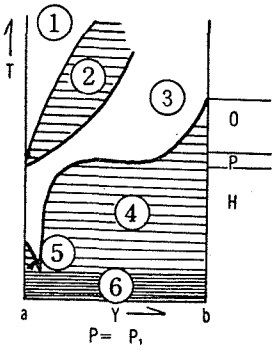

Fig. 12. T-X diagram of the system a-b, at constant pressure P_1 (coexistence of a gas and a solidus phase is not possible). This can be deduced from Fig. 13. (Niggli, 1954).

1: *Gab*
2: *Gab*+*Lab*
3: *Lab*
4: *Lab*+*Sb*
5: *Sa*+*Lab*
6: *Sa*+*Sb*
O: orthomagmatic stage
P: pegmatitic stage
H: hydrothermal stage

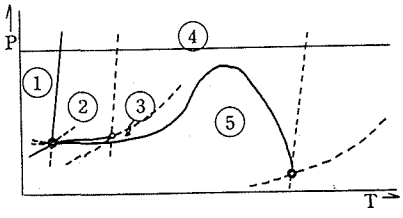

Fig. 13. Entire P-T diagram of a binary four-phase system, with the quadruple point, and the curves leading to the limited unary systems, I and II are the triple points of the unary systems a and b. (Niggli, 1954).

a: Volatile componet
b: Non-Volatile component
1: *Sa*+*Sb*
2: *Lab*+*Sa*+*Sb*
3: *Gab*+*Lab*+*Sa*+*Sb*
4: *Gab*+*Lab*+*Sb*
5: *Gab*+*Sb*

cooling under the condition of pressure corresponding to a depth of about more than 2 km comprises the following stages: (i) The orthomagmatic stage in which is included the formation of igneous rock *sensu stricto*. The volatile components act chiefly as mineralizing agents—i.e., they reduce viscosity, depress the melting points and influence the inner magmatic chemical equilibria. (ii) The pegmatitic and pneumatlitic stages, which form the connecting link between the above stage of silicate melts and the aqueous solutions characteristic in the hydrothermal stage. (iii) The hydrothermal stage.—Here cooling is accompanied by relatively slow crystallization from solutions with high concentrations on the chief volatile component—water.

If we take these conditions into consideration, we shall get a right understanding on the distribution of radioactive elements. The distribution of radioactive elements in granitic rock is studied by several authors such as H. GROSS[25], P. M. HURLEY[26], E. PICCIOTTO[27] and Z. HATUDA and the writer[1)2)3)]. The conclusions deduced from these studies are summarized as follows: (i) The radioactive elements are concentrated in the rock crystallized late. (ii) The radioactive elements are concentrated toward the outer margin of batholiths. (iii) A large proportion of the radioactive elements in rocks occurs in grain-boundaries of the rock-forming minerals. This would be supported also by the following results shown in Table III and Fig. 14.

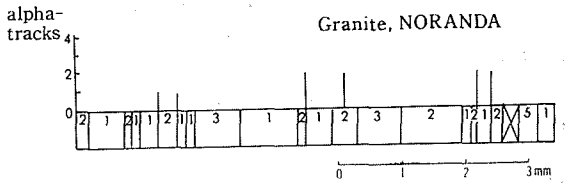

Fig. 14. The relation of alpha-radiation to mineral grains; N. T. A. Plates exposed for 60 days. (Gross, 1952).

1 Quartz 4 Zircon
2 Orthoclase 5 Sphene
3 Plagioclase ☒ Parallel to Contact

Table III. Effect of leaching with HCl aq. on the radioactivity of pulverized rock.

Materials	Alpha-activity (div/min)
Crushed sample of corse-grained granite, Shishitobi.	0.85
The same sample washed with 10 N HCl.	0.42
difference*	0.43

* The difference may be considered attributive to alpha-activity mostly due to materials in the grain boundaries.

The representative radioactive elements in granites are uranium and actinium ($U^{238}+U^{235}$) families, thorium (Th^{232}) family and potassium (K^{40}).

According to GOLDSCHMIDT[28], calcium enters the feldspar lattice more than

sodium does on account of its higher charge, and so is concentrated in the early-formed plagioclase. The potassium ion is considerably larger than sodium ion, and potash feldspar is generally among the last minerals to crystallize. The mgnesium ion is somewhat smaller than the ferrous ion, and magnesium is always concentrated in the early-formed ferro-magnesian minerals. However, GOLDSCHMIDT's rules have had thier greatest utility in predicting the order of removal from a magma not only of the major elements but also of the minor elements. When a minor element has the same charge and an ionic radius similar to a major element, we may speak of it as being camouflaged in the crystal lattice containing the major element. When a minor element has a similar ionic radius but higher charge, or the same charge but a less radius, than that of a major element, it is said to be captured by the crystal lattice containing the major element. Finally, when a minor element has a similar ionic radius but a lower charge, or the same charge but a greater radius, than that of a major element, it is said to be admitted into the crystal lattice containing the major element. But *Th*, *U* and elements having such a character are those which, on account of too greate difference in ionic radius and ionic radius and ionic charge, do not replace the major elements. On account of low concentration in the residual liquid of magmatic crystallization, they are concentrated in pegmatitic minerals.

During the magmatic cycle, uranium as well as thorium is in the tetravalent state, and the crystallization course of both elements are parallel owing to close simillarities in ionic radius[29)30)31)]. Commonly, at a very late magmatic stage, a change takes place which brings the uranium and thorium to a parting of the ways ; the uranium going with the hydrothermal solutions, leaving the thorium to crystallize. It is noteworthy that this process makes possible the formation of thorium free uranium minerals.

In conclusion, it may be anticipated that there is no difference in the variations of K^{40} across each intrusive in any different depths of the earth's crust. Besides, as *U* and *Th* move as volatile matter under large difference in temperature between intrusive and country rocks, *U* and *Th* deposit in quantities within thin film as has been previously mentioned. Near the contact in shallower depth, the temperature falls abruptly, but within the intruding body will be pretty more slow, according the vapour tension is so decreased that diffusion of volatile matter from inner part towards contact zone acts on briskly. On the contrary, with small difference of temperature, *U* and *Th* are diffused widely to country rocks in a long time or they deposit in equaly all over the igneous rocks. Then, it may be shown that the types were classified by the profiles of alpha-activity as has been mentioned.

Summary remarks

(1) From the spectrographical analyses, it has been found that (i) in the Tanakami traverse, the boundary shows sharp distributions of the elements,

but in the Takamizu and Koya traverses the boundaries reveal rather flat distribution of the elements. (ii) Across the contact of Tanakami granite, the increase of volatile matter within the invading rock with decreasing distance from the boundary is conspicuous, but across the contacts of Koya and Takamizu granites, the distributioas of volatile matters are obscure, and (iii) notwithstanding bad condition for analyses of *Th*, the spectra of *Th* are recognized at the position corresponding to the marked hight of radioactivity.

(2) In spite of the fact that the variations of alpha-activity acros Takamizu and Koya granite contacts are quite different, those of beta-activity have much in common.

(3) The beta-activity distribution may be largely dominated by the distribution of K^{40}, and the alpha-activity distribution by those of *U* and *Th*.

(4) In the cooling history of intruded melt, K^{40} may be found in potash-feldpar and mica, and *U* and *Th* are concentrated towards residual portion.

(5) The distribution of volatile matter in intruding body may be concerned with the difference in temperature between granitic melt or partial melt and country rocks.

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