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The Radioactivity of Rocks and Minerals Studied with Nuclear Emulsion
I The Minute Radioactive Minerals of the Tanakamiyama and Mikumo Granites, Siga Pref. Japan

By

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Abstract

The radioactivity of minute minerals in the granite are measured by means of nuclear emulsions. Petrological and geological distribution of radioactive substances of the Tanakamiyama and Mikumo Granites are described. In general, the radioactivity content of the stock type granite is greater than that of the batholithic one, and in either cases, the marginal facies of the granitic mass is more radioactive as compared to the interior. From the mineralogical stand point, the radioactivity of each minute minerals seems to have intimate relation to the deuteric actions.

Introduction

By their pleochroic halos in biotite some species of minute accessory minerals in granitic rocks, such as zircon, allanite, apatite, sphene, etc., were found to contain radioactivity as already studied by Water[1a][1b], in 1909, on some radioactive grain minerals in British granites. His intersting attempt, however, owing to the inefficiency of his herium method in determining the geological ages of those rocks, found no successor for more than a generation. Recently these accessory minerals were once more taken up for "determining the age of igneous rocks" by Hurley[2] and Larsen, Keevil and Harrison[3]. On these method, the total amount of the radioactive elements contained in granitic rocks is, indeed, easily measurable, but the most sensitive electronic instrument cannot determine how much radioactivity each grain contains. Now the nuclear plate more refined than ever is very much furthering this sort of measurement[4][5][6][7]. The difficulties which the autoradiographic method has confronted, can be removed by the technic recommended by the author[8]. The nuclear plate used for this purpose was ET–2E (50 microns in thickness) furnished by Research Laboratory Fuji Photo Film Co., Ltd. The degree of its fading and amount of blank tracks were tested before use. The exposure extended four weeks in ordinary cases, but was varied with the different degrees of radioactivity from one to eight weeks. The author's method was found efficient especially in dealing, by correlating alpha-tracks, with those minute radioactive minerals. The radium contents of Tanakamiyama and Mikumo granites were measured by Mr. Asayama(9), while their total alpha radioactivities by the author on autoradiographic method; and concerning radioactivities thus obtained, the two result relatively agreed with each other. The minute radioactive minerals vary quantitatively relative to their radioactive elements, not only in different granitic rocks, but even in the same thin section. According to Holme's report on Mozambique pegmatite(10), the finer the zircon is, the more uraniferous it is. This tendency is also tracable in all granites the author studied.

General Geology and Petrography

The Tanakamiyama granite (Southern part of Siga Prefecture) is elliptically intrusive and extends 14 km. from east to west and 7 km. from north to south, and the Mikumo granite lies, making batholithic intrusion, connected with the former stock at its southeastern part. (Here, in this paper, by T. G. we understand the Tanakamiyama granite and by M. G. the Mikumo granite respectively.) The northern part of this intrusion is partly overlapped by Plio-Pleistocene sediments, and has partly metamorphosed Palaeozoic formation mainly into hornfels containing cordierite, tourmaline and andalusite. The Palaeozoic sedimentary rocks are the alternation of sandstone, shale, and chert; their general strikes have the same direction as the long axis of the elliptical granitic intrusion. T. G. is chiefly of coarse grains; in its marginal facies, however, partly of medium or of fine grains or porphyritic—invariably containing biotite as their colored mineral. Precious topaz crystals were once found much out of this granite pegmatite, but now are often seen in this granite itself as its accessory minerals with fluorite and topaz. This biotite is reddish brown in the core, and greenish brown in the peripheral zone, showing myrmekitic structure. The greenish biotite contains more fluor and, therefor, less refractive index ($N_\gamma = 1.63$) than the reddish brown ($N_\gamma = 1.64$). The fluorite inclusions are seen mainly in the basic core of sericitised plagioclase. The calcium contained in anorthite was presumably combined with the fluor making these fluorites. The leucocratic facies of T. G. have been furnishing pottery stone. This granite, constituting a stock, is in connection partly with the batholith of M. G. (Ryoke granite) which is coarse-grained hornblende biotite granite. As the contact metamorphic rock, however, the former possesses hornfels, while the latter either schistose biotite hornfels, or schistose granite (lit-per-lit injection gneiss). As located farther away from the neck part of the two (T. G. and M. G.), these granites gradually differ from each other in petrological and radioactive quality.

The relation between radioactivity of rocks and their minute radioactive minerals

According to Mr. Asayama's measurement of radium contents of these rocks, (1) T. G. contains over 1.5×10^{-12}g/g radium (comparatively high among Japanese granites) and M. G. under 1.3×10^{-12}g/g radium (relatively low in Japan) and (2) the radium is more densely distributed in the contact margin of the stock or batholith than in their core.

The pleochroic halos in the biotite of the two granites characterize clearly the different orders of their radioactivity. There lies an intimate relation between the blackness of pleochroic halos and the radioactive elements contained in zircon. Though over-exposed halos are often seen in T. G., under-exposed ones are rather found in M. G. naturally because of the lower radioactivity of its nucleus minerals. Granite is generally said to owe its radioactivity incomparably more to minute accessory minerals (zircon, allanite, etc.) than to essential rockforming minerals (quartz, felspar and biotite)*. Now T. G. contains comparatively rich zircon and scanty allanite, while M. G. contains more allanite and is characterized by the zonally yellowish altered margin. This sort of mineral is often seen also in Ryoke Zone adjacent to M. G. Here the radioactivity of zircon stands in no direct proportion to that of allanite. In T. G. there still exists a certain relation between the halo frequency and the radioactive order. As a rule, the more radioactive the rock is, the more halos it possesses. Leucogranite, however, in spite of its partial richness in radioactive elements, lacks pleochroic halos. The co-exsistence of both over- and under-exposed halos is now of great significance, suggesting the diversity of radioactive orders of those nucleus minerals.

In autoradiographic studies on rocks and minerals, the quantity of Thorium and Uranium contents can be designated by the alpha-track population and the comparative number of the long range of the alpha-track[11a][11b][12]. The author's results there on will be given in another paper. Unlike the ordinary radium measurements e. g. the solution method, for which some 10 grams of it is required, the radioactivity of any rock sample, however little the amount may be, can be determined by autoradiography. The amount of rock sample needed for the autoradiographic measurement is about 0.035 grams of the thin section and this ammount is sufficient enough. On account of the sporadic concentration of radioactive elements in the minute radioactive accessory minerals, two thin sections made out of the same sample of granite seldom show similar order of activity; the radioactivity measurement by the autoradiographic method is applicable only to rocks of homogeneous distribution of the radioactive elements such as volcanic ones. This method, however, must be somewhat modified, when the total alpha radioactivity of granites is measured by means of nuclear emulsion.

Radioactive Geology

Heavy minerals in igneous rocks were often recommended as the means of correlating igneous intrusions. In spite of several unidentified radioactive mine-

* Recently Dr. Hurley made an important discovery[2] about the distribution of radioactivity containd in the rocks, concerning it the author will report his own investigation in another paper.

rals found by the author as sporadic occurrance, zircon is still the most abundant minute mineral of radioactivity in granite. It is a laborious task to count the alpha-tracks on the microscopic stage. The irregurality of grain shapes and the paragenesis of two radioactive minerals of different sorts often make the measurement impossible. Generally speaking, the radioactivity of the minute minerals greatly varies as their host minerals differ. Quartz contains minute minerals covering from the strongest to the feeblest in radioactivity and, consequently, is the most abundant in minute radioactive ones. In biotite they are less in number and medium or feeble in activity. Albitic plagioclase is inferior to biotite both in number and activity; in its basic core, however, no such minerals are found at all. In orthoclase they are both the rarest and the feeblest. The cloudiness of the zircon is due to its suffering deuteric action. Generally, T. G. contains more such zircons than M. G.. In either case, cloudy zircon is often found in the granite near contact, the leuco-granite, and the rock near the xenolith. T. G. has made a concordant intrusion into the Palaeozoic formation as elliptical outcrop, while M. G. a stopping intrusion of batholith in a wide area. Such an interrelation between strong radioactive stock type granite and feeble radioactive batholithic one, can be seen all over the inner zone of the south western part of Japan. The following is the data obtained concerning these granites:

I. Stock Type Granite (T. G.)

(a) Contact Type:—

The assemblage of minute radioactive minerals is abundant in contact facies of the rock, of which the radium contents amount to $2\text{–}3 \times 10^{-12}$g/g, and is constituted mostly of zircon involving 1–3% Uranium equivalent and partly of rare radioactive minerals containing less than 20% Uranium equivalent. This phase of granite is much diverse in radioactivity, the maximum of which lies, however, not exactly in the contact plane but rather in granite a little apart from it. The radioactivity counting instrument also verifies this[(13)].

(b) Central Type:—

This type is mainly of coarse grain biotite granite partly containing muscovite. Here zircon about 3% uraniferous, though less than in the contact rock, still constitutes its chief mineral. Besides, in some rare cases an extremely radioactive mineral like uraninite is found also here. As for radium content, here being 1.5×10^{-12}g/g in average, it is generally less than in the contact facies.

(c) Leuco-granite Type: –

In T. G. there are many sporadic parts of leucocratic rock facies without biotite. As these rocks have severely been weathered, their radioactivity generally cannot easily be determined. According to the author's result, however, their radioactivity seems less than that of the former type.

II. Batholith Type Granite (M. G.)

(a) General Type: –

This granite is of batholith type and, having made a flood type intrusion, extensively distributed. In spite of its Ryoke type, it generally lacks the schistosity. Its radium content is about 0.7×10^{-12}g/g, while its uraniferous zircon contains 0.1—0.34% Uranium equivalent.

(b) Contact Type : –

M. G. of this type is generally more radioactive than that of former type. Near the contact with Palaeozoic formation at Asamiya it contains 1.2×10^{-12}g/g radium. Petrographically it resembles the central type of T. G..

(c) Roof pendant (Schistose biotite hornfels) :—

The pecuriality of rocks of this sort, which are called in Kasagi district "lit-par-lit injection gneiss", is that in radioactivity they greatly surpass the schistosed biotite granite (i. e., normal type M. G.) which accompanies them.

Distribution of Radioactive center

I. (a) In case of fine, medium or porphyritic granite of T. G. (stock type,

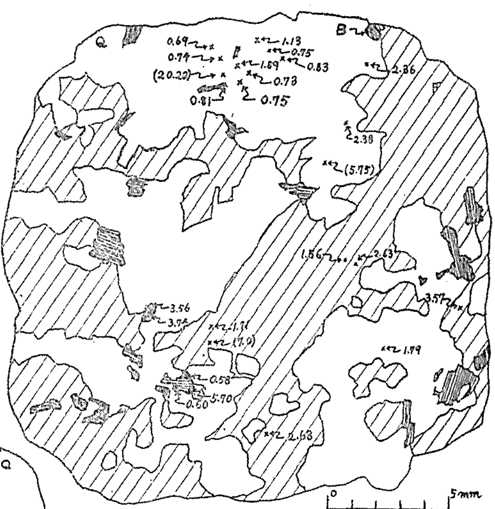

Fig. 1

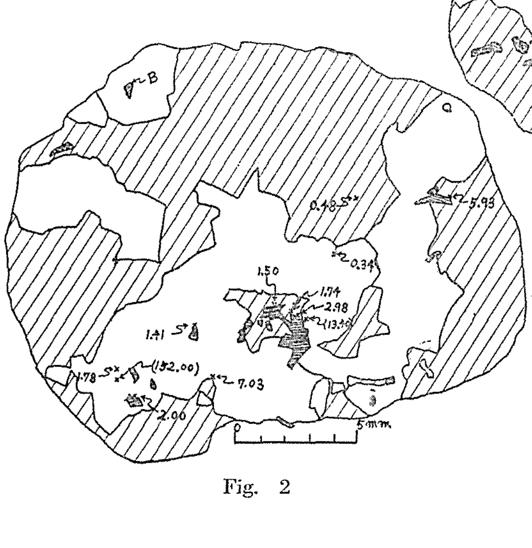

Fig. 2

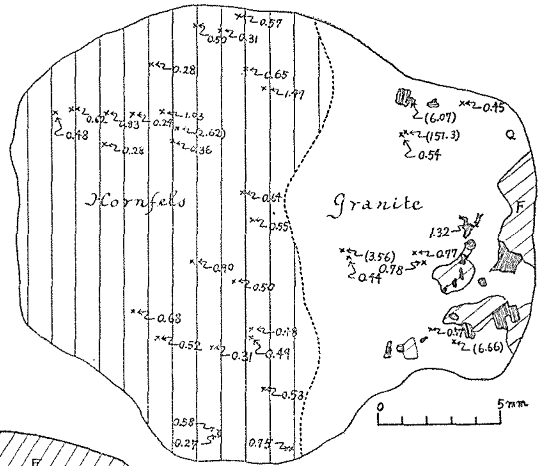

Fig. 3

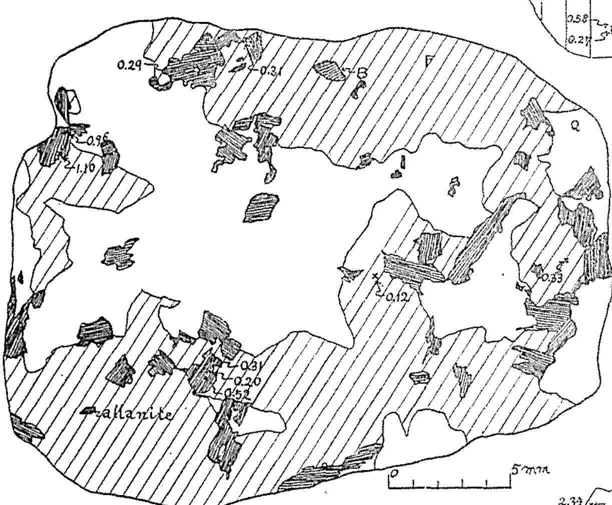

Fig. 4

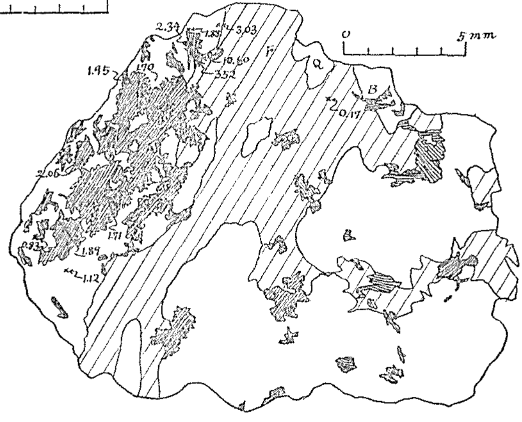

Fig. 5

contact rock facies) as in Figure 1 (T. G. 286, Nango.), the radioactivity of zircons indicates the side range even in the same crystal of quartz, or in the same thin section, of the rock. Figures 1, 2, 3, 4, and 5 show the full space of thin section, "Q" meaning quartz, "B" biotite, "F" felspar, and the numerical values indicating the alpha radioactivity ($T\alpha$) of zircon, namely, the number of alpha particles ejected from the polished surface of radioactive mineral per square centimetre per second. The values put in brackets show those of other minerals than zirconic ones. Though not shown in the figures, these minute radioactive minerals are connected with the fine veinlet of albitic plagioclase running through quartz and felspar. This severely affects the radioactivity of each zirconic grain. Grains of the same order of radioactivity lying near to each other are found upon the same veinlet or fracture in the host rockforming mineral. The difference in the orders of activity is originated, not from the radioactive fractuation by size or shape of the grain, but from the different contents of the radioactive elements. In other cases, the deuteric action accompaning the radioactive elements seems to have been affecting in succession. The radioactivity of zircons containd in the same crystal of quartz (upper part of the Figure 1) amounts mostly to $T\alpha = 0.7$, but in its peripheral or net-work felspar invasion parts, the radioactivity is extremely high ($T\alpha = 2.3$). Biotite also contains diverse groups of zircons whose radioactivities vary from $T\alpha = 0.6$ to 5.7. Figure 6 shows their size, shape, cloudiness

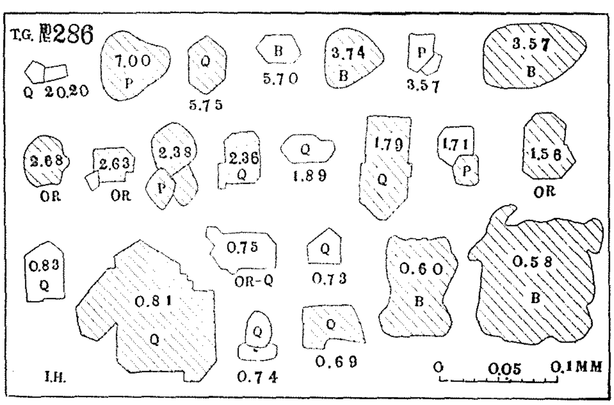

Fig. 6

In Figures 6......12, B, Q, P, etc. show the host mineral. B: Biotite, Q: Quartz, P: Plagioclase, OR: Orthoclase, B–Q: between Biotite and Quartz. Shadow lines show the cloudy zircons.

(represented with shadow lines), host mineral, and the alpha radioactivity ($T\alpha$). In that figure most of such minerals are clouded. As a rule, their cloudiness stands in no intimate relation to their orders of radioactivity and to the species of host minerals. Genereally the contact rocks (T. G.) contain many such cloudy zircons or the colored zirconic minerals. Paragenetic intergrowthes of two kinds of minerals are often seen in the Figure 6 ("OR–2.63": cloudy zircon accompanies reddish one, "P–1.71", "Q–0.74" transparent and cloudy zircons). In these cases, on account of minute grains and diversion of alpha-tracks, it is impossible to determine which grain is more radioactive. The application of coated method of autoradiography[5], however, enables us to determine it exactly. The yellowish mineral "Q–20.20" is lower in refractive index and birefringence than those of zircon. To zircon belong "B–5.70" (yellowish brown), "B–3.74" (with reddish stain), "P–3.57" (pale yellowish brown), "Q–2.36" and "Q–0.73" (brown). Not only their size, cloudiness, and the host minerals, but also their colors have nothing to do with their orders of radioactivity. The colored zircons characterize the T. G. contact granite. Round zircon is generally higher in radioactivity than fine edged and elongated crystal "T. G. 668" and "T. G. 211" containing many considerable radioactive zircons are the contact granites of Sisitobi. In these granites, zircons show two different orders of radioactivities: $T\alpha=1$–4 and $T\alpha=0.5$. Because of its diverse mineralogical features, the one group of higher radioactivity seems to consist of more various minerals. It must be noticed that such a highly radioactive mineral as contained in the central type of T. G. is not found in these rocks. "T. G. 750", namely, the biotite granite with some muscovite, contains clearly and cloudy zirconic grains, and their activities are $T\alpha=2.30$–0.65. As reported by Hutton[14], in its thin section, too, the stronger radioactivity is due to the round grains, while the weaker one is originated from the sharp crystals of regular from. Fairly radioactive are both the yellowish minerals accompaining the yellow micaceous halo in quartz and the brownish stained zirconic ones. Large sized, reddish and apparently rutile-like mineral ($T\alpha=0.78$) is also seen in its section. The autoradiographic measurement clearly designates that the granite of these radioactive characters belongs to the contact margin.

I. (b) In case of the coarse grained biotite granite of T. G. (stock type, central rock facies), the radioactive minute minerals are so distributed in the thin section of T. G. 712 as shown in Figure 2, that those minerals are known, though almost entirely zirconic, to differ greatly in their radioactivities, containing both the strongest and the feeblest. This granite found on the cross-road of Ootorii at the center of that region is representative in T. G. with its greenish brown biotite, much pleochroic halos, small amount of muscovite, and fluorite inclusion in the clouded plagioclase. The chemical analysis and radium measurements of all these rocks will soon be given by T. Asayama[9]. The numerous minute radioactive minerals are shown in Figure 7 and in photographs 1a, 1b. It is known from Figure 2 also that they are not scattered at random, but lie mostly in certain rock minerals; they are assembled like a druse of pegmatite, in quartz, together with biotite and albite etc., but seldom found in orthoclase and oligoclase. One of them (Photo. 1a, 1b) was found highly radioactive after 28 days' exposure, and its alpha-tracks were too numerous to count. B (in Photo. 1a) shows the strongly radioactive minerals and appears magnetite-like opaque mineral, and Bt (in Photo. 1b) shows the alpha-tracks emitted from B after 7 days' exposure. Its radioactive order corresponds to that of uraninite or pichblende. Lt (in Photo. 1b) is the alpha-tracks due to the mineral L (in Photo. 1a) (cloudy malacon type zircon) and this shows how strongly radioactive B is. In Figure 7, "B–Q 13.40" shows the parallel growth of a brownish mineral and the clear type zircon around it. Its activity is due to the former brownish mineral (ferugusonite?). "Q–7.03" has a severely blackening margin. "P–5.93" has reddish stains. The right half of Figure 7 shows another exsample T. G. 707 lying near T. G. 712. This is

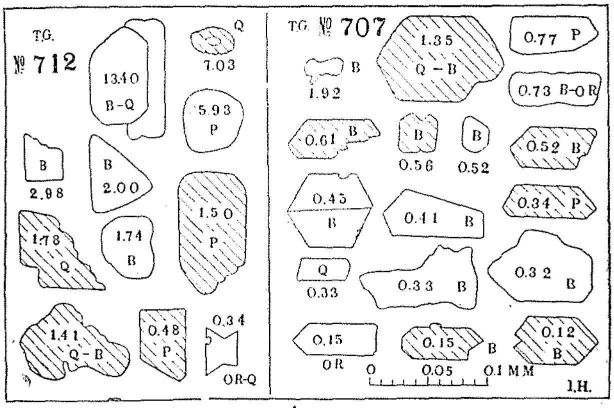

Fig. 7

the porphyritic granite accompanying xenolith. It resembles T. G, 712 except that it contains allanite (B–0.45 zonnary and twinning crystal, B–0.41, B–0.33, B–0.32), "P–0.34" is the purple cloudy zircon, and "B–OR 0.73" may be a certain secondary mineral. Photo. 2a and 2b must be noticed for the strongly radioactive mineral "E" and for its 28 days alpha-tracks "Et". Under the microscope it looks pale yellowish and isotropic; in radioactivity ($T\alpha = 33.4$), however, it resembles betafite. The above-said strongly radioactive minerals may be found, as in case of Kitasirakawa and T. G., mostly in the central type of stock granite.

Figure 8 shows the same type of T. G. 550 and 738. These rocks are seen in south Kirihu and near Maki. Owing to the coarse grained radioactive zircon, these rocks are highly radioactive. T. G. 738 is more or less leuco-granitic, though T. G. 550 belongs to the central type. One grain of zircon in T. G. 550 (B 0.26 in Figure 8) interested the author with the heterogenious distribution of radioactive element in the minute crystal. Firstly, its pleochroic halo is seen only half around this grain and the alpha-track distribution exactly comfirms this heterogeneity. Of large zircons, such a phenomenon was already reported by

Buttler and others[12]. In a minute zircon, however, nothing of this sort is yet depicted. Because the zircon shows the halo only on the side of albitic invasion, the cause of this phenomenon seems, at present, to the author to have been due to the albitic residual solution whose radioactive element was then absorbed into the nearer part of this non-radioactive zircon. P 2.62 and P 2.81: yellowish

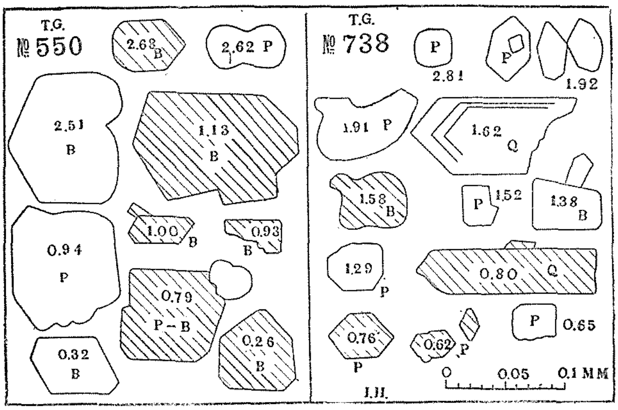

Fig. 8

brown, B 2.51, B 1.13 and P 1.29: yellowish stain, P-B 0.79 (the upper right in Figure 8): yellow. P 1.92 (the left-most one) contains an unidentified brownish nucleus.

In xenolithic rock of T. G. the minute minerals are distributed, as shown in Figure 3 (T. G. 666), always accompanied by biotite and crowded, as fine grained, mostly in the boundary part of xenolith, and their radioactivity $T\alpha=0.3-0.6$. In Figure 9, however, the minute minerals contained in granitic parts are not

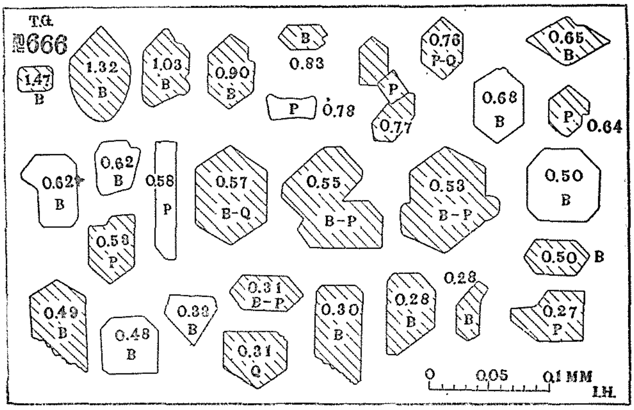

Fig. 9

given. In the hornfelsic xenolith it is note-worthy that they are mostly cloudy zircons of the same order of radioactivity ($T\alpha=0.6$) as the zircons accompanying biotite of contact granite (Figure 1). In granitic parts, those in quartz are higher in radioactivity, and fewer in number than those in the xenolith. The varieties of radioactive minute minerals are chiefly secondary ones, such as yellowish isotropic minerals or zirconic isotropic ones. Further progress in the knowledge of the optical character of minute radioactive minerals, alone, will be able to make clear these questions. T. G. 665 (medium grained porphyritic granite of Sindenba quarry at the same locality of T. G. 666) contains allanites. Its minute radioactive minerals are similar to the T. G. 666. Three purple cloudy zircons were unexpectedly discovered in orthoclase, their size was about 2×10^{-4} mm² and their radioactivities: $T\alpha=3.4-4.3$.

I (c) T. G. 271 (the leuco-granite near the Suisyodani pegmatite) contains a few grains of relatively feeble radioactive minerals ($T\alpha=0.62-1.95$). Its relatively high radium content (1.75×10^{-12}g/g) is due to the fact that the limonite-like mineral and other undetermined reddish ones, though feeble in activity, are abundant in it. In the distribution of their radioactive elements, the unaltered biotite granite, namely, the primary radioactive minerals, essentially differs from the leuco-granite, the secondary ones. Figure 10 is the rocks of the transitional

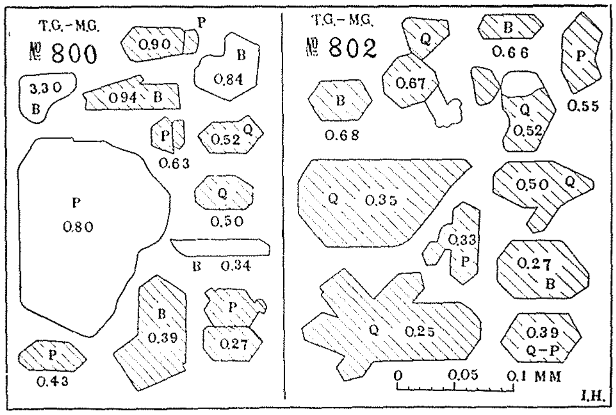

Fig. 10

phase from T. G. to M. G. Here in this facies (T. G.-M. G. 800 and 802) both zircons of feeble activity characterizing M. G. and cloudy ones peculiar to T. G. are found. Under the microscopic appearance, too, the granite contains, like T. G., greenish myrmekitic biotite and at the same time zircon grains are distributed in quartz, as it were, in the form of a druse. Like M. G., however, allanite is contained even in coarse grained biotite granite.

II (a) Such a granite as shown in Figure 4 (M. G. 521) is seen everywhere in Mikumo district. It is characteristic of M. G. that these granites accompany biotites clotted as well as scale-like. These zircons are accompanied with biotite and feebler in radioactivity ($T\alpha = 0.3 - 0.6$). Zircons are fairly large, transparent and pale bluish, beautiful crystal, with feeble radioactivity. Zircon of this granite is shown in the right half of Figure 11. Here what interests the author

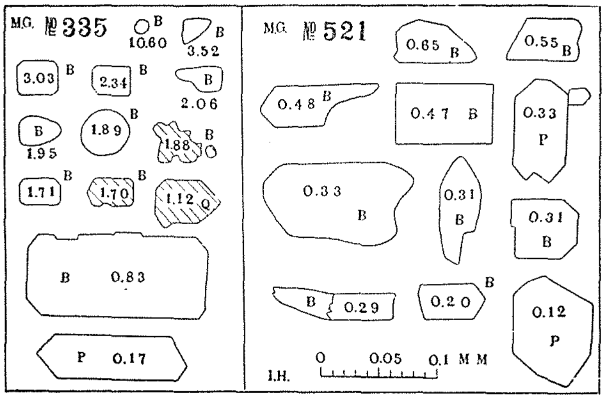

Fig. 11

most is that the three zircons involved in biotites of the three different rocks — M. G., T. G. (in the feeblest radioactivity), and xenolith in T. G. — all belong to the same order of radioactivity. One of characteristics of M. G. is that it always contains allanites even in its main type rocks. In T. G., however, allanite is found rather seldom and only around xenolith. These two facts seem also to suggest the granitizational origin of M. G.

II (b) Figure 5 (M. G. 335) shows the rock in which a schistose biotite hornfelsic rock is crossed by a pegmatitic granite vein, and there the radioactivity

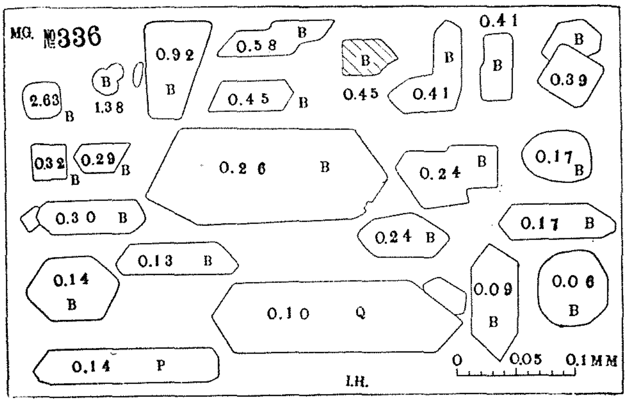

Fig. 12

stands much higher. The left half (No. 335) of Figure 11 shows two groups of zircons, one of which is similar to those of main type of M. G. (II a), and the other of which is rounded minute crystal, fairly high in radioactivity. The sample for Figure 12 (M. G. 336) is of nearly the same granite as the M. G. 335. M. G. 336 contains cloudy zircons as well as rounded ones of higher activity fewer than M. G. 335 does.

Summary and Conclusion

After his geological survey of Tanakamiyama, Mikumo and Sigaragi districts, the author's result obtained from autoradiographic investigations is as follows;

Geological aspect

A. Stock type granite (T. G.)
   - a. In the granites of contact facies, the radioactive elements are concentrated mostly in zirconic minute minerals. ($2–3 \times 10^{-12}$g/g Ra)
   - b. These central type rocks, however, contain the strong radioactive minerals of various species; not only the number of the grains including zircons, but their radioactivity is less than in the former (a). $1.5—2 \times 10^{-12}$g/g Ra)
   - c. Leuco-granite is of m-derate radioactivity originated from the secondary minerals, but a severe fractuation of radioactivity is to be expected. ($1—1.5 \times 10^{-12}$g/g Ra)

B. Batholith type granite (M. G.)
   - a. Contact type of this granite is moderately radioactive. (about 1.2×10^{-12}g/g Ra)
   - b. Main type of this granite seems to have small quantity of radioactive elements. (less than 0.7×10^{-12}g/g Ra)
   - c. Schistosed hornfelsic granite shows moderate radioactivity. (perphaps over 1.5×10^{-12}g/g Ra)

Mineralogical aspect

1. Generally speaking, the radioactive elements in granite are concentrated sporadically in minute radioactive minerals.
2. Its radioactivity depends upon the amount of zircon as well as upon the content of their radioactive elements.
3. Even in the same thin section of rock, the radioactivity extremely varies in respective mineral grain.
4. Though the color of zircon is not necessarily an infallible index of its radioactivity, there is a general tendency that zircons both colored and of low birefringence show more radioactivity. Colored zircons often appear in the stock type granite T. G. and especially in its contact facies.

5. The radioactivity of zircon tends to be stronger in smaller and rounded crystal, but feebler in larger and beautiful one.
6. Cloudy zircons are found always in T. G., in M. G. only in the contact facies, but never in M. G. proper.

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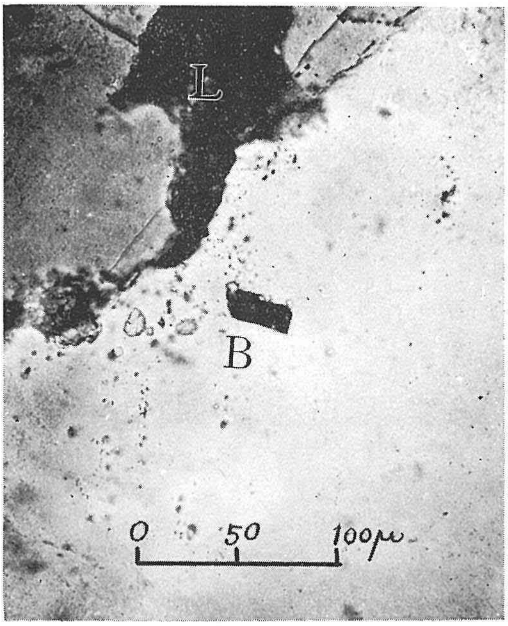
Photo I a



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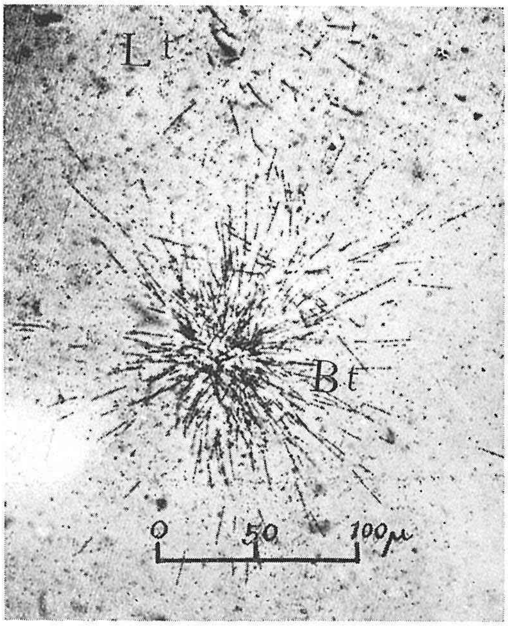

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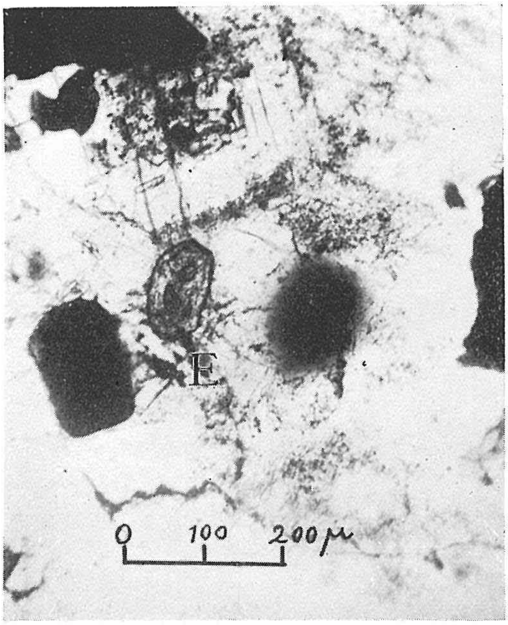

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