MEMOIRS OF THE COLLEGE OF SCIENCE, UNIVERSITY OF KYOTO, SERIES B,
Vol. XXIV, No. 2 Geology and Mineralogy, Article 2, 1957

The Radioactivity of Rocks and Minerals Studied with Nuclear Emulsion V

Radioactive Behavior of Granites

By

Ichikazu HAYASE

Geological and Mineralogical Institute, University of Kyoto

(Received July, 19, 1957)

Abstract

In the present paper the author tries to show the following points: how the contact metamorphism of a pre-existent rock with granite has caused migration of radioactive elements: the more the crystallization gets advanced in hornfels, the more radioactive materials it has absorbed from granite and consequently become more radioactive; while in just contact or near it the radioactive substances are concentrated in brecciated xenolith and dusty parts altered from cordierite porphyroblast.

Minute grains are greatly dependent, both in mineralogical varieties and in radioactive order, upon the type of granite (stock or batholith) which contains them and, also, upon the part of the rock mass (core or periphery) from which the sample is collected.

The quantity (or number) and the respective radioactive order of these grains stand in so simple a relation that it can be illustrated by a neat lognormal distribution.

What sort of radioactive element (whether thorium or uranium or both) the nucleus mineral contains is known from the nature (whether a ThC' ring or a RaC' ring) of the pleochroic halo printed in granitic biotite. The *Th–U* tendency, on the contrary, is deteriminable from the alpha tracks of different length emitted by the nucleus mineral and their divergent distributions. On this latter method it became obvious that, the later the stage of granite is, the more thorium and the less uranium the minute grains contain, as is the case with biotite itself. This uranium leaching and thorium storage must ever have become more remarkable in the later stages of the magmatic differentiation of igneous rock.

Introduction

Since the day of the epoch-making discovery of radioactivity itself at the close of the nineteenth century1), granite, the main constituent of the Earth's Crust, began to be studied from a new point of view. The studies were commenced first with the measurement of its Radium content which remains untill to-day the most precise of all the measurements of radioactive substances of rocks2), 3).

The distribution of radioactive materials in igneous rocks, especially granite4–8), is worth our researches, for

1) granite, being the chief constituent of the Earth's Crust, contains radioactive elements far more than any other rocks,

2) the radioactivity of such elements seems to offer us the clue to all the thermal phenomena in the Earth's Crust⁹⁾,

3) granite and granite pegmatite supply us *Th-U* minerals that enable us to determine the geologic age of rocks and minerals, and

4) granite stands, directly or indirectly, in some relation with the resources of fissionable materials.

Besides, the author hopes that these materials in granite may serve as the trace elements to granitic petrogenesis.

Now in granite these materials are distributed in

a) minute accessory minerals,
b) principal minerals of later stage and
c) deposit at the interstitials of crystals for leachable radioactive element¹⁰⁾.

The often related radioactive behavior of the granite¹¹⁾ as a holocrystalline rock is highly interesting both petrologically and geochemically. In a series of the author's previous papers, firstly, two types of granite, namely, stock and batholith, were divided from each other, so as separately to treat the minute radioactive minerals each of them contains⁸⁾; then the thorium content was deduced from the radioactivity which each of allanite grains often seen in granite is ever emitting¹²⁾; and finally, by pointing out the heterogeneous concentration of radioactive elements partially in granitic biotite, the peculiarity of their migrating concentration to the late deuteric stage was established¹³⁾.

But two matters were left still uncertain: that is, radioactive equilibrium and *Th-U* ratio. These two problems not only require much further investigations, but can hardly be pursued separately. To determine the *Th-U* ratio, therefore, minute crystals seeming to be already kept in that equilibrium were examined¹⁴⁾. This attempt, however, will not be complete, without our careful studies on the micro-distributions of these elements in minute grains. The heterogeneous distribution in each minute mineral¹⁵⁾,¹⁶⁾, which was sometimes alluded to, is worth our attention.

Further investigations of the radioactive distribution in just contact rocks and in xenolith facies, and of the variation of the paragenesis of minute radioactive minerals, will be doing ever-increasing contributions to our science. Our knowledge of the behavior of radioactive elements in granite will not fail to offer us an important clue to its petrogenesis in its post-orthomagmatic stage. In this paper a special heed was taken to the stage of *Th-U* deposition in granite. For the problem of the stage can not be solved as usual by a vague supposition that ascribes it to the pegmatitic. Uranium behavior in granite, as already clear from the hitherto achieved results concerning uranium ore deposites, is suggestive of its origin of much later stage than the pegmatitic. Of course, not so much complete solution, but some small advancement, of this problem is intended in this paper.

Radioactivity of hornfels

Shishitobi contact

Tanakamiyama granite (T. G.) and Palaeozoic shale very clearly come in contact at Shishitobi[17] where, as shown in a map (Fig. 1), the Seta river and the Ooishi brook meet. There stands, as the result of its resistance against erosion, a hill, having been formed by the hornfels along the contact plane on the Seta river bank slightly southward of the Shishitobi bridge.

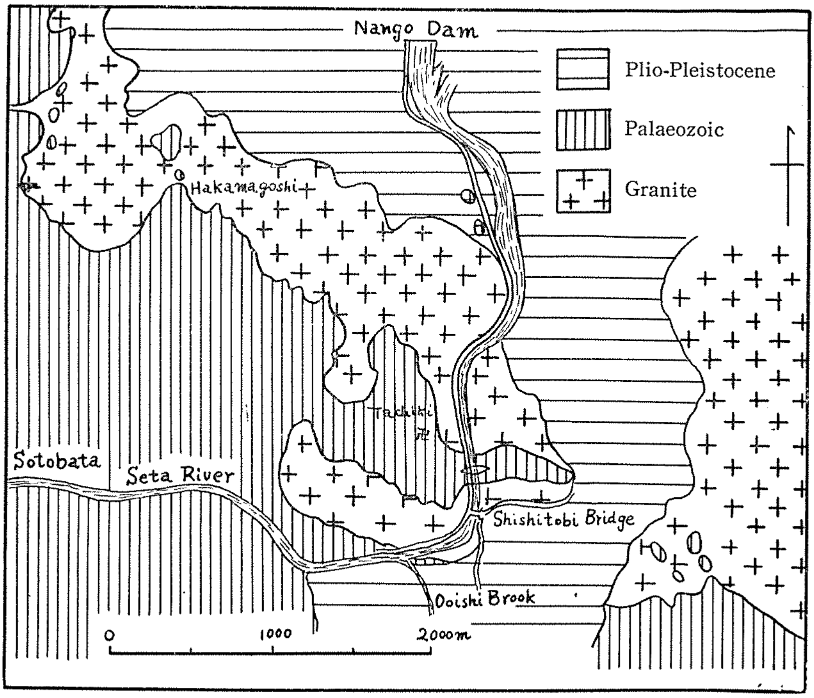


Figure 1 Geological map of Shishitobi and its vicinity

In its just contact facies, the hornfels and granite are separated from each other by a zone of aplite, which includes either pegmatitic parts or brecciated hornfels with a colorless rim (see Fig. 2a). Colorless aplite veinlets are also seen running into the hornfels (see Fig. 2b, 2c).

Fine and colorless grained is aplite especially along its contact part with hornfels, though not uniform in its breadth, but commonly from two to three millimeters wide. Next to aplite zone we see a pegmatitic one about ten centimeters in width, in which largely crystallized biotite lamellae are mixed up with feldspar. This pegmatite zone is followed by a coarse grained biotite granite including considerable

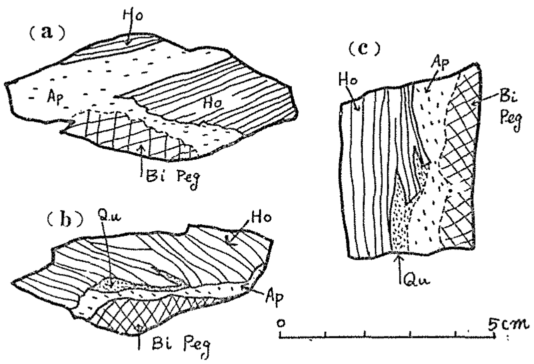


Figure 2 Specimens of contact rock (Ooishigawa)
Ho···Hornfels Ap···Aplite Qu···Quartz
Bi Peg···Biotite Pegmatite

radioactive elements, for instance, more than 3×10^{-12} grammes radium per gramme of rock[18].

In that just contact, the hornfels contains much biotite, many a fine grained tourmaline, and some oval muscovite clots (some three millimeters in size) which seem to have been changed from cordierite, but never fresh one. Fresh cordierite is seen for the first time in hornfels at a distance of a quarter or half a meter from the just contact.

The radioactive distribution was found in hornfels as follows:

a) The radioactive order stands very high in the regions where much biotite assemble, but the highest in the part of biotite and muscovite assemblages.

b) Rather low in the regions where muscovite alone assemble.

c) Generally speaking, radioactivity diminishes gradually from the just contact toward the hornfels.

d) Remarkably high activity observable on both sides of aplite veinlets starting from granite is absent in narrow veinlets.

e) The minute radioactive minerals found in hornfels are zircon-like, rounded, fine grains, a few of which, diverse as they are in shape, often seem as if clustered together; and this is peculiar to hornfels and never happens in granite.

D_α[19] means the total alpha track number which was being emitted from the polished surface of rock per centimeter square and per day. For example, granite involving some 3 grammes of uranium and 12 grammes of thorium per ton, wherever they are kept in a radioactive equilibrium, must be emitting, from its polished surface, alpha particles amounting to about $D_\alpha=60$.

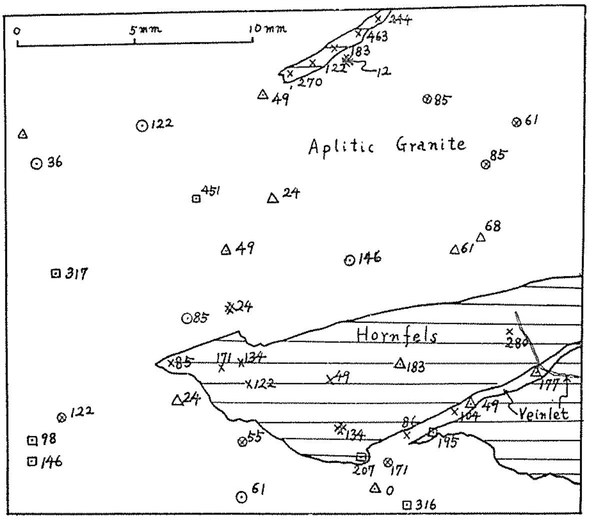
It is very noteworthy that a comparatively high radioactivity in Shishitobi contact rock, as seen in Figure 2, is brecciated hornfels surrounded with aplitic granite.

The track counting was performed at every microscopic field (9.84×0.01 mm square) by calculating point by point thought to require its radioactive value. The value thus derived, which is not the average D_α from the total surface of rock, informs us of the distribution of the radioactivity.

In this xenolithic hornfels (T. G. 770) radioactivity is the highest on both sides of the white aplitic veinlet intruding into the hornfels, especially at the entrance thereinto; not feeble ($D_\alpha=85$) at the edge of that hornfels and fairly strong ($D_\alpha=280$) at its dusty part; in aplite surrounding the xenolith quartz (about $D_\alpha=60$) and muscovite (some $D_\alpha=0$—12) are the feeblest, but albitic part ($D_\alpha=85$—145) is slightly stronger. A fragment of this xenolithic hornfels (the upper small part in

Fig. 3) is highly radioactive ($D_\alpha = 122—463$).

In xenolith radioactivity is strong in the region containing much zircon, and still stronger in the biotite region darkly clouded with imperfect pleochroic haloes due to its strong activity. The region full of quartz is feebly radioactive. We should notice that such exceptionally high radioactivity (D_α 177, 183) is scattered evenly in quartz. The larger xenolith seems less radioactive than the smaller one.



× Xenolithic Hornfels Matrix ⊗ Dusty Albite
⊙ Albite ▣ Biotite ♯ Muscovite △ Quartz

Figure 3 The radioactive distribution in the aplitic granite and its hornfelsic xenolith

(Shishitobi contact T. G. 770-1)
The numbers indicate D_α.

Similar in activity are aplitic veinlets in xenolith and aplite surrounding the xenolith. In aplitic part quartz is feeble ($D_\alpha = 0—68$), albite fairly strong, biotite still stronger (the average value out of 5 cases in aplite $D_\alpha = 265$), and brown biotite the strongest ($D_\alpha = 321$), but pale green one rather feeble, while in xenolithic one the $D_\alpha = 236$ in its mean value out of 5 cases. The result thereof is illustrated in Fig. 3 and 4: radioactivity stands rather high in the parts where biotite and muscovite, probably changed from cordierite, make an aggregate, and where biotite has formed somewhat large crystals, as is often the case with a druse pagmatite in granite.

In the just contact of hornfels, however, the radioactive order is strikingly high in its cloudy part, but rather low in noncloudy part mostly consisted of biotite tourmaline hornfels. This indicates that the formation of hornfels and the migration of radioactive elements into them took place in different stages.

That the radioactive elements were not primodial in the original xenolith, but supplied from the adjacent granite, is evident from the fact that, as shown in Fig. 3, 4, the radioactivity is remarkable especially along the aplitic veinlet that runs through the xenolith. If compared with this aplitic veinlet, minerals and rocks in paragenetic relation to it are divergent in their activity as seen in Fig. 3. In xenolith of biotite hornfels consisted of small grained biotite and quartz, in part, radioactivity stands 2 times as high as the aplite, and in dusty parts containing tourmaline 3—5 times as high as that. In aplite, however, quartz is the lowest, next comes feldspar, and biotite is the highest: 3—5 times as high as the average

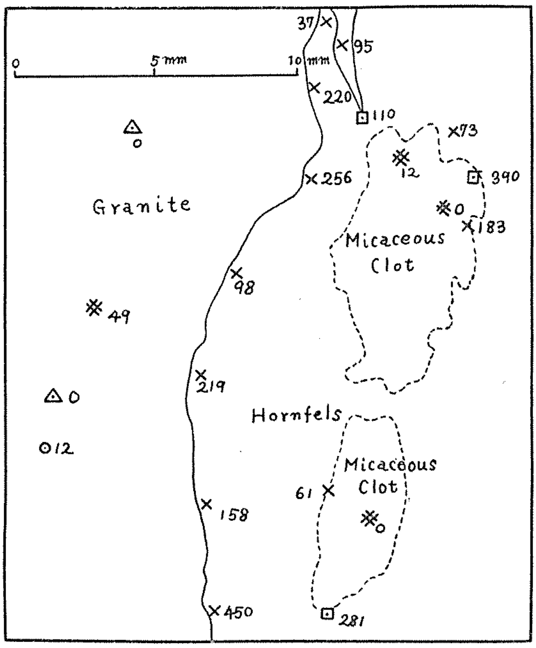


Figure 4 The radioactive distribution in the aplitic granite and its hornfelsic xenolith
(Shishitobi contact T. G. 770–2)
The numbers indicate D_{α}

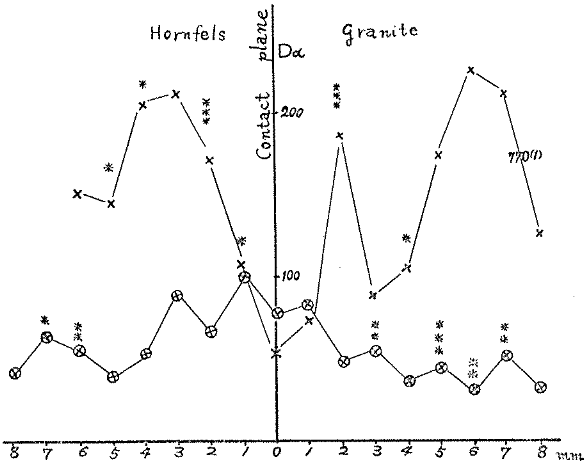


Figure 5 Microdistribution of radioactivity at the just contact
* Minute radioactive minerals

activity of the aplite. Muscovite is generally feeble[13]. Wherever hornfels and aplite are found in contact zone, the activity is much stronger (about 3 times indeed) on the hornfels side than on the aplitic rock. It is often said that the radioactive materials, removed from granitic aplite, were once concentrating in xenolith; and this tendency is found more striking where the xenolith is more coarsely grained. As to the manner of their distribution, in aplite and granite they are concentrated in minute accessory minerals, while in hornfels they are scattered all over. In hornfels, however, their concentration in certain minute grains, as is the case with granite, is seen where the crystallization (or granitization) is more advanced.

Radioactive behavior varies from granite to hornfels as seen in Fig. 3, 4 and 5. The former shows a thin section sample from Shishitobi contact of granite (T.G. 770–1, 770–2) with biotite tourmaline hornfels. Fig. 5 shows the radioactivity of the contact rock in which radioactive elements are not concentrated in some minute minerals but scattered all over. Here, granite and hornfels show little radioactive variation, except the slight feebleness in the just contact. If any noticeable variation is found in contact granite, it is because the rock is more coarsely grained and, therefore, some fluctuation in the distribution of alpha tracks is to be expected. As for the minute radioactive minerals, hornfels exceeds granite in their number, but falls short in their activity. Hence, if these minute minerals of strong radioactivity are taken into account, granite excels hornfels in its total activity.

In the other sample Shishitobi just contact hornfels (T. G. 770 α), a boundary is seen under the microscope between aplitic granite and biotite hornfels. The D_α counts 134.2 at that boundary in the hornfels, but diminishes as is removed from it; for instance, to 129.2 at a spot removed 5 mm from it and to 112.5 at another spot removed 10 mm from it. In most parts within 5 mm from it minute radioactive minerals (chiefly zircon) are comparatively abundant, and 15.5%—10.9% of the total alpha tracks were radiated from them, but in other parts removed 10 mm from it such minerals are less abundant and the percentage is reduced to 3.4% (wherever five or more alpha tracks are radiating from a point in the plate, we suppose there lies in the thin section a minute radioactive mineral corresponding to it.) Thus we see in the hornfels a tendency that its total radioactivity diminishes as is removed from the boundary, but not so abruptly as the number of such minerals does. Generally the radioactivity of a hornfels depends, indeed, upon that of its country rock (granite), but here in the almost perpendicular contact plane of Shishitobi granite the radioactive effect seems to extend over some meters thick, for example, the D_α was 78.3 at a point 4 meters from the contact.

The alpha radioactivity gradually increases in hornfels toward the just contact, and is either concentrated in certain minute minerals or so broadly scattered in the rock that the radioactive origin is hardly discernible. This increase is more striking in the case of concentration than of broad distribution.

Where the Ooishi brook joints the Seta river there lies a phyllitic hornfels in the same dip and strike as its contact plane with crushed leuco granite. This hornfels consists of two parts; the one is almost transparent tiny lens chiefly cordierite porphyroblast blended with muscovite and quartz, and the other dark, opaque part. D_α count in the former 82.2 and in the latter 74.4. This result contradicts, indeed, our expectation of the radioactive concentration always in the dusty parts of the rock. But it is no wonder, if we consider that the often said cordierite porphyroblastic part has been playing the part of drusy pegmatite.

Tachiki-kannon hornfels (T. G. 426) consists, as its thin section of rock (Fig. 6) shows us, of the following parts: the first is a biotite hornfels abundant in quartz, the second and the fourth are dusty biotite ones, while the third is mainly cordierite porphyroblast. The D_α is 41.8 in the first, 25.4 in the second, 137.7 in the third, and 19.7 in the fourth. The first and the third contain minute radioactive minerals, while the second and the fourth are devoid of them. Hence the astonishingly feeble activity of the second and the fourth is ascribable to the excessive concentration of the elements in the third.

The microscopic distribution of radioactive elements in a thin section of hornfels is clearly shown from a sample of contact rocks (T. G. 429-2) obtained from the

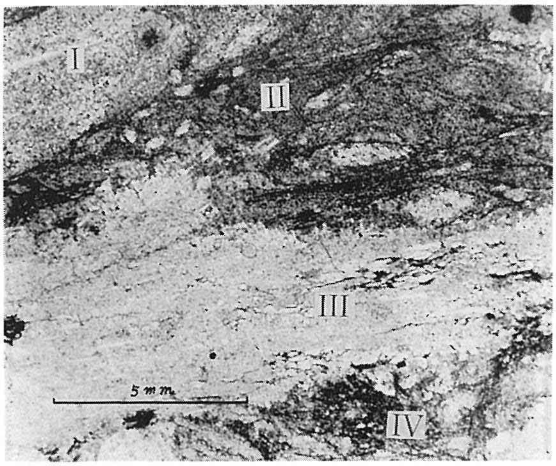


Figure 6 Hornfels of Tachiki-Kannon

roof pendant at the Tachiki-kannon. We perceive in this rock that the cordierite has been changed from place to place into the aggregates of muscovite. The radioactive order of this rock as a whole shows $D_\alpha = 46$, that of the muscovite aggregates changed from cordierite shows $D_\alpha = 182$, and that of the dark cloudy parts shows $D_\alpha = 292$. That is, in such a hornfels as this the radioactive elements are distributed most densely at the dark cloudy parts, next comes the altered cordierite, and the dilutest at the matrix. This suggests us no less than that in the formation of hornfels radioactive materials were concentrated at the dark cloudy parts rich in tourmaline and/or at the porphyroblast of cordierite, in other words, that the above-said parts were influenced by a solution containing radioactive substances, while the quartz and biotite constituting the matrix underwent no chemical change, despite of a recrystallization from shaly material.

Hakamagoshi roof pendant near Shishitobi (See Fig. 1) makes a boundary between Tanakamiyama granite and adjacent hornfels, or more correctly, the latter covers the former as the contact and, having suffered a silicification, is not very strongly radioactive, nor uniform in its alpha radioactivity (D_α) (See Fig. 7): 1) quartzite breccia 13.0 2) quartz biotite hornfels 23.7 3) biotite hornfels 25.9 4) silicified biotite hornfels 31.9. Other samples (T. G. 488—T. G. 490) abundant in quartz and removed several meters from the granite have the activity of $D_\alpha = 34.3$ and 30.2. That is, within several meters from the boundary this hornfels shows no big radioactive variation.

Figure 7 Brecciated hornfels of Hakamagoshi

Daradani contact

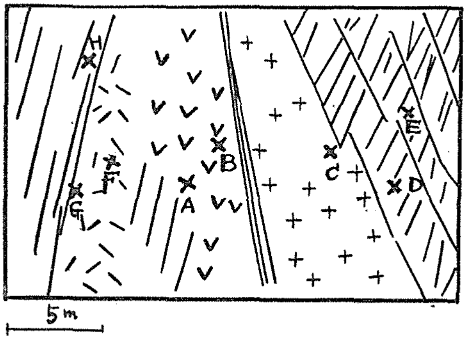


Figure 8 A cutting of Daradani pottery stone quarry

At Daradani, the western boundary of T. G., granite and Palaeozoic formation come in contact, where aplite has been much developed at the just contact and mined as pottery stone because of its scarce ferrugineity. The relationship of various rocks at the time of fresh cutting is given in Fig. 8 which illustrates how it traverses from leuco-granite and quartzite, which has suffered granitization, to hornfels.

Rocks of Daradani, Tanakamiyama, are:

A) In foot wall at the contact facies, fine grained quartzite which, having been changed from chert by virtue of a contact metamorphism, is devoid of its radioactive center, though slightly injected through by sericitic veinlets, along which no remarkable radioactivity is observed. This feeble radioactivity of $D_\alpha=18.4$ is due probably to its situation as a foot wall rock.

B) Muscovite leuco-granite which, having suffered a deuteric action, includes many radioactive centers, namely, either irregularly shaped, dusty, opaque materials or sometimes something like metamorphosed fine zircon grains. The radioactivity of this rock is $D_\alpha=94.2$, and 62 percent of this activities is due to the minute radioactive minerals. Between A and B there is no distinct boundary in the field observation. The micaceous secondary minerals—together with the dark red grains looking like hematite and formed especially between quartz and feldspar—are not only abundant in quantity, but also as strongly radioactive as about $T_\alpha=0.3$. Quartz at Shishitobi contact, Tanakamiyama, often includes zircon-like rounded minerals devoid of their idiomorphic crystals, metamict, similar to zircon in their refractive index, and with the radioactivity of some $T_\alpha=12$; they are too fine in size to be identified as thorite, although their track length suggests us of their thorium content. Slightly more alpha tracks than average are being emitted from the potash feldspar parts which has suffered a considerable kaolinization.

C) The wall rocks, at which leuco-granite, granitized hornfels and biotite granite come in contact with each other, is fairly strong in radioactivity. As to the granitic part, the radioactivity stands fairly high (about $D_\alpha=234.7$) in very dusty albite and biotite parts, but quite low (lower than 10) in slightly cloudy orthoclase and quartz parts, and moderate (some 85) in a part full of sericite. As for the coarse grained granitized hornfels, the clot of biotite is the most radioactive (about 345), while the matrix part, where biotite and quartz are mixed up, is very divergent in activity (some 137); similar in radioactive order is also the part full of muscovite (probably changed from cordierite).

D) Next to C (biotite hornfels $D_\alpha = 60.1$) well developed porphyritic quartz which shows fairly high radioactive order. Noteworthy is the slight radioactivity observable at the dark parts of limonitic deposit. In radioactivity quartz part is feeble $D_\alpha = 12$) but, if the biotite therein is coarse grained with cloudy dust, much stronger (even $D_\alpha = 122$). If the biotite is in intergrowth with reddish limonitic minerals, then often as strong as 342. Generally, slightly feebler (about 42) in smaller grains of quartz and biotite, and very feeble (less than 10) in still finer grains of quartz.

E) Leuco-granite, just like the aforesaid B, has its radioactivity as $D_\alpha = 139.0$, but shows no distinct limit in its outcrop from D. Biotite is fairly radioactive (some 183), quartz very feeble (less than 10), potash feldsper and albite fall just between the former two, unless some minute but strongly radioactive minerals is included in the latter, as is sometime the case with them.

F) As for the biotite hornfels, the radioactivity is strong ($D_\alpha = 84.4$) in a part rich in biotite, and still stronger (even 212) especially in another part full of red limonite-like material (its radioactive origin must be of Thorium, because 25 per cent of the total tracks are over 20 microns), but rather feeble (some 12) in a part rich in quartz, and slightly stronger (nearly 49) in another part full of muscovite probably changed from cordierite.

G) In cordierite hornfels including tourmaline, the radioactivity was found to be some 18 in the cordierite part, but generally about 30.0 in matrix part (biotite hornfels) and stronger than in the former.

H) That quartzite slightly containing biotite has been metamorphosed probably from chert, is completely the same in its petrological character as what was stated under the head A, and very feebly radioactive (8.12).

In brief, the redioactive varieties were found with Daradani hornfels:

1) Where there is abundant quartz, it is weaker than $D_\alpha = 10$.
2) Where there is full of biotite, it is mostly stronger than $D_\alpha = 200$ and, if mixed up with reddish limonite, it amounts even to more than $D_\alpha = 600$.
3) In muscovite clots probably changed from cordierite, it is stronger than in quartz but feebler than in biotite clots. Fresh cordierite hornfels is also generally feeble in activity.

The $D_\alpha = 26.5$ is the quartzite contact rock (S. G. 634 Suzuka granite) near Suzuka Pass, which containes much quartz and almost entirely lacks biotite. But we can find in other parts of this quartzite that the activity shows $D_\alpha = 31.9$ in red-brownish parts (probably alteration product from biotite). The quartz brecciated biotite hornfels (S.G. 635), $D_\alpha = 44.6$—42.4 in activity, much resembles Hakamagoshi rock both in appearance and in radioactivity. As for the mylonitic rock near that Pass, the activity is not much different ($D_\alpha = 129.1$) from that of the granite in low grade mylonite (S.G. 636) still retaining its granitic texture, but much lower ($D_\alpha = 59.0$) in leuco-granite (S. G. 625) whose biotite was altered under the influence of breaching. But such a rock contains reddish stains whose activity is remarkably, though not invariably, strong: $D_\alpha = 357$, 780, 1726, 1892 and so on, and such a thing is never expected in quartz and feldspar.

The radioactive degree of contact metamorphic rocks of Tanakamiyama granite can be designated on the autoradiographic method by D_α as shown in Table 1. It generally stands lower than that of original granite but higher than that of sedimentary rocks of Palaeozoic Formation. In the thin sections of rocks we perceive that the radioactive elements are scattered more uniformly in hornfels than in granite. But it is likely that the coarser the quartz, feldspar and biotite grains in hornfels

Table 1 Radioactivity of Tanakamiyama hornfels and others

Locality	Rock	Radio-activity D_α	Sample
Daradani	hornfels	60.1	D
		84.4	F
		30.0	G
	quartz hornfels	10.0	C
	quartzite	18.4	A
	quartzite	8.4	H
Hakamagoshi	hornfels	23.5	488
	hornfels	34.3	489
	hornfels	30.2	490
Tachiki	hornsels	73.8	431
	hornfels	48.1	426
	hornfels	60.0	424
	hornfels	65.2	420
	quartz hornfels	84.0	767
	quartzite	69.0	767
Shishitobi	hornfels	82.0	670
	hornfels	74.4	670
	hornfels	30.0	673
	hornfels	125.5 (3)	770α
	hornfels	179.9 (2)	772
Kurahone	hornfels	39.1 (2)	216
	quartzite	33.7	219
	quartzite	40.8	225
	hornfels	81.6 (2)	1114
Tashiro	hornfels	50.8 (2)	821
	hornfels	51.6	823
	hornfels	55.7	825
	hornfels	51.0	830
	quartzite	44.9 (3)	822α
Ryuozan	hornfels	47.0 (3)	811
South Konshoji	hornfels	47.4	715
	silicified hornfels	48.2	710
Ootorii	hornfels	45.8	793
Hata	hornfels	51.0	830
Asamiya	quartzite	21.3	841
Kitashirakawa	quartzite	24.3	30–1
	quartzite (red stains)	64.3	30–2
Hira Takashima	hornfels	94.2	
Nishitani Shizo Hyogo Pref.	grey wacke	18.2	
Narutaki Kyoto City	siliceous slate	26.9	
Yunoyama Mie Pref.	hornfels	62.7 (2)	
	quartz hornfels	31.6 (3)	
Shinooka Aichi Pref.	hornfels	43.1	
Ajioka Aichi Pref.	hornfels	44.3	
Suzuka Pass	quartzite	65.0	634
	quartzite	31.9	623
	hornfels	47.4	637
	hornfels	44.6	635
Gyojayama	quartzite	52.6	A
Sakuratenjin	quartzite	56.1	B
Tsuge-Kabuto Mie Pref.	hornfels	24.4	851
	hornfels	70.7	852
	hornfels	17.8	848

are, the more abundant are zircon and other minute minerals into which the radioactive elements are condensed. The radioactivity of hornfels depends 1) upon that of the original granitic rock and 2) upon its distance from the just contact with granite and their relative situation.

The radioactive characteristic of minute minerals in granite

It was often said in previous papers[16] (I[8] and II[12]) of the great varieties of these minute inclusions which differ in the amount of radioactive elements, and in other papers[14],[20] of their divergent *Th-U* tendencies.

Now, are the volumes and the radioactivities of these minute grains kept in the same relation through all different samples? Here let us take up zircon, monazite and some other accessory minerals and, for convenience's sake, give up coarse grained and feebly radioactive ones like allanite, sphene and so on.

For, if a coarse grained allanite or sphene, say 1 mm in diameter, in a thin section was compared with a much finer zicon or monazite crystal, our results would be little trust-worthy.

In Fig. 9, which illustrate the volumes and radioactivities (T_α) of the above said minerals, the abscissa indicates their radioactive order, for instance, $T_\alpha = 1.26$ is to be found at the point of 1.2, while the ordinate their respective volumes.

Note: the number of sections constituting a histogram informs us of the number of minute grains of the same radioactive order. (Grains higher in radioactivity than $T_\alpha=14$ were as follows: $T_\alpha=15.50$ (0.041×0.01 mm²), $T_\alpha=16.80$ (0.09×0.01 mm²), $T_\alpha=26.10$ (0.014×0.01 mm²), $T_\alpha=29.40$ (0.012×0.01 mm²).

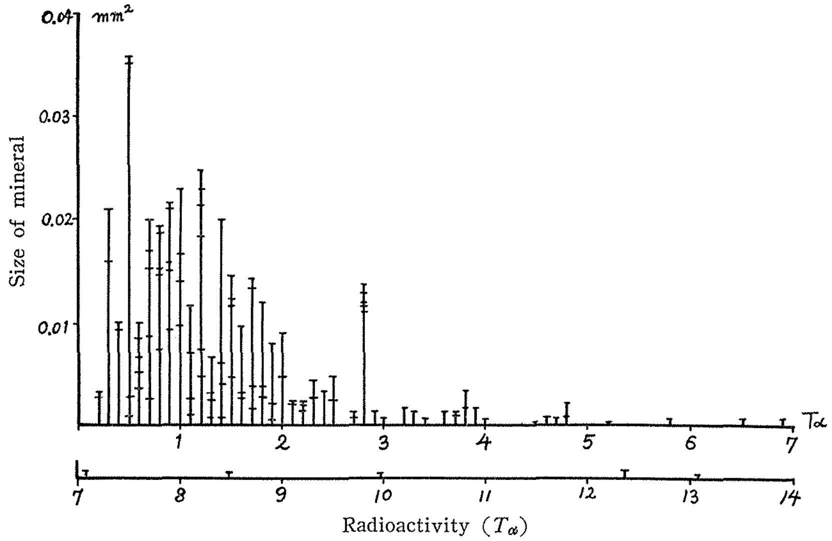


Figure 9 Grain size and radioactivity of minute radioactive minerals (Shishitobi contact rocks).

Eight Shishitobi (Tanakamiyama) samples (all lacking allanite) are exemplified in Fig. 9, from which we can clearly perceive a lognormal distribution between the appearance frequency of those minute minerals and their radioactive order. Though rare and generally fine grained, minute crystals of extraordinary high activity are found among a multitude of them.

Now, these minute minerals can be divided into some groups of different radioactive order. Then, which of these groups is *most* contributing to the total radioactivity of the sample? From Fig. 9 it is evident that the group of some $T_\alpha \doteqdot 1.4$ is the most dominant radioactive source of it; that next come the groups from $T_\alpha = 0.5$ to 5; and that other groups higher and lower than that play no important part in constituting the rock radioactivity. Especially in granitic rocks lying near the contact, zircon, monazite and xenotime grains belong to the strongly radioactive group, though not uniform in their *Th-U* ratio. In number zircon seems far to surpass monazite and xenotime.

As for Shindenba quarry, xenolith-like granite sample contains 1.72×10^{-12}g/g radium, and feebly radioactive zircon grains are found there in the dark ovoid xenolith. (Compare Fig. 9 with Fig. 10. Generally the rock radioactivity depends chiefly upon those which are represented by the densest and highest parts in Fig. 9 and 10). Four granitic samples (a, b, c and d) were collected from near Shishitobi contact, and their minute inclusions under $T_\alpha = 7$ are shown in Fig. 11, while those over $T_\alpha = 7$ are five grains: (a) $T_\alpha = 8.5$ (0.029×0.01 mm^2), (b) $T_\alpha = 12.4$ (0.081×0.01 mm^2), $T_\alpha = 16.8$ (0.094×0.01 mm^2), (c) $T_\alpha = 10.04$ (0.035×0.01 mm^2), (d)

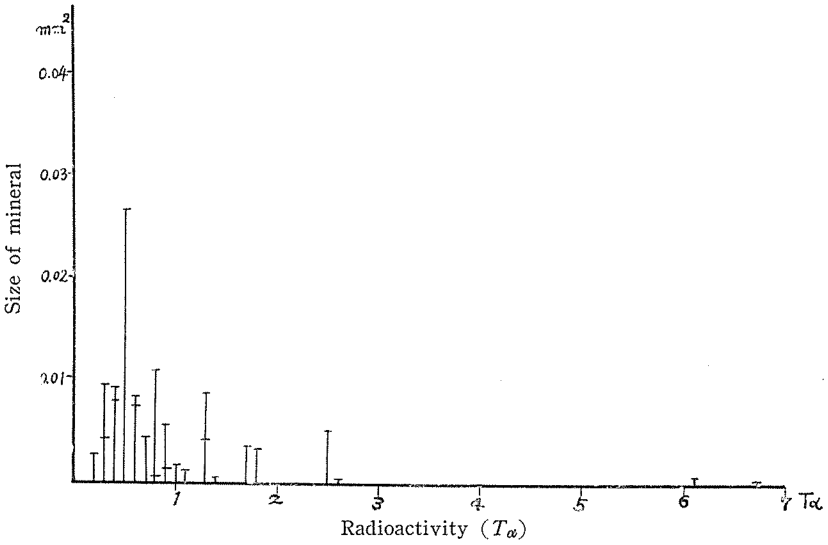


Figure 10 Grain size and radioactivity of minute radioactive minerals (T. G. 665, 666)

$T_\alpha = 7.14$ (0.037×0.01 mm^2). Now it becomes evident from Fig. 11 that (a) and (b), farther removed from that contact, are much wider in radioactive range than (c) and (d), in which feebly radioactive minerals are abundant. As a rule, these rocks are highly radioactive and, according to Asayama[18], sample (b) (T.G. 224) contains 2.07×10^{-12}g/g of radium.

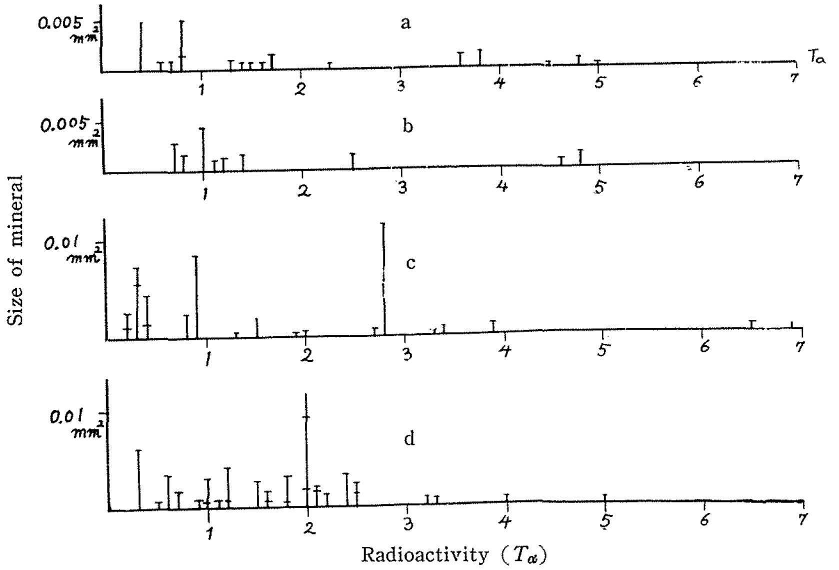


Figure 11 Grain size and radioactivity of minute radioactive minerals (T. G. 674, 224, 211, 668)

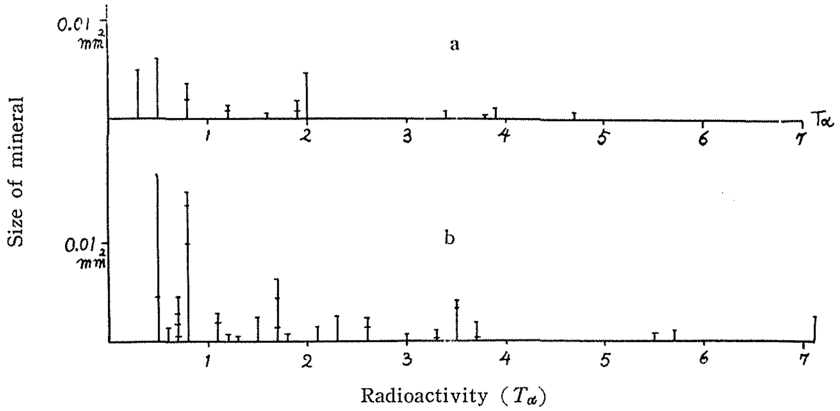


Figure 12 Grain size and radioactivity of minute radioactive minerals (T. G. 890, 286)

As for Akao (a) and Nango (b) samples at the north west part of Tanakamiyama, see Fig. 12 which, like Fig. 11, informs us that highly radioactive grains are existent in them, namely, so high as $T_\alpha = 15.6$ (0.041×0.01 mm^2) in the former and $T_\alpha = 20.3$ (0.056×0.01 mm^2) and $T_\alpha = 14.7$ (0.073×0.01 mm^2) in the latter.

Fig. 13, 14 and 15 illustrate Mikumo granite samples whose minute minerals, with very few exceptions, are of a quite uniform tendency ($T_\alpha < 1$): a tendency entirely different from that of Tanakamiyama granite (Fig. 11, 12 and 20). This is due, as is evident from the radium content distribution map,[18] to different radium content in them, namely, over 1.5×10^{-12} g/g in Tanakamiyama granite, but under 1.0×10^{-12} g/g in Mikumo samples. In such granite whose radium content is under 1.0×10^{-12} g/g, as a rule, the minute zircon grains buried in biotite flakes are their sole radioactive inclusions.

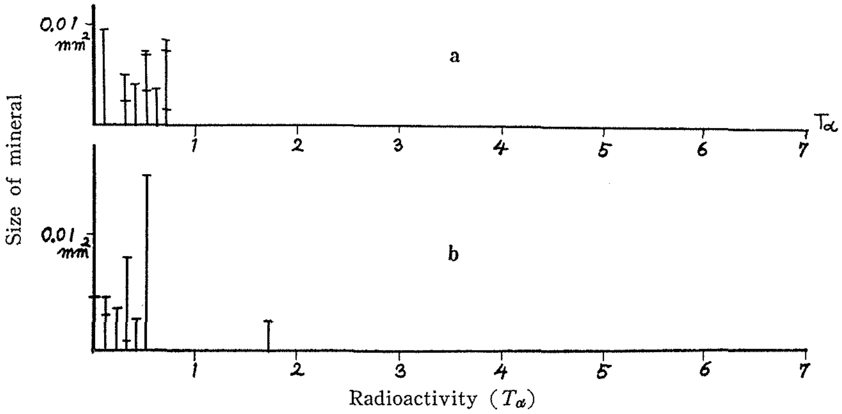


Figure 13 Grain size and radioactivity of minute radioactive minerals (M. G. 529, 654)

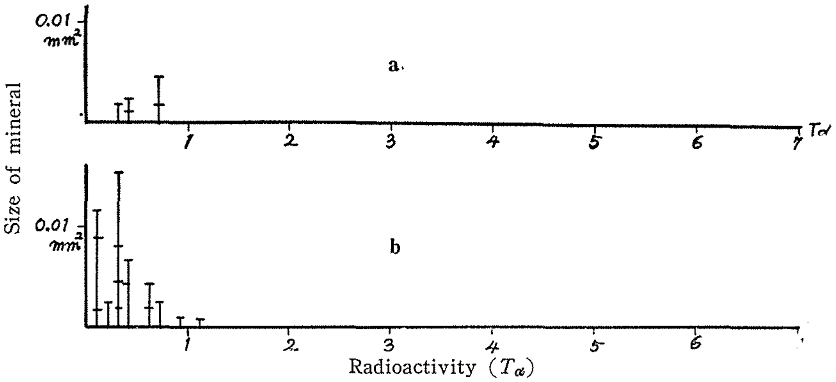


Figure 14 Grain size and radioactivity of minute radioactive minerals (M. G. 323, 521)

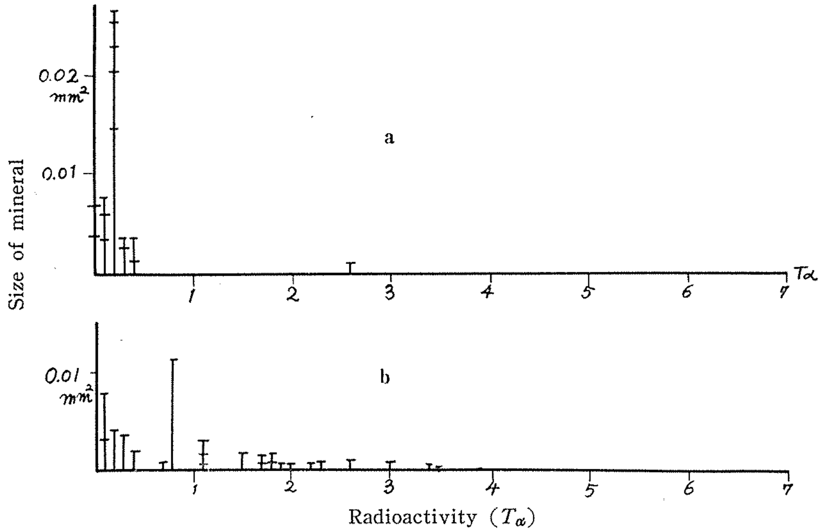


Figure 15 Grain size and radioactivity of minute radioactive minerals (M. G. 336, 335)

Somewhat aliniated from this rule, nevertheless, are Mikumo samples (see Fig. 15b) which were invaded through by some veinlets of pegmatitic solution greatly to affect the radioactive content of their minute inclusions (over $T_\alpha > 7$ grains: $T_\alpha = 10.5$ (0.06×0.01 mm²) $T_\alpha = 10.6$ (0.012×0.01 mm²)). As for their distribution, see Fig. 5 in report I,$^{8)}$ in which it is quite noteworthy that, the population of grains and their radioactivity are the least in granitic pegmatite, more in adjacent schistosed granite, and the most in their contact part.

Samples (a) (M. G. 529) and (b) (M. G. 654) in Fig. 13 much alike in their tendency and in their petrographic characteristics were exemplified in Report I under the head of Mikumo type granite; sample (a) (Fig. 14) illustrates biotite syenite known as Murayama pottery stone. The fact that this pottery stone belonging to so different a sort of rock shows still a quite similar inclination, simply because it lies near these granite, deserves our attention. Sample (b) shows the result from two thin sections of M.G. 521 ($Ra = 0.53 \times 10^{-12}$ g/g).

Ootorii samples T.G. 712, 750 and 778 (see (a) in Fig. 16) containing minute crystals surpassing $T_\alpha = 7$ in their activity: $T_\alpha = 8.6$ (0.0135×0.01 mm²) $T_\alpha = 10.0$ (0.0165×0.01 mm²) $T_\alpha = 13.4$ (0.371×0.01 mm²) $T_\alpha = 14.8$ (0.0427×0.01 mm²) and $T_\alpha = 16.0$ (0.073×0.01 mm²), are similar in its tendency to Tanakamiyama granite (T. G. 712—1.86×10^{-12}g/g *Ra*, T. G. 750—1.48×10^{-12}g/g *Ra*) Samples 802 and 800 (see (b) in Fig. 16) have much in common with Mikumo type granite, though in sample 800 zircon grains contained in biotite alone are $T_\alpha < 0.6$ and more radioactive inclusions are found in quartz and feldspar. (T. G. 800—0.92×10^{-12}g/g *Ra*).

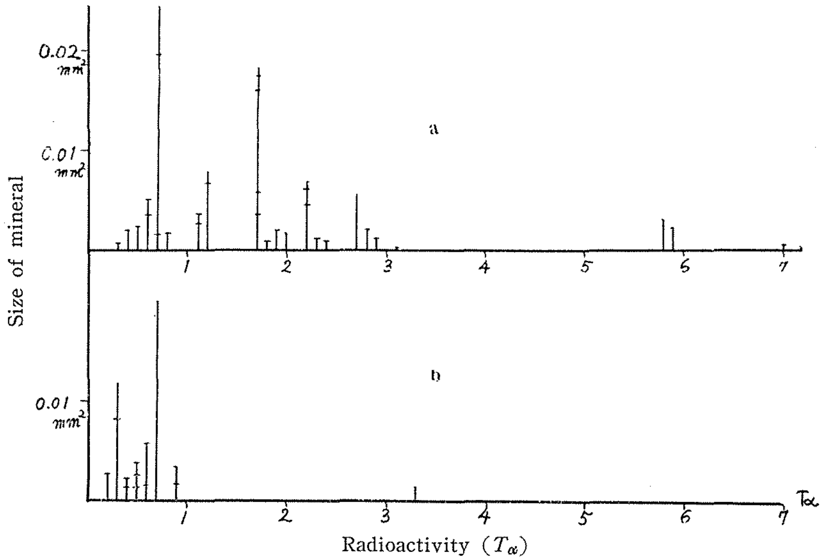


Figure 16 Grain size and radioactivity of minute radioactive minerals (T. G. 712, 750, 778 and T. G. 800, 802)

Such a difference as this, despite their related granitic mass as well as their similar microscopic appearance, seems to be due, as already mentioned in Report I, to their different rock types, namely, stock (like Tanakamiyama granite) versus batholith (like Mikumo granite). (The high radioactivity of Tartiary granite is

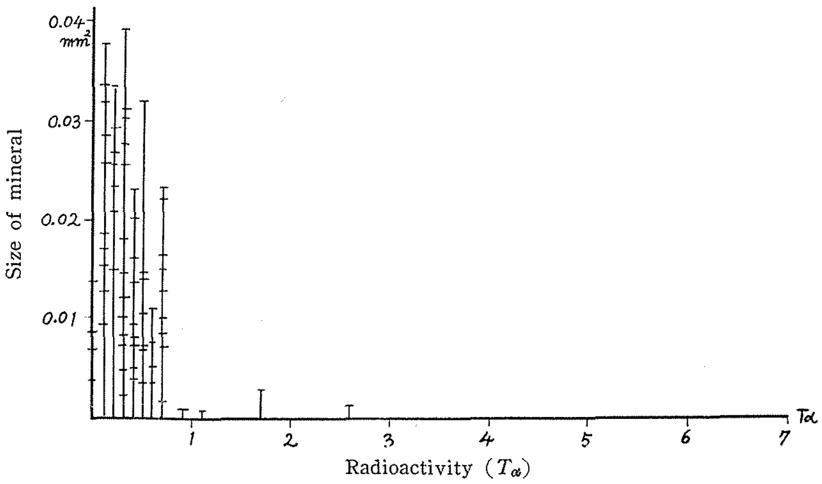


Figure 17 Grain size and radioactivity of minute radioactive minerals (Ryoke type granite)

attributed to the fact that it belongs to the stock type, and available samples are from the outcrop where the stock apex is exposed to our eyes.) Compare (a) with (b) in the Fig. 16: what was absent in Fig. (b) is added in Fig. (a); and Fig. 13, 14 and 15 except Fig. 15 (b) are summarized in Fig. 17. The radioactive characteristics of minute inclusions in Ryoke type granite will be quite clear from comparison of the two Fig. 9 and 17.

Schistosed or not as they are, Konze, Asamiya and Kabuto rocks—adjacent to Mikumo granite and all belonging to the Ryoke type—are much resembling one another (see Fig 18). By the way, Asamiya sample (M. G. 843) near the contact is rather rich in radium (order of stock type granite) for a granitic rock of batholith type, but in its radioactive inclusions nothing distinguishes it from the rest of the Ryoke type granite (Fig. 19a).

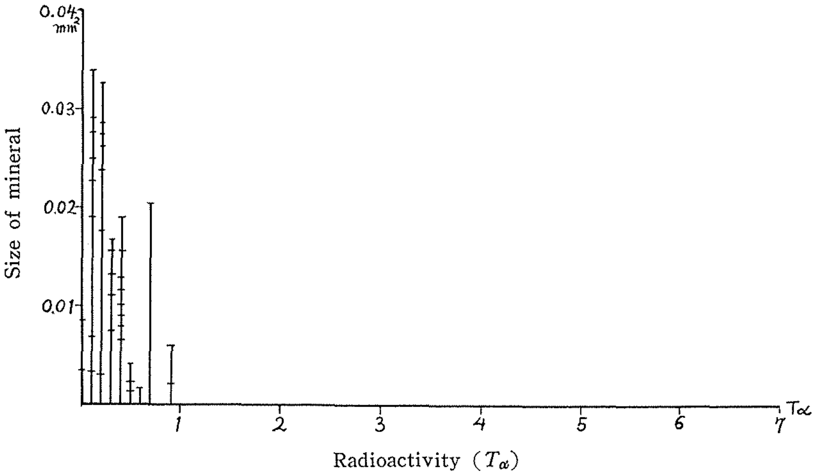


Figure 18 Grain size and radioactivity of minute radioactive minerals (Mikumo Granite 4 thin sections)

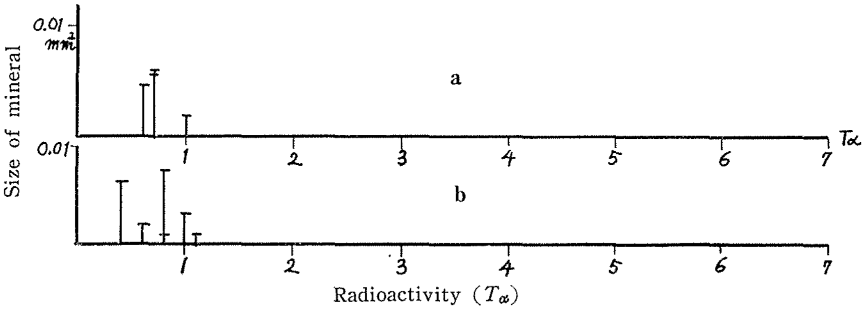


Figure 19 Grain size and radioactivity of minute radioactive minerals (M. G. 843, 466)

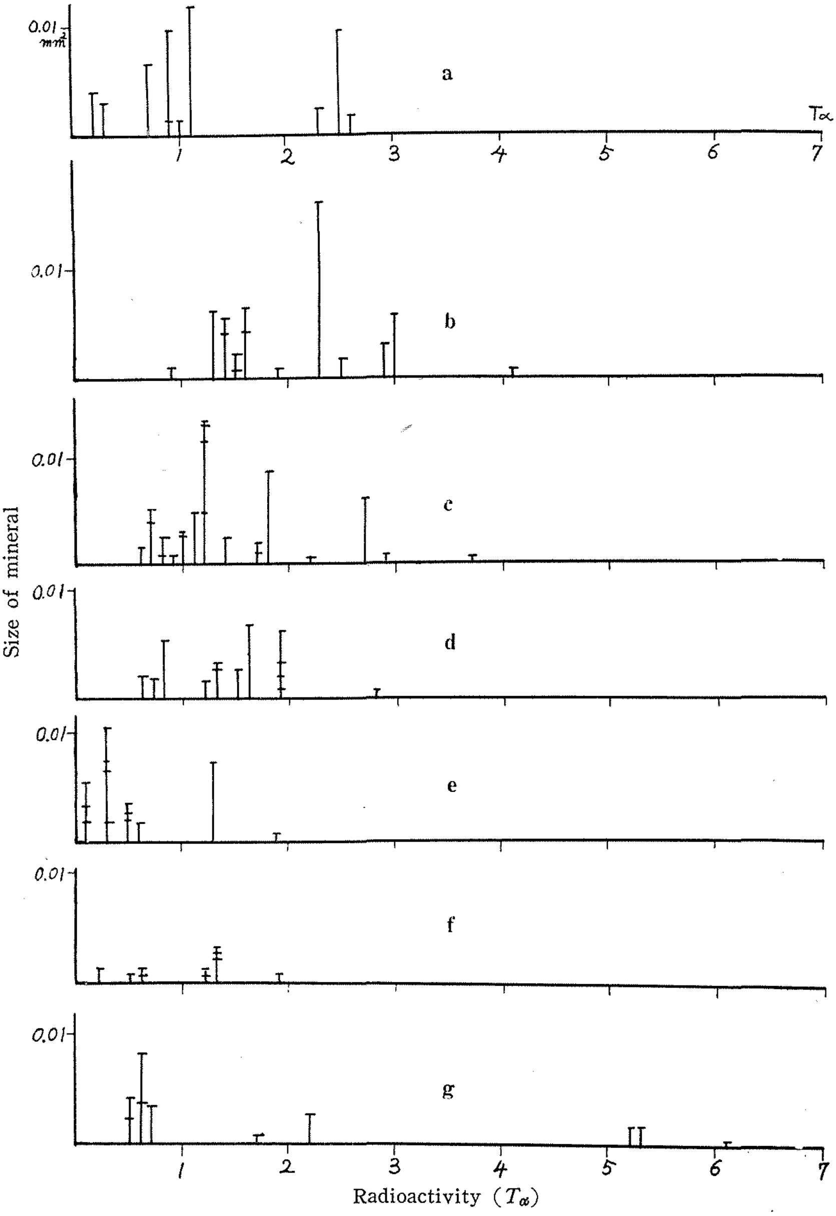


Figure 20 Grain size and radioactivity of minute radioactive minerals
(a) T. G. 550, (b) T. G. 812, (c) T. G. 440, (d) T. G. 738,
(e) T. G. 707, (f) T. G. 271, (g) T. G. 461

Fig. 20 illustrates some Tanakamiyama granite samples (a—550, 1.87×10^{-12}g/g *Ra*, b—812, c—440, 2.06×10^{-12}g/g *Ra*, d—738, 3.23×10^{-12}g/g *Ra*, e—707, 1.81×10^{-12}g/g *Ra*, f—271, 1.75×10^{-12}g/g *Ra* and g—461). But, as for radioactive minerals over $T_\alpha=7$: b—$T_\alpha=18.7$ ($0.023\times0.01\,\text{mm}^2$), $T_\alpha=21.9$ ($0.194\times0.01\,\text{mm}^2$), c—$T_\alpha=21.5$ ($0.014\times0.01\,\text{mm}^2$), $T_\alpha=29.4$ ($0.012\times0.01\,\text{mm}^2$), e—$T_\alpha=26.4$ ($0.63\times0.01\,\text{mm}^2$) and g—$T_\alpha=23.0$ ($0.043\times0.01\,\text{mm}^2$). Despite its 1.75×10^{-12}g/g radium content, in sample f (from near Suishyodani pegmatite) rather few minute crystals are radioactive, probably because of the abundance of coarse grained secondary radioactive minerals. In sample e (T. G. 707) minute minerals under $T_\alpha=0.6$, all contained in biotite, are mostly zircon and the rest are allanite. (In Tanakamiyama granite, as is the case with this sample, radioactive allanite grains are seen only in the xenolith).

Minute radioactive minerals and their pleochloic haloes

As already stated in a previous paper (IV)(14), the thorium and uranium content of minute radioactive minerals contained in thin sections of granite can be measured on our autoradiographic method. In other words, so far as the radioactivities of these minute minerals are in their radioactive equilibrium, the distribution of alpha tracks of different length informs us the *Th-U* ratio. But are such *Th-U* values thus measured really reliable? To answer this question, let us turn to the pleochroic haloes found in biotite flakes in granite.(21), (22), (23), (24)

By the way, the minute radioactive minerals in granite thin sections which we now deal with are of 10—100 microns in diameter and mostly of 30—50 microns in size, while the thin section which contains them is about 30 microns thick. Therefore, the most preferable for our measurement are the minute radioactive minerals of at least over 30 microns in dimensions.(24)

Such a radioactive mineral that happened, early enough in geologic age, to be contained in a biotite flake must have created around itself a pleochroic halo. And whether the halo is of uranium or of thorium or of binary origin is known by the peculiar size of that halo. For, a thorium halo of *Th C′* origin is about 41 microns in radius, that of *Th A* some 27 microns, while an uranium halo of *Ra C′* origin is about 32 microns in radius and that of *Ra A* some 23 microns. This enables the semiquantitative determination of the thorium and uranium ratio now in question.

The author took up a minute mineral whose thorium and uranium contents had already been known on some other methods and then made an autoradiographic examination of the respective distributions of alpha tracks of different length. (The ET-2E photo plates then used had 50 microns thick emulsion.) The ratio of the number of alpha tracks longer than 30 microns to the total track number was found, in the case of an allanite grain (Kitashirakawa granite) whose radioactive content was mostly thorium, 5.4% but the case of a zircon (the same granite) whose radioactive substance was greatly uraniferous, 1.7%.

But the *Th-U* tendency in a nucleous mineral (Gyojayama granite) which carries a thorium halo around it is illustrated in Fig. 21 and Table 2 in which the above said ratio is most often falling between 5.0 and 5.5 per cent. Even after the possible fluctuation caused by the comparative scarcity of alpha tracks longer than 30 microns has been taken into consideration, this very cleary informs the tendency that thorium is the unique radioactive content therein.

Otherwise, Table 3 illustrates *Th-U* composite haloes and uranium ones. The often said ratio is smaller than 2.4% in the latter, namely, alpha tracks longer than 30 microns are found less than one; once 2 in 110. Therefore, if the exposure is prolonged till the total track number may be some 200, the ratio in question is supposed to be nearly 1.7%.

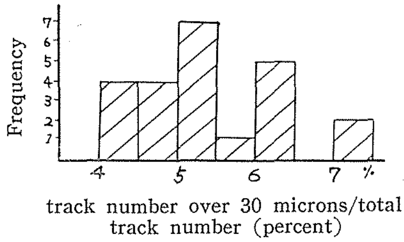


Figure 21 Track length distribution from the nuclei of thorium haloes

Table 2 Thorium haloes and their minute nuclei (Kameoka granite)

Sample	Total alpha track number	Track number over 30 microns	Ratio %	Radio-activity T_α
Yunohana	83	4	4.82	1.30
Y 4	68	3	4.41	0.96
	20	1	5.00	1.55
	48	3	6.25	1.13
	180	9	5.00	2.78
	151	6	3.97	2.07
Y 3	64	3	4.70	2.90
	36	2	5.56	1.06
	98	6	6.12	2.65
	93	6	6.45	1.37
	93	5	5.38	3.59
	114	7	6.16	3.52
Y 5	104	5	4.75	3.06
	46	2	4.35	4.26
	37	2	5.40	1.52
Y 9	160	10	6.26	2.47
	81	6	7.40	6.95
Y 6	56	3	5.36	4.05
	71	5	7.04	3.04
	95	5	5.26	11.0
	67	3	4.48	3.45
Y 7	81	4	4.94	5.55
Y 16	58	3	5.17	3.70

Table 3 Uranium haloes, composite haloes and their minute nuclei (Kameoka granite)

Sample	Total alpha track number	Track number over 30 microns	Ratio %	Radio-activity T_α
Kojin (a)	57	1	1.75	0.60
	46	0	0	0.32
	52	0	0	1.47
	15	0	0	1.16
	40	0	0	3.43
Y 3	50	1	2.0	3.28
	49	1	2.1	6.97
	25	0	0	11.5
	38	0	0	4.22
	48	1	2.1	0.74
Y 6	102	1	1.0	4.19
Y 12	41	1	2.4	4.75
	18	0	0	2.28
Y 16	110	2	1.8	0.97
Y 16*	112	3	2.68	2.08
Y 5 *	200	4	2.00	4.11
	31	1	3.30	4.57
Y 3 *	49	1	2.10	6.97
Y 4 *	123	4	3.25	2.37

* composite haloes

Now come the composite haloes (see, also, Table 3) which occupy a medium position between the genuine uranium and the pure thorium mineral. And all this assures us of the reliability of our track length method concerning the *Th-U* tendency in each minute mineral. In other words, if its mineral size, its weathering and metamorphism, the total alpha track number ever emitting from it, are taken into consideration, its *Th-U* ratio will be quite evident from the peculiar distributions of alpha tracks of different length. The rate of alpha tracks longer than 30 microns among the total track number amounts, for instance, to about 17.8% in allanite (Kitashirakawa granite) whose radioactive tendency in chiefly thorium, but to 6.4% in zircon grains (the same granite) in which uranium is its main radioactive substance. It counts 7.7—10.0% in minute grains (Tanakamiyama granite: T.G. 800) each of which carries composite haloes: a *Ra C'* halo (32 microns in radius) includes inside it another faint *Th A* halo (27 microns in size). This clearly informs us the great supremacy of uranium content over thorium. It shows, on the other hand, 8.6—9.5% in other grains (the same granite: T. G. 895) which also carry composite haloes: but this time a faint *Th C'* halo is seen around an overexposed *Ra C'* halo. This also affirms us the same supremacy of uranium over thorium.

Th-U tendency of the minute accessory minerals

It was stated in another paper (IV)⁽14⁾ that the *Th-U* ratio in various minute minerals contained in granite are rendered measurable by counting the distributions of different alpha track length peculiar to each of these two elements. (Before alluding some results concerning these ratios, it must be remembered that the values given here indicate the ratios, not of the real quantities of, but of the alpha activities emitting from, these two elements contained in minute grains. Hence, for example, *Th*-36 means that 36% of the total activities are being originated from *Th* and 64% are from *U* content.) Many an autoradiographic attempt⁽15⁾,⁽35⁾ was done to know the ratio between uranium and thorium content by the track distribution of different length. But, so far as minute radioactive minerals are concerned, two assumptions are essential, namely, the uniform distribution of the radioactive elements in those grains and the radioactive equilibrium of the active elements in them. The former requires a careful microscopic observation of the thin section of rock containing them. As for their inequilibrated condition, the matter is two-fold: in case of an equilibrium soon to be restored we have to observe only the emanating power from the minerals, but in case of an equilibrium to be restored in numerous years the uranium radium ratio becomes more important.

Specimens availed from Shishitobi contact of granite and hornfels (T.G. 770) show that fine zircon grains are contained in the above said aplite. (See Fig. 18, p. 268 and p. 266 in report IV.)

It was already made clear from allanite samples⁽12⁾ that higher percentage of this ratio suggests us that, in that grain, thorium supersedes uranium in amount. In minerals whose radioactive element is chiefly thorium, in fact, the percentage lies between 16.6% and 19.1%⁽12⁾.

It is usually said that the granitic zircon contains various *Th-U* ratio⁽25⁾, but we do not know their petrological meaning. Here in this case, it is evident that *Th* is, qualitatively, incomparably higher in these zircon grains. In this respect they are much different from Kitashirakawa zircon grains.[14] Though it is hard to explain why the percentage is so high as 34.7% in one of them, these zircon must be rich in thorium. Fig. 14 in the report IV[14] illustrates the *Th-U* tendency observed in

Table 4 Thorium uranium tendency of the minute minerals in granites

	Sample Number	Locality	Host Mineral	Total track number	The track number longer than 20 microns	%
a	295	near Daradani	biotite	81	9	11.1
				70	8	11.4
	697–2	Konze	biotite	107	5	4.7
	750	Ootorii	quartz	96	9	9.4
	864–3	Fudoiwa	biotite	55	4	7.3
				27	2	7.1
				26	2	7.7
b	750	Ootorii	muscovite	25	5	20.0
				23	6	26.2
			altered biotite	55	11	20.0
	440	Kiriu	muscovite	142	27	19.2
c	278	Daradani	leuco granite	31	6	19.3
				17	6	35.3
				117	23	19.6
				30	8	26.6
				16	4	25.0
				(65	3	4.6)
	281	Daradani	leuco granite	69	16	23.2
				52	8	15.4
				83	11	13.2
d	772–1	Shishitobi	leuco granite	64	10	15.6
				26	4	15.4
				34	6	17.6
				33	11	33.3
				59	8	13.6
				23	4	17.4
				44	8	18.2
				12	5	23.1
	770–2	Shishitobi	leuco granite	71	13	18.3
				95	15	16.0
				134	20	14.9

two minute minerals contained in leuco-granite B at Daradani contact zone. Of these zircon-like grains (a) exists at the boundary between quartz and albitic plagioclase, while (b) in orthoclase. Both are rich in thorium, for their longer tracks over 20 microns occupy 14.6 and 19.7 percent of their total track number. (Note: line 9, page 265, Report IV "—to be uraniferous" reads "—to be thorium rich") These zircon-like and thorium rich minute minerals are not rare. So is it in the leuco-granite; to the *Th-U* tendency in zircon grains in normal granite, see Table 4.

All the minute minerals shown in Table 4 (a) are rich in uranium and either inclusions in fresh biotite or minute grains devoid of muscovite. The minute grains of (b) are rich rather in thorium and either accompany muscovite or exist in altered biotite as its inclusions. It was already reported of the monazite inclusions contained in Tanakamiyama muscovite as its minute radioactive minerals. In Table 4 (b) they seem to be also minute monazite. Important is the fact that muscovite and altered biotite contain minute minerals rich in thorium, while fresh brown biotite and quartz contain tiny inclusions rich in uranium, and that both cases are observable often in a single thin section of the same rock. But it must not be misunderstood that fresh biotite always lacks accessory minerals rich in thorium. On the contrary, the Daradani contact samples c (278, 281), both being leuco-granite, tell us that nearly all the minute minerals are thorium rich, though in one of them the percentage is as low as 4.6. These minerals tend to be thorium rich also in leuco-granite at Shishitobi contact of hornfels with just contact. But obviously uranium rich are the zircon-like grains in fresh brown biotite flakes of Fudoiwa granite on Tanakamiyama.

In rocks in their deuteric stage the uranium thorium behavior demands our further investigations. But thorium seems to constitute their leading radioactive content of the minute grains contained in rocks accompanying muscovite, for examples, in leuco-granite, leuco-aplite, low grade mylonite and common two mica granite. Generally, such granitic rocks abundant in muscovite or sericite are feebler in their radioactivity than their cognate adjacent rocks. This appears that, due to an hydrothermal alterations, *uranium* and iron were *bleached out*, when *thorium* was *remaining*.

Table 5 *Th-U* tendency of zircon in the granitic biotite

Sample	Color of biotite (Host)	*Th-U* tendency
T. G. 894	brown	16
T. G. 895	brown	17, 20, 23, 24
T. G. 846-2	brown	10, 24
T. G. 440	brown	10
Shindenba	brown	17, 20
M. G. 521	brown	0
M. G. 505	brown	7
Shigaragi 831	brown	0, 13, 23, 28, 31, 50
T. G. 896-3	brown	53, 43
Ookawara 609	brown	49, 62
K. G. 31	brown	0
T. G. 770 (Hornfels)	brown	100
Chofukuji 242 (Quartz porphyry)	brown	67
T. G. 800	green	15, 25, 30, 31, 49, 56

Zircon grains found in

biotite flakes are first taken up to exemplify the *Th-U* tendency therein (see Table 5). The tendencies are more fluctuating where the host of these grains is brown biotite.

In comparison to Tanakamiyama granite (T. G.), it is characteristic of Mikumo rock (M. G.) that *U* excels *Th* in the ratio, so far as zircon grains in biotite flakes are concerned. A sample of Shigaragi 831 in Table 4 is the contact granite at Hata which belongs to M.G. type.

But the opposite *Th-U* tendency is seen in this sort of zircon grains where their biotite host lies either in hornfels or in quartz porphyry. And the latter tendency seems to be common among granitic rocks lying near the contact.

It must be noticed, however, that the *Th-U* ratio is never uniform even in one sample (T. G. 831) and in the same thin section of it (see Table 6). As already said, in the same sort of minute minerals, for instance, *Th* exceeds *U* in ratio more when their host is muscovite than when it is biotite.

Table 6 *Th-U* tendency of zircon in the sample T.G. 831

Radioactive mineral	Host mineral	*Th-U* tendency
zircon	biotite	23
		0
		13
		28
		50
		31
zircon or monazite	muscovite	93
		100
		68
		100

Table 7 *Th-U* tendency of zircon in the granites

Sample	Host mineral	*Th-U* tendency
T. G. 550-2	sericitized albite	36, 46, 100(3), 94(4)
T. G. 298	quartz and albite	64
T. G. 889	orthoclase	9
T. G. 211-1	quartz	18(4)
T. G. 895	quartz	14, 22
T. G. 674	quartz	16(3)
T. G. 750	quartz	23
K. G. 21	quartz	2
K. G. 16	quartz	8
K. G. 24	quartz	19
Suzuka Pass 628	quartz	54
Kasagi 1152	albite	4(2)
Mushima	green hornblende	8(3)

( ): number of measurements

Such complexity of fluctuations becomes still more remarkable (see Table 7) in zircons contained in quartz where *U* exceeds *Th* in ratio as is the case both with Tanakamiyama and with Kitashirakawa granite. But when sericitized albite is their host, rather the opposite tendency is common among zircon crystals.

The notably radioactive minerals shown in Plate I (found in the medium grained biotite granite collected from Unkoji quarry in Seto City, Aichi Prefecture) is cubic opaque minerals and their radioactivity are $T_\alpha = 179$ and 143 (A and A′ in Plate I Fig. 1a). "M" in Fig. 1a is the malacon type zircon grain which lies between quartz. Mt, At and A't correspond with M, A and A' in the Plate I Fig. 1b. They seem to be the same uraninite grains[26),27)] already mentioned in the first report of this paper[8)] on the Tanakamiyama minute minerals ($T_\alpha = 152$). These opaque minerals contain only uranium as the radioactive element as is evident from the alpha track length distribution.

A pale yellow mineral shown in Plate I Fig. 2 is isotopic and its radioactivity is $T_\alpha = 75$ which was included in the biotite of Minamisakura granite, Shiga Prefecture.

Ooro granite (Naka County, Kyoto Prefecture) contains a pale yellow minute mineral. As is often the case, here in this rock, too, radial fractures are seen around the radioactive mineral. This granite is coarse grained leucocratic one containing comparatively abundant magnetite. The radioactivity of this mineral is $T_\alpha = 29.8$.

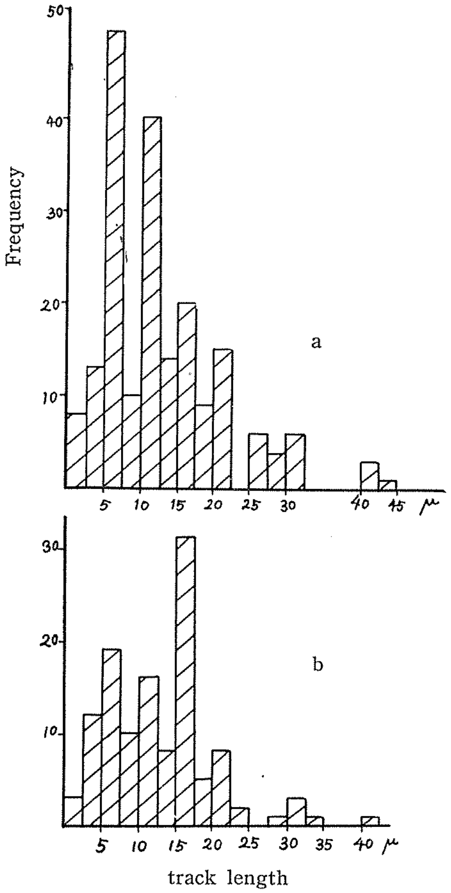


Figure 22 Histogram of the alpha track length from biotite flake

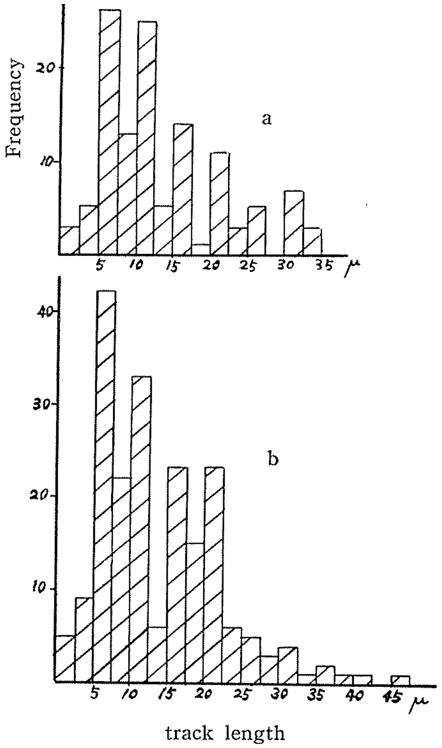


Figure 23 Histogram of the alpha track length from biotite flake

(Plate I Fig. 3) These strongly radioactive minerals exist, though very rare, in every granitic thin section, just as large crystals of radioactive minerals happen to be seen in pegmatite.

Radioactivity of principal minerals, and the radioactive meaning of magmatic differentiation

The microscopic observation of the tracks emitted from biotite give us the impression of greater abundance of long tracks among them than among those ejected from zircon grains; and this tendency was verified by measuring the track length one by one. The four biotite flakes (Shishitobi 896) have left during twelve weeks' exposure 198 (Fig. 22a), 121 (Fig. 22b) 137, (Fig. 23a) and 202 (Fig. 23b) tracks, respectively, numbers quite favorable for the microscopic track counting and length measurement. Alpha tracks of different length were found to be distributed as graphically illustrated in Fig. 24 by a, b, c and d; their comparison with the theoretically presummed uranium and thorium curves is given in the same figure (upper curve of the shadow part: *Th*, under the curve of it: *U*). Despite the occasional discordance between the theoretical presumption and the experimental result as already stated in the previous reports II, III and IV, it is well determinable as to whether uranium or thorium is the principal radioactive element. How many tracks longer than 10, 15, 20 and 25 microns were found among all the tracks longer than 5 microns, is illustrated in Fig. 24 in which the curve, in close resem-

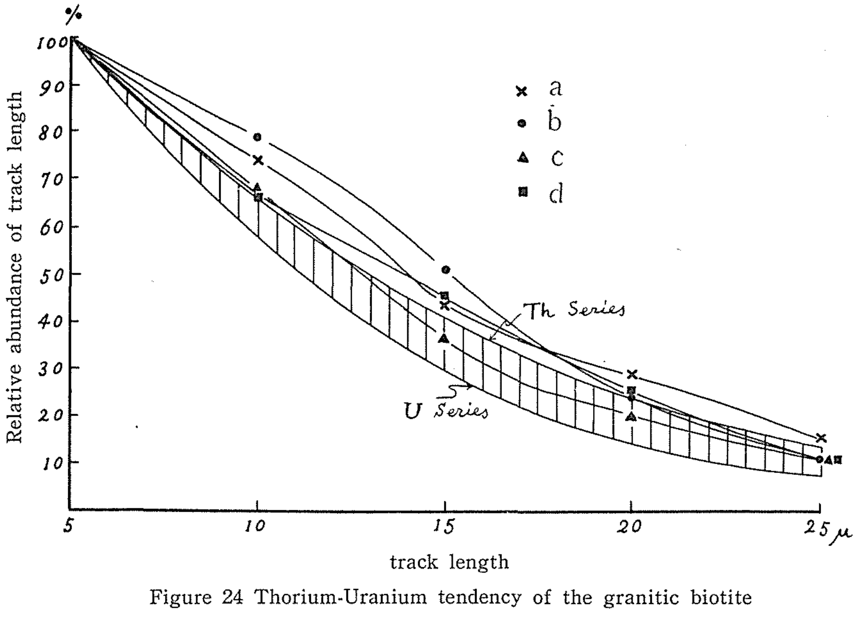


Figure 24 Thorium-Uranium tendency of the granitic biotite

Table 8 *Th-U* tendency of granitic minerals

Sample	Mineral	*Th-U* tendency
846–2	sericitized albite	100
896–3	sericitized albite	100(3)
896–1	green biotite	100(3)
	brown biotite	66
	brown biotite	42
668	green biotite	100
846–1	green biotite	100
831	green biotite	100
Mikumo	brown biotite	15
	brown biotite	13
	brown biotite	7
831	secondary mineral	65, 92, 100

Table 9 Radioactivity of granitic biotites

Sample	Color of biotite	Radioactivity (T_α)
800	brown	0.0025, 0.0034
	green	0.0032
550–1	brown	0.00088
750	brown	0.00075
844(Asamiya)	brown	0.0015
440	brown	0.0012
697–2	brown	0.00025, 0.0005 0.00043
762	green	0.0014
1152(Kasagi)	brown	0.00024, 0.00018

Table 10 Radioactivity of orthoclase and quartz in granite

Sample	Mineral	Radioactivity (T_α)
224	orthoclase perthite	under 0.2×10^{-3}
	quartz	under 0.2×10^{-3}
800	orthoclase	under 0.1×10^{-3}
	orthoclase*	0.72×10^{-3}
	quartz	under 0.1×10^{-3}
	quartz	0.15×10^{-3}
550	orthoclase perthite	0.88×10^{-3}
	orthoclase	under 0.2×10^{-3}
	quartz	under 0.2×10^{-3}
750	orthoclase*	0.5×10^{-3}
	quartz	under 0.2×10^{-3}
271	orthoclase	0.50×10^{-3}
	quartz*	1.2×10^{-3}
844	orthoclase	under 0.2×10^{-3}
	quartz	under 0.2×10^{-3}
440	orthoclase	1.0×10^{-3}
697	quartz*	0.25×10^{-3}
	quartz*	0.06×10^{-3}

(* with red stain)

Table 11 Radioactivity of granitic plagioclase

Sample	Radium content of rock (10^{-12}g/g)	Mineral (altered)	Radioactivity (T_α)
224	2.09	sericitized	2.5×10^{-3}
		sericitized	1.4×10^{-3}
			0.5×10^{-3}
800	0.92	sericitized	1.5×10^{-3}
			0.25×10^{-3}
550	1.87		0.5×10^{-3}
750	1.48		1.1×10^{-3}
271	1.75		1.1×10^{-3}
			0.25×10^{-3}
			0.88×10^{-3}
844	1.22		under 0.2×10^{-3}
440	2.06	sericitized	0.75×10^{-3}
			0.5×10^{-3}
697	0.77**	*	0.46×10^{-3}
		*	0.56×10^{-3}
		*	0.1×10^{-3}

(* with red stain)
(** similar rock (696) near this sample)

blance to that of allanite grains included in Kitashirakawa granite, indicates us of the indisputable thorium superabundance in these biotite flakes. The longer tracks than 20 microns confirm the same tendency which is obtained from the result by the similar treatment in the previous paragraph. Table 8 tells us that in sericitized albite, like its inclusions, *Th* is dominant, while *U* is almost entirely lacking; that biotite shows no general tendency, namely, *Th* excels *U* in Tanakamiyama granite, whereas *U* is alone dominant in Mikumo rock just as we saw in zircon inclusions there; and in secondary minerals, where the radioactive equilibrium remains doubtful, *Th* generally tends to be rich.

Biotite in Tanakamiyama granite, especially in its green biotite (Report III)[13], far exceeds, in radioactive order, those obtained by any other authors' results (for instance, O. H. MERLIN and others[36] concerning those in granitic biotite (see Table 9). Of brown biotite (sample T. G. 800) one flake ($T_{\alpha}=0.0025$) contains minute radioactive inclusions, while another flake ($T_{\alpha}=0.0034$) does not.

As to the radioactive order of the essential minerals of granite, namely, of quartz, orthoclase (Table 10) and plagioclase (Table 11) which exemplifies that of plagioclase: as a rule, sericitized one is more radioactive than the fresh, and sodic one is higher in activity than the calcic. Because of their much larger size, however, they play a bigger role in the total activity of the rock than minute radioactive grains they contain. (ASAYAMA's measurement of granitic radium content is listed also in Table 11).

As for orthoclase and quartz, see Table 10 which tells us, despite the diverse periods of exposure (sometimes even for 111 days), at least that the red stained ones are more radioactive than the not stained.

Table 8 shows us that, while green biotite of a later stage is rich in thorium, brown biotite of an earlier stage than that is rich rather in uranium.

Thorium and uranium behavior during the granitic differentiation is no doubt much complicated. Generally, as the granite grows more acid, its *Th-U* content increases.

J. A. S. ADAMES[26] offered us a figure: "Relative alpha activity (in counts per hour) plotted as a function of the uranium content of volcanic rocks from Lassen National Park, California" (97 P). "The plot", he says further, "suggests that the *Th-U* ratio is roughly constant in these rocks". Now let us examine this figure more closely, and ask: Can the *Th-U* ratio be nearly *constant* during the magmatic differentiation?

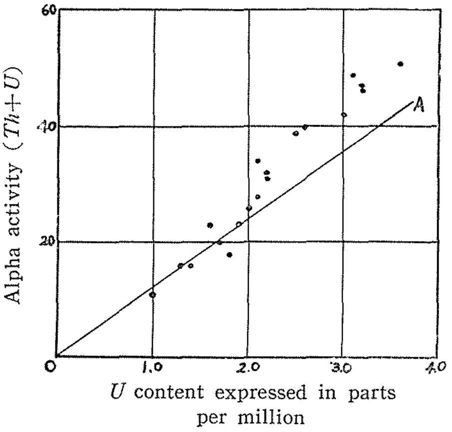


Figure 25 The *U–Th* ratio during the magmatic differention
(from Nuclear Geology p. 97 Fig. 2.5 p. 97)

Dots in upper position above the line OA in Fig. 25 clearly indicate a tendency— a tendency usual both in a series of volcanic rocks— that the *Th-U* ratio increases from basic rocks toward acid ones, in other words, that in volcanic rocks this ratio is magnified as the magmatic differentiation gets more advanced.

Radioactive alteration of granits

Granitic rocks have suffered the hydrothermal alteration at the Mikumo Hyakumaidani pottery stone mine.28) Quartz porphyry was once affected by granite into a leucocratic rock, to produce this pottery stone. The following two points distinguish, in radioactivity, such a leucocratic rock from ordinary granite, quartz porphyry and aplite :

A) In leuco aplite and leuco granite we see feebly radioactive dark as manganoan earthy stains whose chief radioactive source must be thorium, if judged from its alpha track length distribution.

Here, where the emulsion used for autoradiography was 15 microns thick, alpha tracks over 20 microns were 13.6 and 17.8 per cent of the total tracks. This distribution ratio is diminished from 12.9 per cent in the North Corean thorium rich monazite to 1.9 per cent in Ogamo mine uranium rich coffinite.

The dark spots in Mikumo soft leuco aplite vary in their radioactivity from $T_\alpha = 0.018$ to $T_\alpha = 0.136$. Now these spots are due to the impregnation of Fe and Mn into quartz or feldsper and, because the rock itself is porous, the measured radioactive order must be an apparent one and in reality somewhat higher than that. Anyhow, if thorium be its sole radioactive source, 0.014—0.104 per cent of the rock must be assumed to be thorium. But, owing to its eminent emanating power, the loss of *Tn* and *Rn* must also be taken into consideration.

B) Here the kaolinized biotite aplite, altered into leuco aplite, has produced the above said pottery stone along the joints left in the fresh biotite aplite. Deprived of *Fe* by hydrothermal solution, it has been turned into dusty kaoline. The radioactive superiority of fresh biotite ($T_\alpha = 0.018$—0.011) to kaolinized one ($T_\alpha = 0.013$—0.011) confirms us of the big loss of radioactive materials leached away by hydrothermal solution. This radioactive difference is quite remarkable even at a few centimeters' distance.

Thus the radioactive elements leached away (with *Fe* and *Mn*) by hydrothermal solution from fresh granite are redeposited in leucogranite or in leucoaplite (pottery stone) into fissures and dark spots. Such rocks, pulverized to test the emanating power of these gases, reveal us the superabundance of *Tn*, that is to say, while *U* is washed away as uranil, *Th* thus leached from granite is co-precipitated as ferromanganoan material. (Leucogranite and leucoaplite containing these dark spots are gangue for pottery stone.)

Pulverized granite served for the radioactive determination of gases like *Rn* and *Tn*.29), 30), 31), 32) The following three points were made clear from the determination of their emanating power by counting the alpha tracks which these gases issued into the air left on our photo plates:

1) Their emanating power greatly depends not only on the amount, but also on the condition, of their original radioactive elements.

2) The comparative distribution of two branched tracks33) (though seldom, two tracks starting from one point in V form) semiquantitatively determines which of these gases is issuing more.

3) Despite their nearly similar diffusion velocity in the air, the distance between the pulverized sample and the emulsion affects much in radioactivity, because of the great difference in the half life of *Rn*, *Tn* and *An*.33) And this difference enables us to tell which gas or gases was affecting the emulsion. The radioactivity of Actinon whose half life is extremely short is rendered determinable on this method. This in turn suggests something of the original amount of their parent elements (*Ra*, *U* and *Th*) and also of the original situation of these elements in the rock (for instance, whether concentrated in minute inclusions or as a hydrothermal product or again as a weathered deposit). Years long studies of this sort has confirmed the author that, while *Th* is apt to remain in, *U* is likely to be leached away from granitic rocks—as is evident in remarkably acid rocks like muscovite granite.

The fact that some granitic rock, if pulverized into grains finer than 60 mesh, reveals the same emanating power34), indicates no less than that the original materials producing the emanating gases are not minute crystals, but some secondary earthy materials likely to issue these gases.

Summary and conclusion

1 Rock radioactivity gradually decreases from just contact to hornfels.

2 In the hornfelsic rock, dusty parts and biotite after cordierite are distinguished in activity.

3 In minute radioactive minerals, the relation of their quantity to their radioactivity is in lognormal distribution.

4 Generally speaking, in batholith type granite these minute radioactive minerals are mostly feeble, but in stock type granite they are mainly strong in activity. In contact rock and near xenolith, however, grains of high as well as low order are found—in both types of granite.

5 The *Th-U* tendency registered in a pleochroic halo and the distributions of different length alpha tracks emitted from its nucleus mineral are in full agreement.

6 Minute zircon grains are uranium rich in earlier stages of magmatic differentiation, but grow thorium rich in its later stages.

7 The same trend is seen also in biotite, and sericitized plagioclase in merely thorium rich.

8 In acid igneous rocks in general uranium leaching and thorium storage, which took place in later stages of magmatic differentiation, have made the *Th-U* ratio higher than in earlier stages.

Acknowledgement

In completing this report, the author wishes to express his sincere thanks to Prof. A. HARUMOTO, Prof. S. MATSUSHITA, Assist. Prof. H. YOSHIZAWA, Assist. Prof. Z. HATUDA and Mr. T. TSUTSUMI of Geological and Mineralogical Institute, Kyoto University, Prof. K. KIMURA of Physical Institute, Kyoto University, and Dr. T. ASAYAMA and Mr. N. YOSHIDA of Kyoto Technical University for their constant aid and encouragement in the course of this work. Thanks are also due to Dr. S. FUJIZAWA and Dr. S. KOSEKI, Research Laboratory, Fuji Photo Film Co. Ltd. for supplying him with ET-2E plates. Fund necessary for this investigation has been defrayed by the Scientific Grant of the Ministry of Education and by the Grant of the Hattori Hokokai, to which the author wishes to express his thanks.

References

1) BECQUEREL, H.: Sur les radiations émises par phosphorescence. Compt. rend. **122** 422 (1896).
Sur les radiations invisibles émises par les corps phosphorescents. Compt. rend. **122** 501 (1896).
Sur les radiations invisibles émises par les sels d'uranium. Compt. rend. **122** 689 (1896).
2) DAVIS, G. L.: Radium content of ultramafic igneous rocks I Laboratory investigation. Am. J. Sci. **245** 677–693 (1947).
3) DAVIS, G. L. & HESS, H. H.: Radium content of ultramafic igneous rocks II Geological and chemical implications Am. J. Sci. **247** 856–882 (1949).
4) HÉE, A.: Recherches sur la radioactivité dún granite des Vosges par la méthode photographique. Ann. Geophy. **4** 242–252 (1948).
5) HÉE, A., DERVILLE, R. P. & JAROVOY, M.: Determination of the radioactivity of Quincy granite by the photographic method. Am. J. Sci. **252** 736–744 (1954).
6) PICCIOTTO, E. E.: Distribution de la radioactivité dans les roches éruptives I, II, III, IV Bull. Soc. belge Géol. **LIX** 170–198 (1950).
VI Bull. Soc. belge Géol. **LXI** 215–220 (1952).
7) COPPENS, R.: Étude de la radioactivité de quelques roches par l'émulsion photographique Thèses (1949).
8) HAYASE, I.: The radioactivity of rocks and minerals studied with nuclear emulsion. I The minute radioactive minerals of Tanakamiyama and Mikumo granites, Shiga Pref. Japan. Mem. Coll. Sci. Univ. Kyoto ser. B **XX** 247–260 (1953).
9) STRUTT, R. J.: On the distribution of radium in the Earth's crust and on the Earth's internal heat. Proc. Roy. Soc. London A **77** 472–488 (1905).
10) HURLEY, P. M.: Distribution of radioactivity in granites and possible relation to helium age measurements. Bull. Geol. Soc. Am. **61** 1–7 (1950).
11) HAYASE, I.: Radioactivity of minute radioactive mineral in granite. (in Japanese) Kagaku **21** 422–423 (1951).
Radioactivity of allanite from the Kinki granites. (in Japanese) Kagaku **21** 543–544 (1951).
Some minute radioactive minerals from the Tanakamiyama granite. (in Japanese) Kagaku **21** 91 (1952).

12) HAYASE, I.: The radioactivity of rocks and minerals studied with nuclear emulsion II Thorium content of granitic allanites. Mem. Coll. Sci. Univ. Kyoto ser. B **XXI** 171–182 (1954).

13) HAYASE, I.: The radioactivity of rocks and minerals studied with nuclear emulsion III Radioactivity of biotite of the Tanakamiyama granite, Shiga Pref. Japan. Mem. Coll. Sci. Univ. Kyoto ser. B **XXII** 177–184 (1955).

14) HAYASE, I.: The radioactivity of rocks and minerals studied with nuclear emulsion IV Thorium uranium ratio measurement of the minute radioactive minerals by the photographic method. Mem. Coll. Sci. Univ. Kyoto ser. B **XXIII** 255–274 (1956).

15) VON BUTTLAR, H. & HOUTERMANS, F. G.: Photographische Messung des *U*- und *Th*-gehaltes nach der Auflagemethode. Geochi Cosmochi. Acta **2** 43–61 (1951).

16) HAYASE, I. & TSUTSUMI, T.: The minute radioactive minerals of the Gyojayama granite, Kyoto Pref. (in Japanese) Jour. Jap. Assoc. Min. Pet. Econ. Geol. **40** 216–223 (1956).

17) HAYASE, I.: Thorium halo in the Shishitobi granite, Shiga Pref. (in Japanese) Kagaku **21** 143–144 (1951).

18) ASAYAMA, T.: The radium content and the chemical composition of granitic rocks in Japan, especially in the Tanakami-Mikumo and the Hiei regions, Shiga and Kyoto Prefectures. Mem. Fac. Ind. Arts, Kyoto Tech. Univ. Sci. Tech. **3** (B) 25–54 (1954).

19) HAYASE, I. & TSUTSUMI, T.: Autoradiographic studies on some lamprophyre and mudstone (in Japanese) Jour. Jap. Assoc. Min. Pet. Econ. Geol. **39** 15–21 (1955).

20) HAYASE, I.: Uranium contents of zircons in granite. Mineral. Jour. **1** 147–159 (1954).

21) HAYASE, I.: Extinct halo "type A" found in the biotite in the contact zone of Tanakamiyama Shiga Pref. Japan (in Japanese) Chigaku (Kyoto) No. 3 19–23 (1951).

22) HAYASE, I.: The distribution of thorium halo on the Kinki granite (in Japanese) Chigaku (Kyoto) No. 4 7–11 (1951).

23) HASAYE, I.: The development of pleochroic haloes and the alpha radioactivity of the nucleus minerals. Mineral. Jour. **1** 213–223 (1955).

24) HAYASE, I.: Relative geologic age measurements on granites by pleochroic haloes and the radioactivity of the minerals in their nuclei. Am. Mineral. **39** 761–772 (1954).

25) FAUL, H.: Nuclear Geology: a symposium on nuclear phenomena in the earth sciences. John Wiley (1954).

26) YAGODA, H.: The localization of uranium and thorium minerals in polished section. Part 1: The alpha ray emission pattern Am. Mineral. **31** 87–124 (1946).

27) YAGODA, H.: Radioactive measurements with nuclear emulsions John Wiley (1949).

28) HAYASE, I.: Feldspasic pottery stone from the south Shiga Pref. (in Japanese) Chigaku (Kyoto) No. 1 83–84 (1949).

29) HATUDA, Z. & HAYASE, I.: A new radioactive prospecting method by means of alpha particle photo plate (in Japanese) Rep. Res. Lab. Fuji Photo No. 1 113–115 (1953).

30) HATUDA, Z.: A new method of radioactive exploration using nuclear emulsions and comparison with other methods. Mem. Coll. Sci. Univ. Kyoto ser. B **XX** 89–92 (1952).

31) HATUDA, Z.: Radon content and its charge in soil air near the ground surface. Mem. Coll. Sci. Univ. Kyoto ser. B **XX** 285–306 (1953).

32) HATUDA, Z.: Radioactive method for geological exploration. Mem. Coll. Sci. Univ. Kyoto ser. B **XXI** 231–271 (1954).

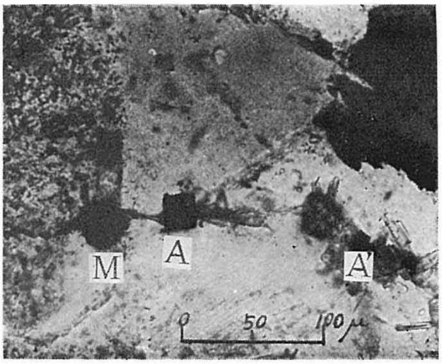
33) HAYASE, I.: Radioactive characteristics of Misasa granite and the radioactivity of Misasa sinter deposit (in Japanese) Rep. Balneo. Lab. Okayama Univ. No. 18 3–10 (1957).

34) HAYASE, I.: Direct measurement of radioactive gases emanating from the powdered granite (in Japanese) Kagaku **24** 365 (1954).

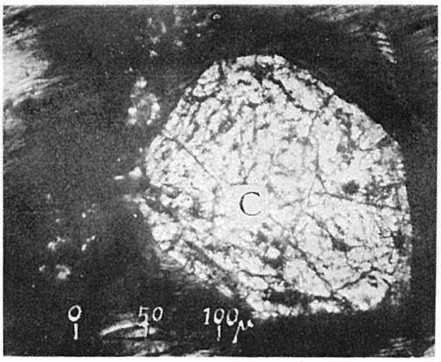
35) POOLE, J. H. J. & MATTHEWS, C. M. E.: The theory of the use of alpha ray ranges in nuclear emulsions for the determination of the radioactive contents of materials. Sci. Proc. Royal Dublin Soc., **25** 305–316 (1951).

36) MERLIN, G. H., PICCIOTTO, E., et WILGAIN, S.: Étude photographique de la distribution de la radioactivité dans la granodiorite de l'Adamello. Geochi. Cosmochi. Acta **11** Acta **11** 141–161 (1957).

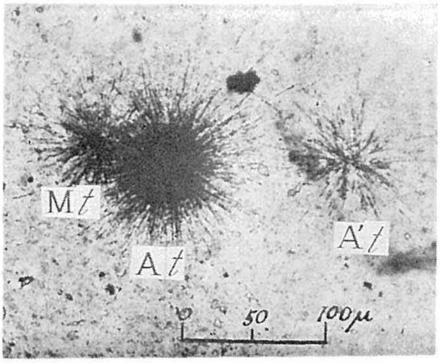
1a



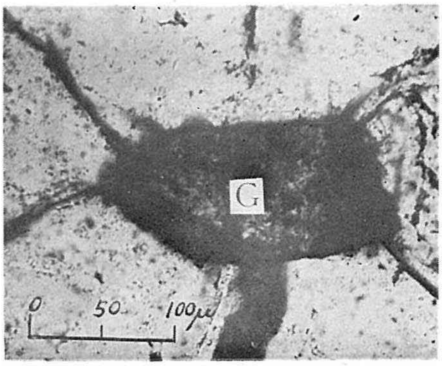
2



1b



3



1a. minute uraninites on the Seto granite, Aichi Pref.
1b. their autoradiographic pattern
2. minute radioactive mineral on the granite from Yasu-minamisakura, Shiga Pref.
3. minute radioactive mineral on the Ooro granite