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The Radioactivity of Rocks and Minerals Studied with Nuclear Emulsion

II Thorium Content of Granitic Allanites

By

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Abstract

The alpha radioactivity of minute granitic allanite was measured by the autoradiographic method. Allanite contains mostly thorium as the radioactive element, and the thorium can be calculated by the alpha track length and population. Granitic allanites treated here in this paper varied in thorium content from 0.5 to 1.6 percent even in the same thin section of the rock. The thorium content of allanite is peculiar to each granite mass, but has no direct relation with the radioactivity the host granite itself contains.

Introduction

The uranium and the thorium content can be measured, at first qualitatively and then quantitatively, by determining the radioactivity of rocks and minrals by the photographic method.[1)-10)] Uranium, actinium, thorium and samarium are the substances of natural alpha radioactivity, among which, however, samarium is rather negligible because of its rarity in nature and of its feeble radioactivity, while actinium needs no special measurement, for it is always contained in uranium in a certain constant ratio. Therefore, what is to be measured is thorium and uranium alone. For simplicity's sake, however, these two elements are provisionally supposed to be in radioactive equilibrium, to which thorium attains, in reality, in some 70, and uranium in about 10^6 years.

Now the alpha track number recorded on the nuclear emulsion is proportional to the uranium and the thorium content as well as to the alpha particles' permeability (ψ) through the minerals.[2)] Hence, between arbitrary two minerals either of the same species or of similar permeability, the alpha track populations can be regarded, without any further consideration, as the functions of the content of their radioactive elements. The alpha track length, in turn, denotes the alpha range peculiar to each radioactive element. In ET-2E nuclear emulsion, Fuji Photo Film Co. Ltd., for instance, ThC' alpha track length is 54.6 microns and RaC' alpha track length is 44 microns. Only the alpha tracks of full length from ThC' and RaC', which passed almost parallel to the surface of the emulsion, show the above-said two ranges, whereas those that injected it either at high angles or in residual ranges are much shorter. In either case, therefore, any track apparently longer than 45 microns can be ascribed to ThC'.* Hence, the track number between 54.6 and 45 microns in length designates the thorium content in the mineral. Thus, in minerals either of the same species or of the same alpha permeability, the number of long tracks (per given unit area and unit time exposure) must be proportional to the thorium content; and hence the uranium content in question can be measured from its total track number, if the above-mentioned thorium content is taken into consideration. In the same manner, if the permeability of rocks and minerals are known, this method is applicable also to the measurement of the uranium and the thorium content involved in them. In this paper, however, the above-said method is applied chiefly to granitic allanite.

Counting of alpha track number

In applying this method, care must be taken so that the surface of the section may be polished as evenly as possible; for the different orders of radioactivity cannot theoretically be compared except in case of geometrical plane.[11a, b] Naturally this method is inapplicable to the minerals whose radon and thoron are easily diffusible from the section. In case of thin section of rock, it is desirable that its thickness may be larger than the range of ThC' emitting from it.

For the exposure the nuclear emulsion must be put, without cover glass, directly upon the polished surface or upon the thin section. The exposure must be varied, according as the content of the radioactive element differs, till the alpha track number becomes the most convenient for the microscopic counting. Broadly speaking, the exposure is variable, in case of radioactive minerals, from hours to days, whereas, in case of rocks, from days to weeks. During the exposure the nuclear emulsion must be carefully guarded from excessive heat and moisture, lest the latent image should fade out. The fading, nevertheless, is not inevitable, even if the radiographic camera, filled with nitrogen, is kept at about −7°C in a refrigerator. A calibration by means of counting the blank tracks printed on the unexposed emulsion manufactured and kept in the same condition furnishes the result with a higher accuracy. The cross marks (thin section) or the outline of the mineral piece (polished section) printed on the photo-plate give much aid to the localization of the sample on the emulsion. The latter is a technic of exposing the photo-plate to a dim light for a while, so that the outline

* It was treated in this way and in detail by POOLE.[7]

of the sample may be printed negatively on the emulsion. Here the first thing to be certified is whether or not the alpha tracks were almost equally distributed throughout the portion of the emulsion on which the mineral was fixed. A remarkably uneven distribution of alpha tracks indicates the presence of the heterogeneous inclusion in the sample.

Distribution of alpha track

The alpha track distribution ejected from allanite into the photo-plate depends upon the following conditions:

1) the homogeneity of radioactive elements in allanite,
2) the absence of inclusion in it,
3) the freshness of it,
4) the evenness of the surface of the thin section of rock,
5) the close contact of the allanite with the photo-plate without air gap during the exposure.

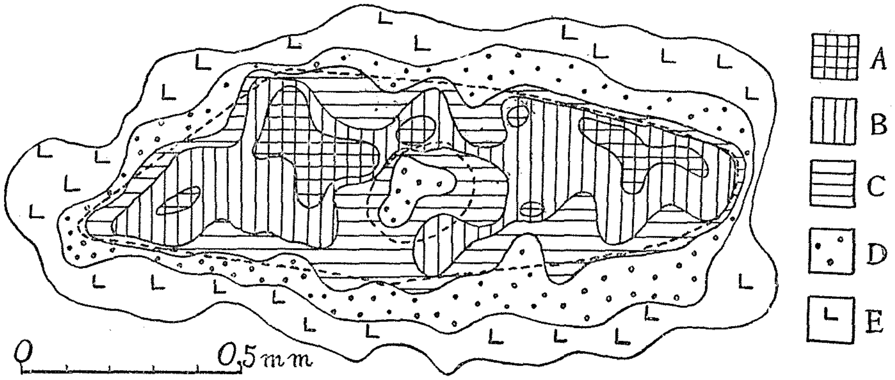

Fig. 1 The alpha track distribution from an allanite of Tanakamiyama granite, Shindenba Quarry. A–over 80 tracks, B–70∼80, C–60∼70, C–30∼60, D–5∼30 per 0.01mm^2 for 28 days' exposure.

Figure 1 illustrates an allanite sample of Tanakamiyama granite, at Shindenba quarry, exposed for 28 days.[10] The alpha track distribution beyond the boundaries of allanite indicates some air gap; probably the minute groove at the grain boundary induced it, which was made during the polishing, owing to the different hardness of the minerals. The outer dotted line in the figure is allanite boundary and the inner one is a quartz inclusion. The alpha tracks are insufficient on the non-radioactive quartz inclusion, but the radioactivity of surrounding allanite is fairly effective. The alpha tracks located on these non-radioactive minerals (either an inclusion or its outer part of allanite) are discriminable at a glance under the microscope, for these alpha tracks are starting generally at low angles from the original allanite. In some rare examples, the brownish stain in the granitic allanite shows fairly radioactive concentration; this heterogeneity will be reported in another paper. Though the average track number per $0.01mm^2$ is 110 in the case of the Figure 1, the real track number left on the photo-plate, in loose contact with the allanite, is always found less than that.

Comparative thorium content in allanite measured by long alpha tracks

As stated above, the thorium content in each mineral is proportional, if it is of the same species, to the long alpha track population printed on the emulsion. Figure 2 shows the relation between the number of their alpha tracks

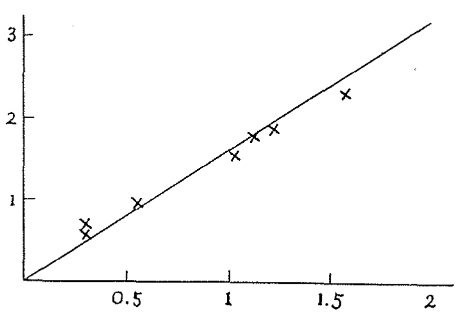

Fig. 2 The relation between the number of long alpha tracks longer than 46 microns and the thorium content of seven pegmatitic allanites. Ordinate—number of long tracks ($L\alpha$—per mm^2 per a day) Abscissa—percent of thorium

apparently longer than 46 microns ($L\alpha$—per mm^2 per day) and the thorium content of other allanites whose thorium contents are already known. The result, however, is reliable only after the calibration of the blank tracks from the emulsion itself; but it must be remembered that nearly full length of ThC' tracks are seen oftener among blank tracks than among ordinary ones. (The thorium stars are relatively abundant in the emulsion itself.) The alpha tracks from the mineral, on the other hand, can be distinguished from the blank tracks, because the former never form a thorium star in the emulsion. The blank tracks from the plate glass itself are also distinguishable, if we observe from which side of the emulsion such tracks were projected on it. Allanite grains obtained at Otowadani (Shugakuin, Kyoto City), though varying from 5 to 0.5mm in size, are similar in track number. These minute allanites, implying biotite, apatite and sometimes zircon, show no remarkable radioactive heterogeneity *, as is often the

* The heterogeneous distribution of radioactive elements was rarely detected in granitic allanite, though commonly seen in pegmatitic ones.

case with pegmatitic ones. In the same manner, granites of various districts were tested for the determination of the thorium content of the minute allanites contained therein. In such a measurement, those allanites whose size was smaller than 0.1mm square, or those which had too small a number of long alpha tracks, were excluded. Experimentally, from Figure 2, the following relation was established between the thorium content in allanite and the number of alpha tracks longer than 46 microns :

$$L\alpha = K\psi Th$$

Here Th=1/100 (one persent), $\psi = 6.3$ (alpha permeability of allanite), and $L\alpha = 1.6$ (from Figure 2)

$$\text{Then : } K = 25.4$$

The value "K" remains constant concerning almost all the rocks and minerals. Now the value "K" may be applied also to any other mineral than allanite, and its result may be compared to the already analysed results. *

Thorium equivalent of granitic allanite

As already stated, thorium is prominent, but uranium is negligibly small in quantity, especially in granitic allanite.[12] According to the pioneering work of Yagoda,[2] alpha track population left upon the autoradiography projected from the polished mineral is :

$$T\alpha = \psi(25.73U + 7.80Th)$$

Here $T\alpha$ is the alpha track number per sec. per cm^2, ψ the permeability of the mineral, and U or Th the uranium or thorium content in the mineral. In allanite, $\psi = 6.3$[2], and U is negligibly small as compared with Th.

$$\text{Hence } T\alpha = 6.3 \times 7.8\ Th \qquad Th = T\alpha/49.14$$

TABLE 1

Shugakuin allanites

Grain size $\times 0.01\ mm^2$	$T\alpha$	Th equivalent
24.0	0.80	1.62
45.9	0.69	1.41
1.13 (A)	0.61	1.24
2.15	0.35	0.72
1.40 (B)	0.29	0.59
7.87	0.22	0.46

* As pointed out by Poole[7], no satisfactory result is obtainable from the calculation by long alpha tracks themselves.

Table 1 illustrates the thorium content in allanite of Shugakuin granite. This allanite with idiomorphic crystals was found in the Kitashirakawa granite at Shugakuin quarry. In the same thin section of granite, two allanite grains A and B in Figure 1 have pleochroic haloes; the halo of the grain A (28 microns in width) is darker than that of the grain B (18 microns in width). This confirms that the radioactivity of A and B is $T\alpha=0.61$ and $T\alpha=0.29$ respectively. The tendency of this radioactive divergence between allanite grains, in the same thin section of granite, is not so extraordinary as between zircons. Generally many allanite grains in the same locality have the same order of radioactivity.

TABLE 2

Daimonji allanites

Grain size $\times 0.01mm^2$	$T\alpha$	Th equivalent
0.56	0.55	1.12
5.77	0.49	1.00
1.54	0.39	0.80
0.65	0.38	0.78
* 380	0.31	0.63
* 725	0.36	0.72

* Placer allanites suffered with weathering

Daimonji allanite (Table 2) shows, by its smaller grain size, the same decreasing tendency of radioactivity. The placer allanite offers a somewhat smaller value of the radioactivity; perphaps because they have suffered surface weathering and other leaching of radioactive elements. The values given in Table 2 well conform with those already measured by the analytic method;[13) 14)] and the table informs us that, in the range of the radioactive contents, the thorium in granitic allanite is much narrower than the uranium in granitic zircon.[16)] This narrower range of the former suggests, in turn, the earlier crystallization of, that is, the shorter duration of thorium migration into, those allanites than the zircons rich in uranium. The thorium migration was, no doubt, aided by cerium and other rare earthes in allanite, which are more similar to thorium than to uranium in their chemical behavior.

The length of alpha tracks from the allanites

The ratio between thorium and uranium was measured and calculated by such investigators as Poole,[7)] von Buttlar,[6)] Hée,[16)] Picciotto[8)] and Coppen,[9)] from the ratio between the track number of longer range and that of shorter range. Some samples of allanite obtained from Kitashirakawa granite are illustrated in Figure 3, in which the total track number is compared with the three track numbers whose horizontal range is (A) longer than 45 microns, (B) between 35 and 45 microns, and (C) longer than 35 microns respectively. Here the total track number, however, designates the alpha tracks ejected from each allanite grain during 28 days' exposure.

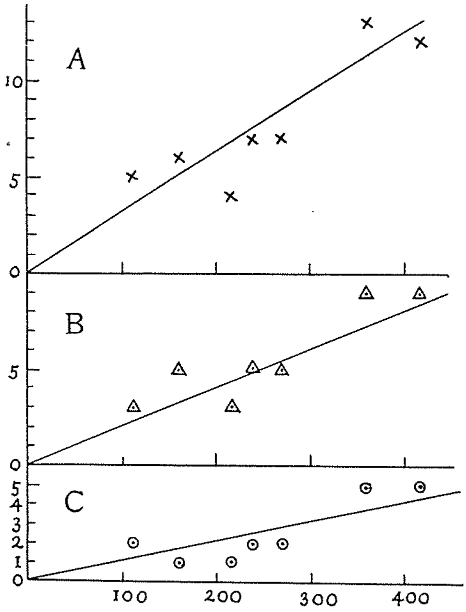

Fig. 3 The relation between the number of long alpha tracks and that of total alpha tracks of some allanites from the Kitashirakawa granite. Ordinate—number of long tracks Abscissa—number of total alpha tracks

The tendencies illustrated as B and C in Figure 3, showing a close approximation to the value given above, prove qualitatively that thorium is the chief radioactive element in those allanite grains. Figures 4 A and 4 B indicate how many tracks of divergent length two grains of allanite ejected. And if Figures 4 A and 4 B (of allanite) are compared with Figures 5 A and 5 B (of zircon), it is evident that in allanite the chief radioactive element is thorium, while in zircon uranium. Two allanite grains included in Kitashirakawa granite samples left upon the photo-plates, after 4 weeks' exposure, 162 and 217 alpha tracks (A and B in Figure 4), whose length was carefully measured one by one. Among tracks of so moderate number, however, we found only one or two long tracks, too few to enable us to determine, with strictness, the thorium content in an allanite grain. *

As shown in Figure 5, on the other hand, two zircon grains included also in the Kitashirakawa granite samples left 157 and 132 tracks. To some degree it is certain, through experiments of another sort,[16] that the principal radioactive element of zircon is not thorium but uranium. If Figure 4 (allanite) and Figure 5 (zircon) are compared with each other, it becomes clear that long tracks are found more in the former than in the latter. But by means of so few long

* The fact that more thorium than uranium is contained in allanite, was perceived from various investigations, for intance, Marble's. [15]

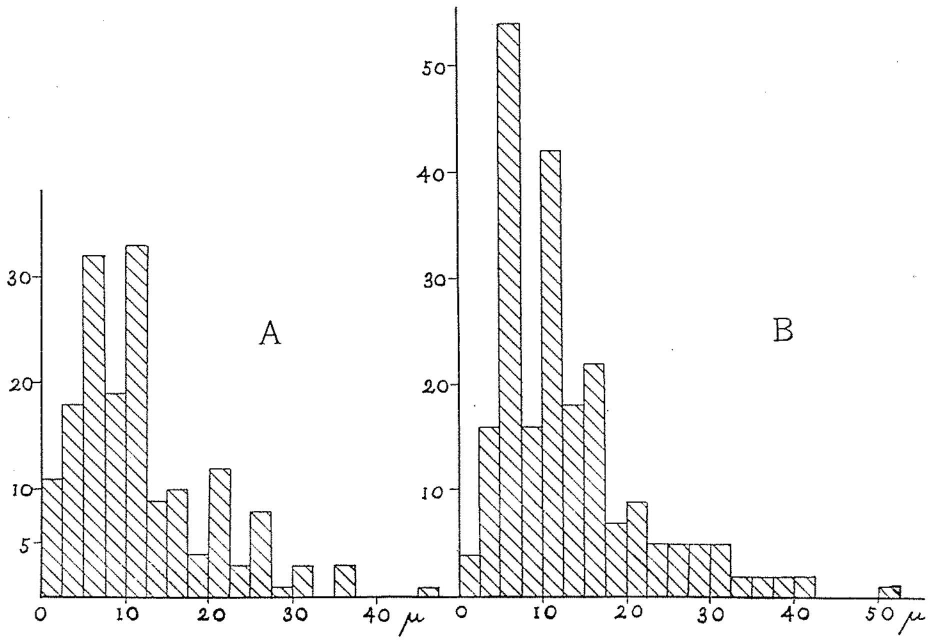

Fig. 4 Alpha track lengths distribution of two allanite grains from Kitashirakawa granite. Ordinate—number of alpha tracks
Abscissa—horisontal lengths of alpha track in microns

tracks the thorium-uranium ratio is hardly determinable. And this, at last, led the present author to the counting of tracks longer than 20 microns alone, so as to compare them with the total track number. As the ThC' track length has been found 54.6 microns in ET-2E plate, other ranges of alpha tracks are easily determinable. Now the ratio of the tracks longer than 20 microns to the total track number is 31/162=0.191 (Figure 4 A) and 36/217=0.166 (Figure 4 B), whereas 10/157=0.064 (Figure 5 A) and 13/132=0.099 (Figure 5 B). Thus it is obvious that, judging from the total track number, the former (allanite) is far more abundant than the latter (zircon) in long range tracks.

More detailed results concerning track length, long track number and thorium-uranium content, will be stated in another paper. Here the author emphasizes, qualitatitvely, only that tracks longer than 20 microns are emitted far more from allanite than from zircon.

What must not be left unsaid here, by the way, is that the idea of the so-called "least discernible length of track", which many investigators hold concerning tracks of extremely small length caused by their high angle or their

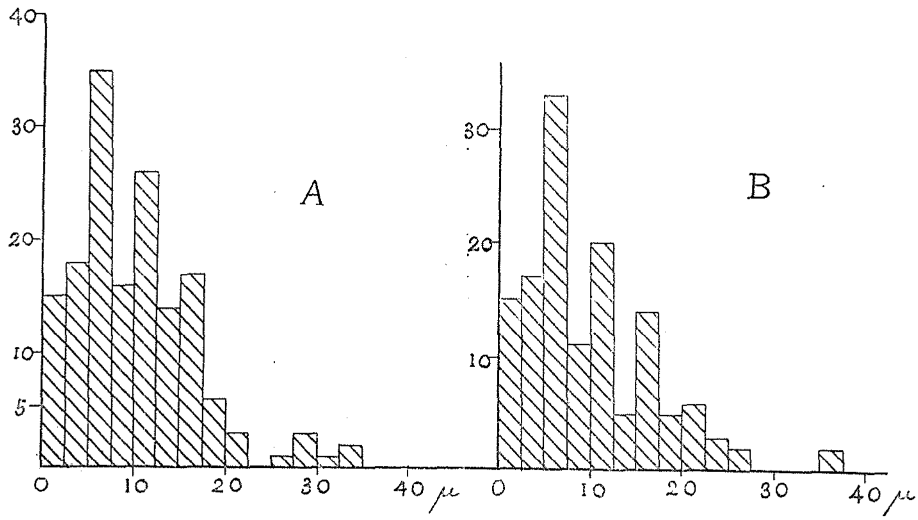

Fig. 5 Alpha track lengths distribution of two zircon grains from Kitashirakawa granite.

horizontally residual range, was practically not so helpful to investigators. For short tracks of, say, 3 microns may show, under the microscope, very different degrees of discernibility, if their angles or positions in the emulsion differ. High-angle alpha tracks can, by moving the microscopic tube up and down, easily be pointed out as such. And where alpha tracks are scattered in a residual form, there can be counted a very short, low angle track among them, if it is in process of coming from the center of the radiation; but hardly determinable, if it is not from the center. Though there is always some room for personal error, such alpha track number must be measured experimentally before all other things.

Thorium equivalent of some Japanese granitic allanites

Table 3 illustrates the radioactive orders of allanite of various granite samples–mostly biotite granite except the quartz porphyry obtained at Oomi-Hachiman and liparite at Yanahara—contained in their thin sections. The size of the allanite is generally smaller than 1 mm.

Allanite is generally more abundant in fine or medium grained granite than in coarse grained one, and richly involved especially in the granite accompanying some pale greenish hornblende, and also in the xenolith lying near the contact margin. Rocks containing allanite are acid rocks, like granite porphyry, quartz porphyry and liparite which are not rare. Idiomorphic fine crystals of allanite are seen rather in granite, but ovoidal porphyroblastic ones more in quartz por-

TABLE 3

Locality	Sample number	Size of allanite ×0.01mm²	Tα	Thorium equivalent (percent)
A	S-31	4.06	0.46	0.94
	S-31	4.30	0.45	0.92
	S-29	2.00	0.45	0.92
	S-32	2.38	0.41	0.83
	S-6	1.30	0.38	0.73 *
	S-27-E	3.34	0.33	0.67
	S-9	4.48	0.30	0.61
	S-27-E	5.57	0.27	0.55
	S-27-E	2.44	0.26	0.53
B	707	0.38	0.48	0.98
		2.01	0.44	0.90
		0.26	0.42	0.85
		2.89	0.37	0.75
		0.26	0.37	0.75
		0.35	0.36	0.73
	700	4.12	0.35	0.71
C		1.47	0.50	1.02
		1.80	0.48	0.98
		1.56	0.42	0.85
		6.91	0.16	0.33
D	Y-7a	0.92	0.57	1.16
	Y-76	1.33	0:56	1.14
	Y-5	1.33	0.52	1.06
	Y-9a	0.52	0.41	0.83
	Y-100	0.92	0.24	0.49
	Y-126	7.20	0.18	0.37
E	O-154	1.80	0.25	0.51
		1.13	0.22	0.45
		3.62	0.21	0.43
		1.47	0.19	0.39
		0.58	0.19	0.39
		0.28	0.18	0.37
		1.77	0.17	0.35
F	O-179	4.92	0.35	0.71
		5.68	0.25	0.51
G		2.62	0.40	0.81
		1.65	0.40	0.81
H	857	1.50	0.75	1.53
	878	0.69	0.60	1.22
	881	0.53	0.17	0.35
I		5.13	0.39	0.79
J	251	0.39	0.48	0.98
		1.47	0.39	0.79
		0.71	0.39	0.79
	245	2.60	0.35	0.71
	249	1.25	0.24	0.49
	245	0.92	0.23	0.47

J	251	7.04	0.20	0.41
		12.0	0.20	0.41
	242	6.07	0.14	0.29
K	T-294-a	0.16	0.44	0.90
		0.33	0.38	0.77
		0.38	0.27	0.55
		1.93	0.19	0.39
		0.70	0.16	0.33
		0.38	0.11	0.22

A Loc. Kitashirakawa, Sakyo Ward, Kyoto City.
Rock: Adamellite rich in fine allanite crystal (stock type intrusion). It is well known that the contact margin or xenolithic schlieren, such as Daimonji and Shugakuin, includes much allanites.
* This allanite sample is surrounded by epidote with zonal intergrowth.

B Loc. Tanakamiyama, Kurita County, Shiga Pref.
Rock: Mainly coarse grained biotite granite containing some muscovite (stock type intrusion). Allanites are seen only in the xenolithic schlieren and in the medium or fine grained biotite granite including some greenish hornblende. Some radioactive minerals had been reported of this granite pegmatite.

C Loc. Ishigure, Inabe County, Mie Pref.
Rock: Coarse grained biotite granite (stock type intrusion). Some radioactive or rare earth minerals had been reported of this granite pegmatite.

D Loc. Katanoyama, Kitakawachi County, Osaka Pref.
Rock: Coarse grained biotite hornblende granite (batholith type as a part of Ryoke granite). The large xenolithic quartz diorite accompanies this granite.

E Loc. Mushima Islet, Oda County, Okayama Pref.
Rock: Ryoke type coarse grained biotite granite (batholith). The outcrop of roof pendant of Palaeozoic formation is seen at the south eastern part of the islet.

F Loc. Manabe Islet, Oda County, Okayama Pref.
Rock: The same petrographic and geologic feature as the granite of Mushima.

G Loc. Rokko-zan, Nakanohata, Kobe City.
Rock: Biotite granite with pink potash felspar forming a stock type intrusion.

H Loc. Kabuto, Suzuka County, Mie Pref.
Rock: Coarse grained biotite hornblende granite partly schistosed as a part of Ryoke granite (batholith).

I Loc. Maruyama. Inago, Ayama County, Mie Pref.
Rock: This granite is geologically continuous to the rock (H) and the same in rock feature as (H).

J Loc. Chofukuji, Gamofu County, Shiga Pref.
Rock: Quartz prophyry partly including biotite granite porphyry. This rock is developed widely in the south-east part of Shiga Pref. and constitutes a part of extrusive facies of Hanazonoyama granite.

K Loc. Yanahara, Kume County, Okayama Pref.
Rock: Liparite of various rock facies.

phyry and liparite. Owing to this irregularity of their crystal forms of extrusive rocks, their size is hardly determinable. Though the values even in the same thin section of granite differ from each other, allanites of a different rock mass offer still more different radioactive order.

Conclusion

1) The range of radioactive content of granitic allanite is from 0.3 to 1.6 percent for the equivalent of thorium. Generally, allanite including less than 1 percent thorium is the commonest, and that of over 2 percent is never seen in this study.
2) Allanites that contain different degrees of radioactive element are seen even in the same thin section of granite; but this tendency is not so remarkable in allanite as in zircon.
3) Different granite masses include each different degrees of allanite in their thorium content.

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