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The Radioactivity of Rocks and Minerals Studied with Nuclear Emulsion IV

Thorium Uranium Ratio Measurement of the Minute Radioactive Minerals by the Photographic Method

By

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Abstract

Thorium uranium ratio measurement of the alpha radioactivity of minute mineral was studied by the photographic method. The preliminary result of these studies was presented by total alpha track length distribution and by the absorbing method of metalic foil. The former method can be aplied on the feeble radioactive minerals, and the latter on the high radioactive ones.

At the short time exposure, care must be taken to the radon or thoron leakage from the minute radioactive minerals.

Introduction

The range in the radioactive order of minute accessory minerals included in granite is large. The radioactive elements contained in each grain of these minerals, more especially the uranium-thorium ratio in it, have remained almost uninvestigated. In this paper, therefore, the above said ratio in each grain and the non-equilibrated radioactive condition of these two elements therein are treated. Their different alpha track lengths printed on the nuclear emulsion render this ratio determinable. Here simply some data are given, while their petrological and mineralogical interpretation is left for another chance.

The ideal (or theoretically presumed) alpha track distribution of different lengths emitting from the polished surface of minerals

In case uranium and thorium are in their radioactive equilibrium, Pool's[1)]

* Read at the meeting of the Geological Society of Japan held in Kyoto on April 2 1956.

and von Buttlar's[2] results can be illustrated by such an alpha track distribution of different lengths as seen in Figure 1, in which the ordinate designates the alpha track number emitted, especially, from uranium and thorium of the same amount, while the abscissa denotes the apparent alpha track lengths left on the ET nuclear emulsion.

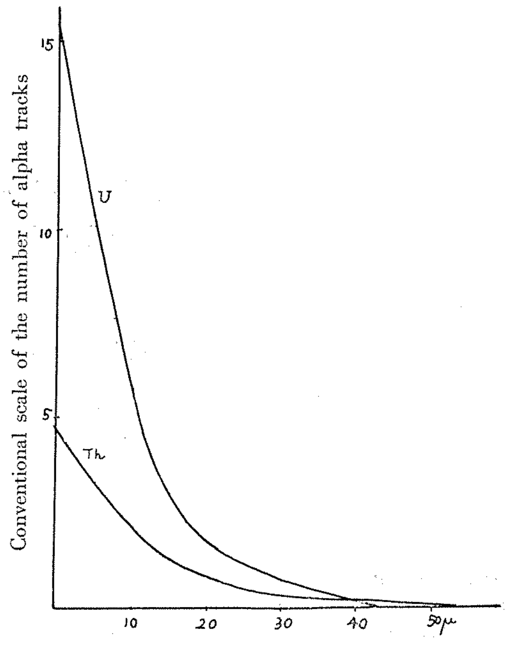


Figure 1

The ratio between the total track number and the longer track population has been the unique clue for the autoradiographic studies on the uranium thorium ratio contained in rocks and minerals. Nevertheless, the scarcity of longer tracks among ordinary ones is so remarkable that their plenitude must be searched for among countless tracks beyond our enumeration. To evade this dilemma, tracks of moderate length of between 5 and 25 microns were taken up, as a rule, for the comparison.

Not that the comparison of total track number with tracks longer than those emitted from ThA is less necessary, but that the liable errors in counting total track number seem to the author more serious. The abundance of the least descernible variety of tracks, which, as already said,[3] often endangers our track counting, no longer disturbs our investigation, if tracks longer than 5 microns alone are chosen.

Now Figure 2 illustrates how much different percentage of tracks longer than 10, 15, 20 and 25 microns, respectively, the uranium and thorium series were emitting, when tracks longer than 5 microns were 100 percent. Since the tracks of ThA and ThC' origin surpass in length those of RaA and RaC' origin, the comparative plenitude of long tracks indicates the greater abundance of thorium content. This figure is nothing but a presumed result from an ideal surface of rock.

When the track distribution of different lengths is examined in a figure, the fact that the line representing it falls, namely, whether near the uranium curve, or close to the thorium curve, or again between the two curves, informs us that the radioactive content in a rock or mineral is uranium or thorium or both.

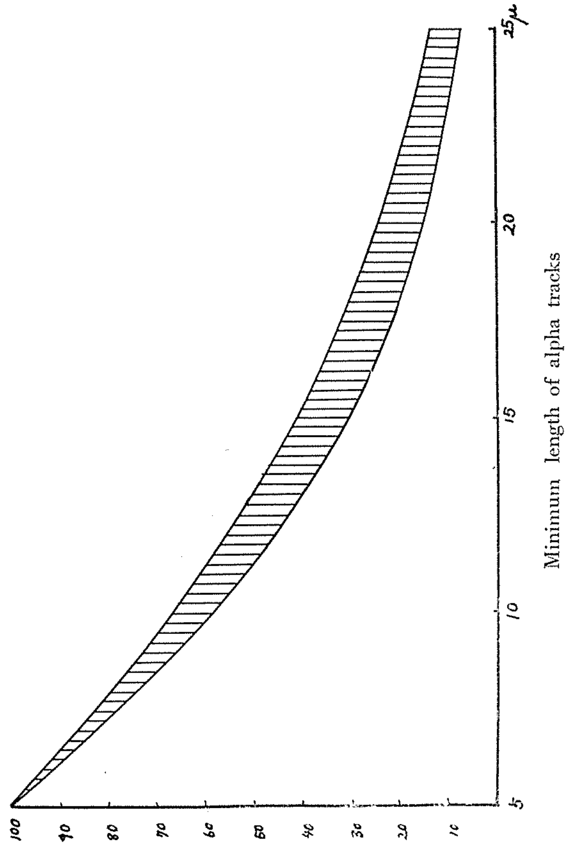


Figure 2

Such a result as given in the foregoing paragraph can be expected only when, firstly, the polished surface of the rock or mineral is so geometrically even that there exists no air gap between it and the nuclear emulsion, secondly, the alpha permeability of all the minerals on the polished surface is strictly the same, and, thirdly, the uranium and thorium contents are in their perfect radioactive equilibrium. Firstly, however, the geometrical evenness is hopeless in the artificially polished surface of a thin section some 30 microns in thickness. Secondly, some air gaps are always inevitable between the emulsion and the imperfectly evened surface, for example, the air gap caused around an allanite grain, as already pointed out in the former report II,[3] an alpha track diffusion of some 0.1 millimeter. But an air gap of one millimeter is nearly nothing, if the incomparably long alpha range in the air is considered, and the radioactive effect depends chiefly upon the surface magnitude of the mineral. The different hardness of the minute mineral keeping the surface uneven against the polishing as shown in Figure 3, makes the radioactive effect more remarkable than in an even surface, so that our radioactive comparison of two grains by means of only their surface magnitude under the microscope is rendered more difficult. The radioactive comparison is, nevertheless, effective, when the two grains are found in the same or nearly the same surface conditions, namely, when they are of the same mineral kind and included in the same host mineral. If one may stands higher, while the other rather lower, than the even surface A, as B and C in Figure 3, generally, the B condition makes long tracks more, while the C condition renders them less, than the A condition does. The discrimination of their surface condition is often rather easy, if the radial distribution of alpha tracks and the size of the grain ejecting them are carefully observed. That is, owing both to thus enlarged polished surface and to the air gap between it and the emulsion, a mineral in B condition scatters the multipled long tracks farther away, whereas a grain in C condition rather concentrates, on the emulsion, high angled tracks almost vertically around it. Even in case a grain in A condition, its track length must be carefully observed, apart from the afore said theoretical or ideal value, due to the different permeability between the grain itself and its host mineral. Rather the big role that the grain size and shape play herein, should be remembered. The best comparable is of course between either two inclusions or two hosts belonging to the same mineral kind. Among quartz, feldspar and biotite, the difference in permeability is not remarkable. The distribution of track length of uranium and thorium out of their radioactive equilibrium resemble those in Figure 1 and Figure 2. And a considerable deviation from Figure 1 or Figure 2 in their distribution, in turn, confirms us of the remoteness of these two elements from their radioactive equilibrium. On this supposition, the minute radioactive inclusions in Kitashirakawa granite were studied.

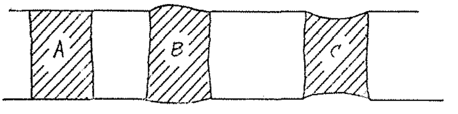


Figure 3
Minute radioactive minerals of various hardness in a thin section of rock.

The minute radioactive minerals in Kitashirakawa granite

This biotite granite is adamellite.

1) (Sample K-21-e) A transparent zircon grain *a* (52×122 microns in size) included in quartz offered, after 28 days' exposure, 74 tracks of diverse length distributed just as illustrated by *a* in Figure 4, which, if plotted upon Figure 2, produces Figure 5 and makes its radioactive content reducible mainly to uranium. Figures 4 and 5 designate how the tracks of different length,

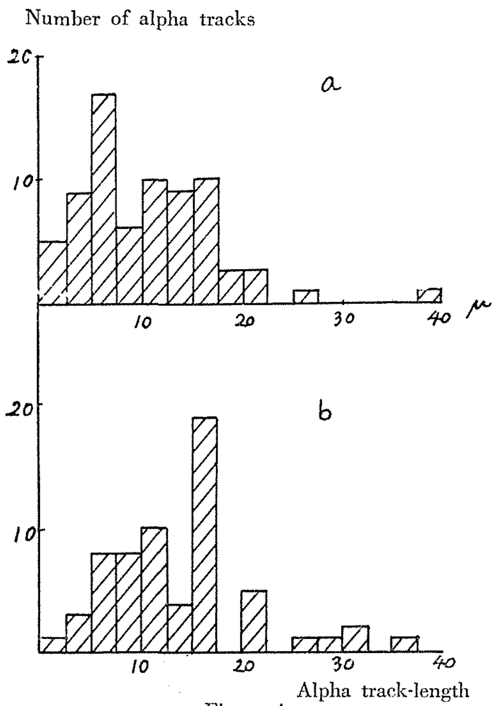


Figure 4
Histogram of the alpha track-length
(a) zircon (b) red mineral

whose sum total is fewer than about 100, are distributed. Another grain *b* red and transparent (28 × 28 microns) with no definite form and adjacent to *a* left, after the same exposure, 63 tracks distributed as designated by *b* in Figure 4 and *b* curve in Figure 5. If these two grains are supposed to be in their radioactive equilibrium, the greater plenitude of long tracks, and consequently the greater abundance of thorium content in *b* grain than in *a*, is clear from the comparison between the two curves in Figure 5. The possible fluctuations in the distribution of diversely lengthened tracks often prevent the curves of this sort from falling between

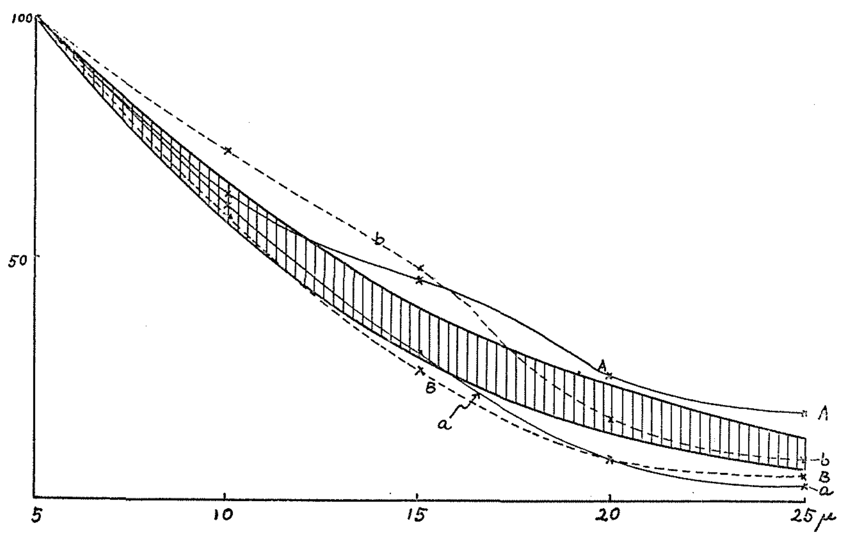


Figure 5 Thorium-Uranium tendency of the minute minerals

the uranium and thorium curves.

2) (Sample K–31) two transparent grains A and B found in intergrowth around a biotite flake have emitted their differently lengthened tracks as shown by A and B in Figure 6. The grain A, granular and $0.165 \times 0.01 mm^2$ in size, offered after 28 days' exposure 35 tracks, whose distribution curve indicates more thorium content than uranium one in it. Despite its zircon like appearance, this grain may be identified either as monazite or as xenotime, because of its small birefringence and of its abundant thorium content. The other grain B, with zircon like fine crystal form and $0.85 \times 0.01 mm^2$ in size, ejected during 28 days' exposure 157 tracks, whose distribution informs us that of uranium to be super-abundant in it.

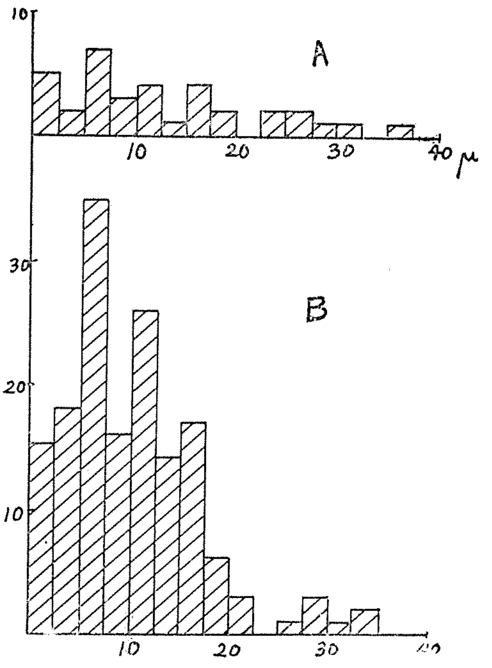


Figure 6 Histogram of the alpha track-length

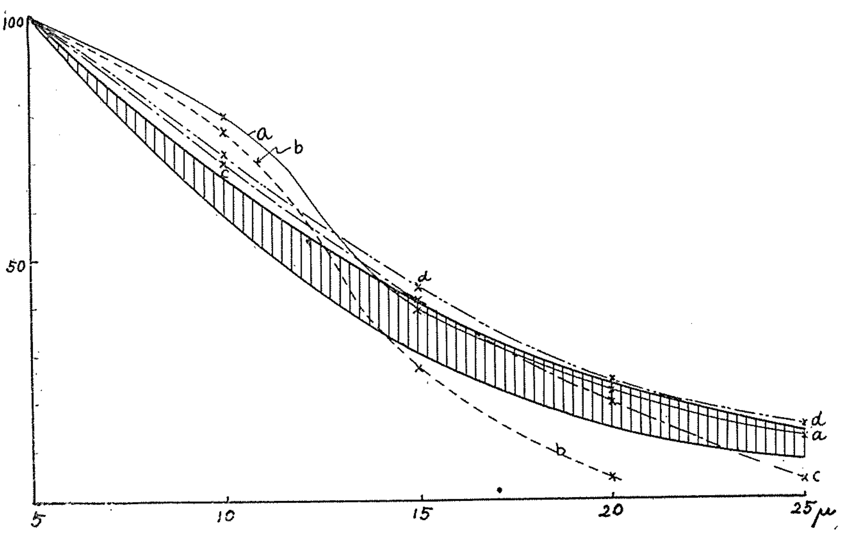


Figure 7 Thorium-Uranium tendency of the minute minerals

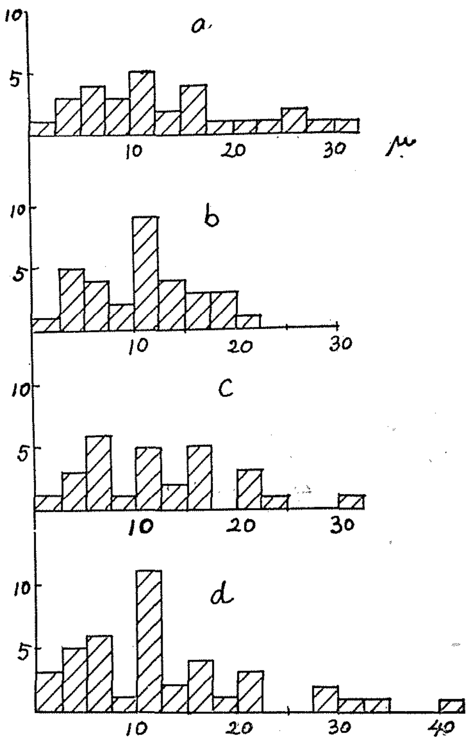


Figure 8 Histogram of the alpha track-length

3) (Sample K–29) Three zircon like grains (*a*, *b* and *c* in Figure 8) emitted during 4 weeks' exposure 29 and 32 and 28 tracks, respectively, whose distribution curves are illustrated in Figure 7 by *a*, *b* and *c*. Although the greater thorium abundance in *a* grain is obvious from *a* curve, the uranium thorium ratio in two other grains remains still indeterminable from *b* and *c* curves. Grain *c* promises us a more definite result obtainable from the far greater track number by much prolonged exposure, while grain *b* interests us with its greatly deviated distribution curve, somewhat resembling curve *b* in Figure 5, and probably due either to the nonequlibriated radioactive condition, or to the extreme heterogeneity in radioactive distribution, in the grain itself. Now grain *d* in Figures 7

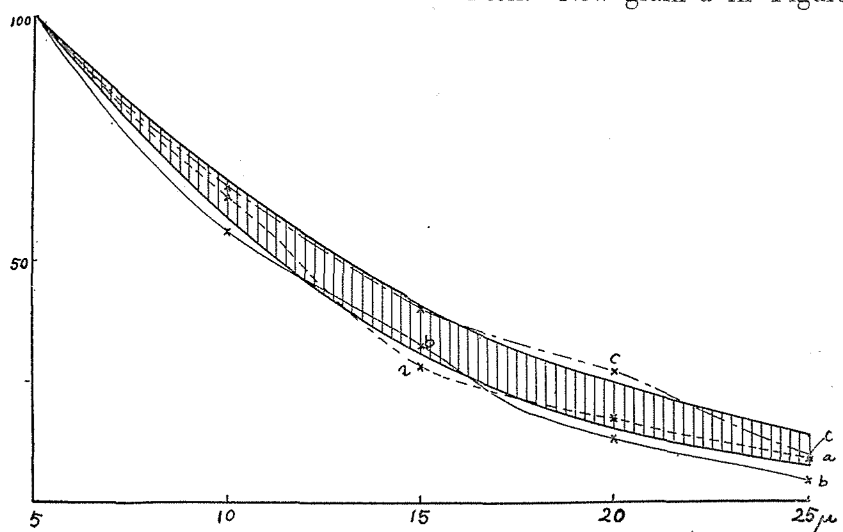


Figure 9 Thorium-Uranium tendency of the minute minerals

and 8 is included in plagioclase, rather flat and rhombic in crystal form, low in birefringence, monazite like in appearance, has emitted during 4 weeks' exposure 42 tracks, and yet it is evident, from this small total track number, that its radioactive content is mostly thorium, as curve *d* in Figure 7 shows it.

4) (Sample K–21–f) Of two zircons like grains *a* and *b* in Figure 9 contained in the same thin section, the former is $0.69 \times 0.01\ mm^2$ in size, situated between quartz and biotite, $T\alpha = 0.49$ in radioactivity, ejected during 28 days' exposure 82 tracks, whose distribution curve is designated by *a* in Figure 10. The latter, namely, zircon grain *b* in Figure 9 is included in albite, $1.22 \times 0.01 mm^2$ in size, and $T\alpha = 0.45$ in radioactivity. It is well exemplified in this grain that, in case the total track number surpasses 100, the uranium thorium ratio is determinable with far greater accuracy. Both these grains are abundant in uranium, but grain *a* seems to include also some thorium. This tendency of including some thorium besides uraniferous content is still more clearly seen in a grain (K–27) which is 0.15 $\times 0.01 mm^2$ in size, contained in biotite, seeming from its appearance under the microscope to be something else than zircon, $T\alpha = 3.04$ in radioactivity, and 114 in total track number, whose

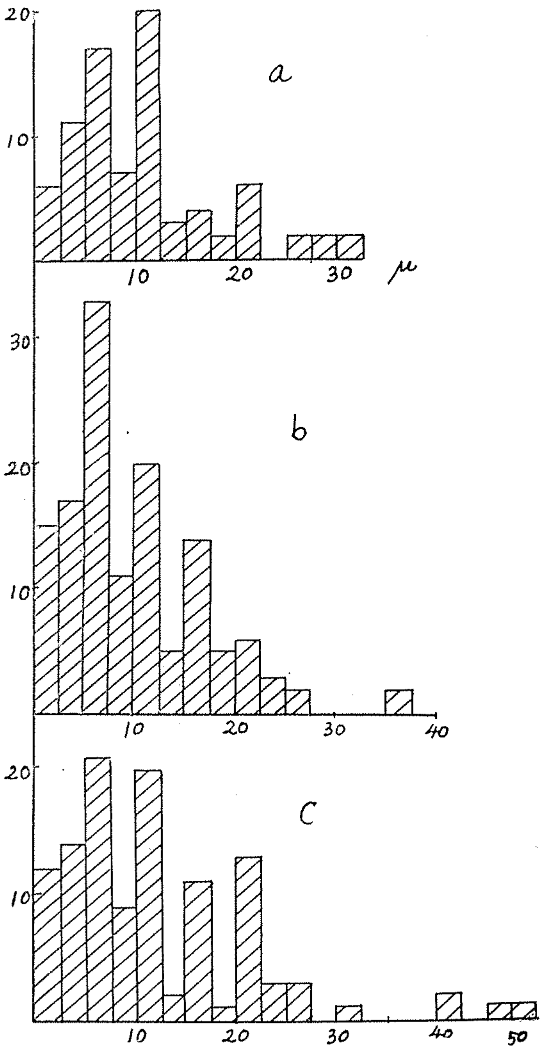


Figure 10 Histogram of the alpha track-length

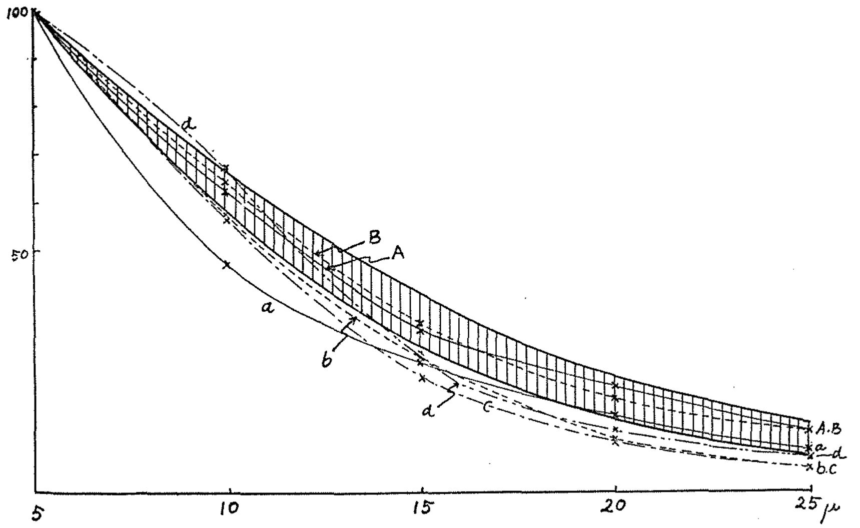
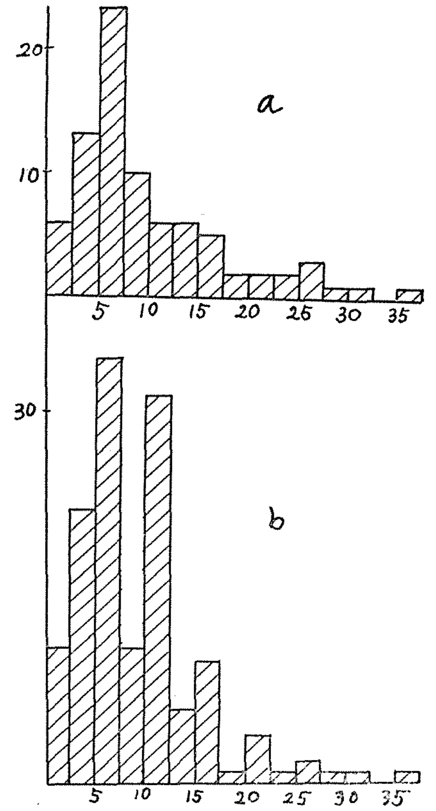


Figure 11 Thorium-Uranium tendency of the minute minerals

distribution curve is given as *c* in Figure 10. Its two long tracks between 45 and 50 microns seen in Figure 10 naturally prove thorium contents in it, while the distribution curve of its diverse tracks designates thorium as the principal radioactive element in it.

5) Of the two zircon grains *a* and *b* in Figures 11 and 12 contained in a granite (K–16), the former is included in biotite, while the latter in quartz; $T\alpha=1.27$ and $T\alpha=2.26$ in radioactive order, and 81 and 136 in total track number, respectively. Uranium is principally regarded as radioactive element alone from the entire absence of tracks longer 36 microns among so many tracks. Two zircon gains (K–24 and 26) are 90 and 77 (Figure 13) in track number and, like zircon samples (K–16), of uraniferous tendency in radioactive content. It is known by the four examples (*a, b, c,* and *d*) that the experimented uranium curve of differently lengthened track distribution stands somewhat lower than the theoretical uranium curve. Distribution curves A and B in Figure 11, the results obtained from two allanite grains (K–15 and 29), fall rather near the uranium curve, despite our expectation of their approximation to the thorium curve, as thorium is their almost sole radioactive content. Like the above-given four zircon grains, here is seen also the tendency of the experimental curve falling rather lower in figures than the theoretical curve.

Minute crystal with thorium pleochroic halo in Kitashirakawa granite



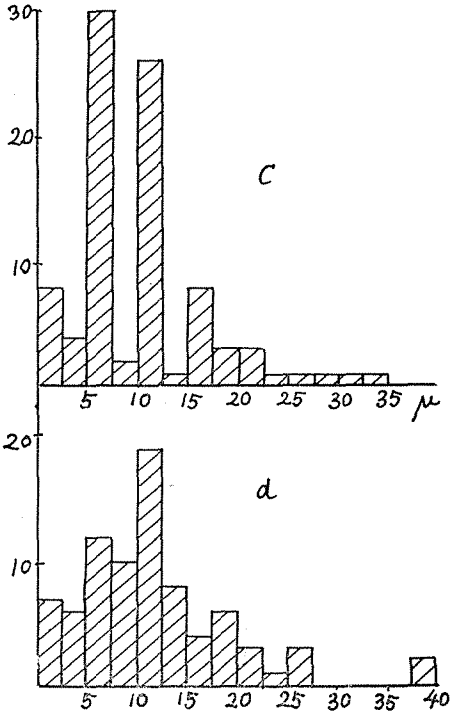


Figure 12 Histogram of the alpha track-length Figure 13 Histogram of the alpha track-length

(K–16–2) shows the track length ratio 19 percent (track number longer than 20 microns/total track number), and consequently it seems mineral rich in thorium. Minute monazite in plagioclase with $T\alpha=0.89$ shows the ratio 19 percent. Zircon in plagioclase from the dyke granite of Ikenojizo, Kita-shirakawa granite, shows the ratio 7.7 percent and it may be uranium only as the radioelement. Zircon grain in the granite of K–9 (2) near the contact margin has the ratio 17 percent and it contains thorium and (uranium) as the radioelements.

From the Figure 1, if thorium uranium ratio is infinite, the long alpha track over 20 microns must be 17 percent of the total alpha track and if the ratio is zero (uranium only) the long alpha track must be 8.6 percent. Experimental result shows about the same order.

The minute radioactive minerals in Tanakamiyama and Mushima granite

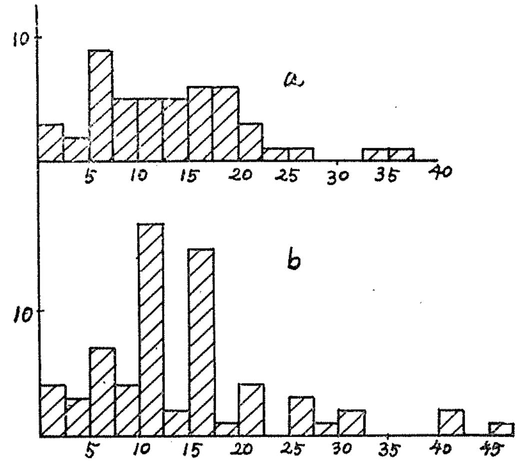


Figure 14 Histogram of the alpha track-length

The leucocratic coarse grain granite (T–298) from Daradani contact margin of Tanakamiyama contains zircon ($0.048 \times 0.01\ mm^2$, $T_\alpha = 1.38$) interstitial between quartz and albitic felspar. The track length distribution of this zircon shows Figure 14 (a) to be uraniferous from the ratio between the number of alpha tracks longer than 20 microns, and the number of total alpha tracks is 14.6 percent. The other grain of zirconic minute mineral in orthoclase ($T_\alpha = 12.1$, pale yellowish color, low baief.) shows thorium rich from the ratio as 19.7 percent. (Fig. 14 b). The same thin section of rock has the reddish brown ferro-manganoan deposit at the interstitial boundary of rockforming minerals. These material (Fig. 15) seems from the shape and size as secondary origin. The alpha activity of these four grains A, B, C and D are $T_\alpha = 0.307$, 0.258, 0.246 and 0.154 included in the micaceous part. The ratio of the alpha tracks shows 15.9, 14.4, 17.8 and 16.8 percent, showing it a mineral rich in thorium but fairly in uranium. The alpha track length distribution shows Figures 16 and 17. These data clearly indicate the same result protting

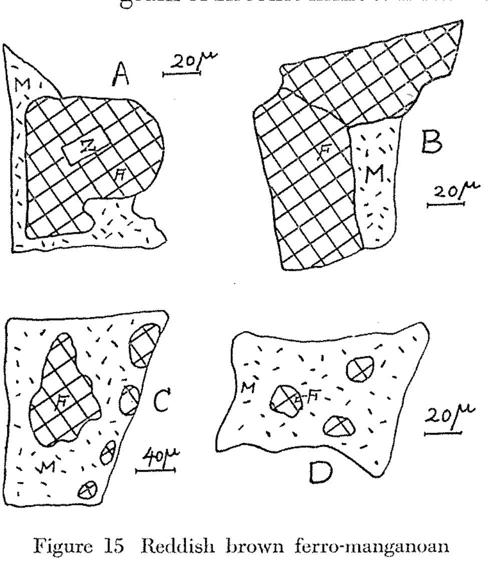


Figure 15 Reddish brown ferro-manganoan mineral (F) Z—zircon M—micaceous material

on the curve in Figure 2.

The Tanakamiya aplite (T–770 I) near the contact hornfels has radioactive zircons, $T_\alpha=1.18$ (a), 3.56 (b), 3.43 (c), and their track length distribution shows Figure 18 which indicates (a) uraniferous, (b) thorium-rich, (c) thorium-rich in the track length ratio (a) 14.6 % (b) 34.7 % (c) 17.6 %.

Minute zircon from the granite of Mushima island, Oda County, Okayama Pref., shows feeble radioactivity despite of the 84 days exposure. Their radioactivities are (a) $T_\alpha=0.15$, (b) 0.12, (c) 0.13 and (d) 0.19, and the track length ratios are (a) 20.0 % (b) 6.6 % (c) 5.1 % and (d) 10.0 %. The grain (a) is included in orthoclase, (b) (c) (d) in green hornblende, and the first shows rich in thorium and the others uraniferous (Figure 19).

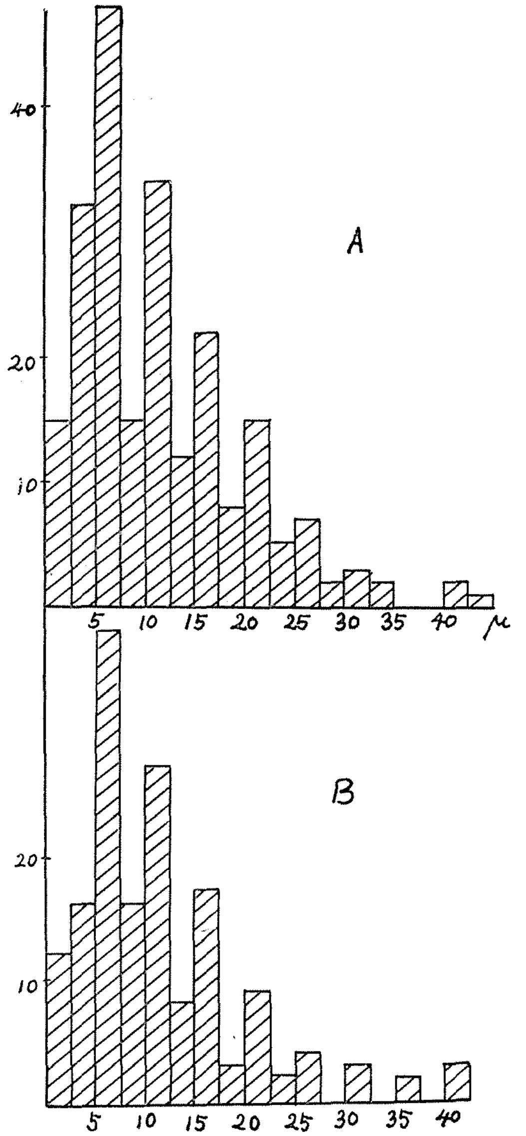


Figure 16 Histogram of the alpha track-length

The mineral determination by their uranium thorium content in minute grains

It became evident from the foregoing examples that different length of alpha tracks renders the uranium and thorium content semi-equantitatively determinable. The present studies on granites are concentrated in the uranium thorium content determination in so feebly radioactive inclusions that each of which will probably remain beyond all our chemical measurement of those two

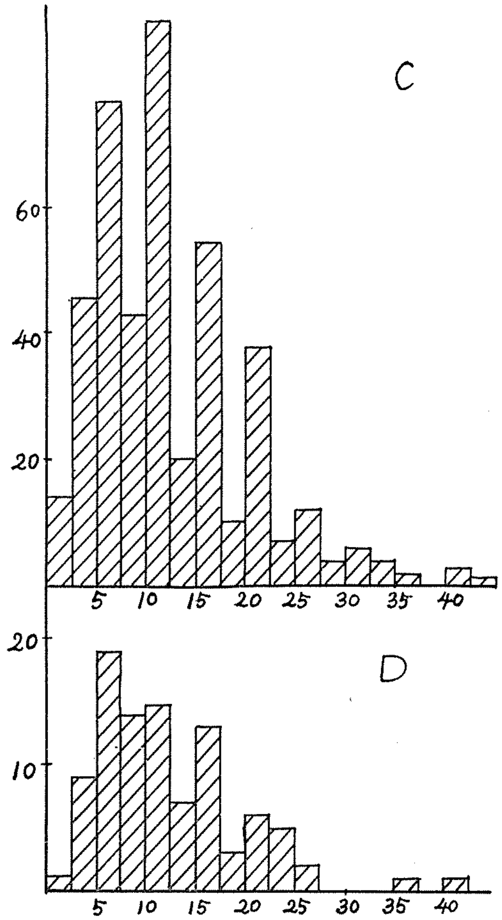


Figure 17 Histogram of the alpha track-length

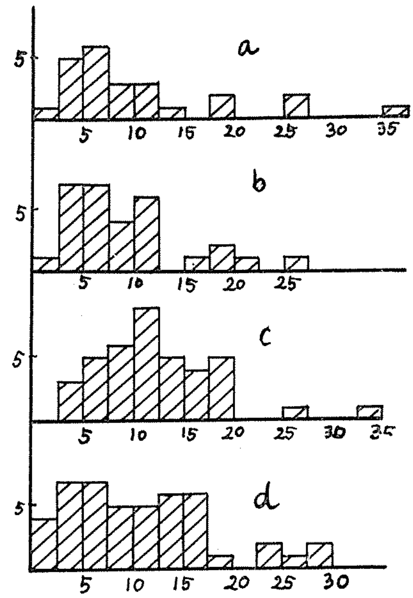


Figure 19
Histogram of the alpha track-length

elements in it.

The results of our experiment have shown us that the zircon-like minute grain fresh in their microscopic appearance seem, with the good harmony between the real track distribution of different length and the theoretically presummed one, to be in their perfect radioactive equilibrium.

At the same time a few secondary seeming mineral grains remain doubtful of their radioactive equilibrium owing to their peculiar distribution curves of differently lengthened tracks.

For the thorium uranium ratio determination at least 50 tracks are needed, while 100 or 200 are the most favourable, though in some rare cases

20 or 30 are sufficient.

So far as Kitashirakawa granite is concerned, zircon grains were almost invariably found in content of uranium series and in radioactive equilibrium, whereas thorium superabundance was remarkable in monazite and allanite grains. By way of the measurement of different track length, the most interesting are the secondary or weathered minerals out of their radioactive equilibrium, the interpretation of which will follow to some extent, as soon as the data thereof are gathered sufficiently enough for it.

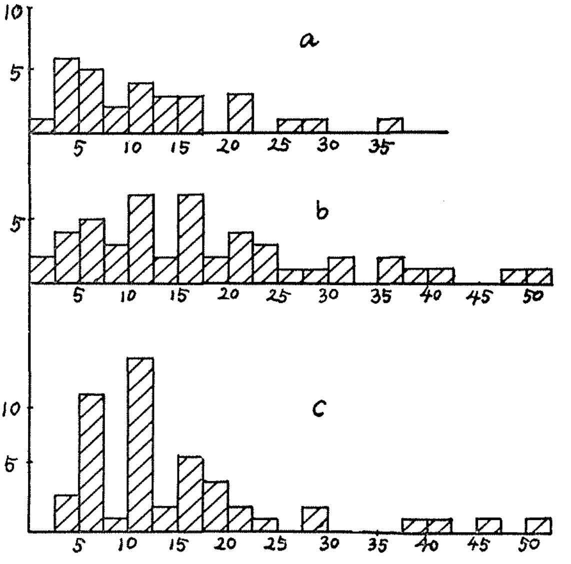


Figure 18 Histogram of the alpha track-length

Fluctuation of alpha track number by the exposure interval

The number of alpha track must be proportionate to the exposure interval by the track autoradiography of radioactive minerals. These experiments were tried with the same thin section in the same or different exposure time. On the several samples of Tanakamiyama granite, the satisfactory results were obtained from the 28 days exposure (Table 1 a). Repeated 28 days exposures, the alpha track number is comparable with fairly quantitative exactness. The data cross to some degree between 7 days exposure and 28 days one. (Table 1 b) They cross, still more, between the data of several hours exposure. The data from Table 2 (a) shows the remarkable fluctuation. This fluctuation appears to be caused by the secondary mineral with high radioactivity, not on the fresh zircon grain of feeble radioactivity.

The effect of radon leakage, nevertheless, does not appear by the long time exposure such as 28 days. Radon and even thoron leakage must affect on the short time exposure.

One minute radioactive mineral ($T_\alpha = 18.85 \pm 1.24$) with yellowish color

Table 1

28 days exposure		(a)
Sample	1 st trial	2 nd trial
K-894	93	98
	47	46
	45	47
	64	67
	27	23
	75	74
K-889	85	87
	49	48
K-423	32	31
	21	23
	21	20
	25	29
	55	58
7 days and 28 days exposures (b)		
	7 days×4	28 days
K-211 (1)	68	46
	84	54
	60	67
	128	112
	68	70
	24	25

Table 2

Sample	Grain size (Microns)	Exposure time (hours)					
		24 h	18 h	15 h	18 h	18 h	20 h
K-10(2)	22×58	11	19	11	19	17	22
K-2	70×53	55	31	51	44	46	55
K-17	34×37	45	21	24	33	35	37
K-23	52×47	41	21	21	37	33	40
K-25	34×22	16	22	15	7	17	17

Table 3

Sample	Exposure time	Total track number	Track number longer than 20	%
K-2	24 h	55	13	23.6
	18 h	31	5	5.2
	15 h	51	10	19.6
K-10(2)	24 h	11	2	18.2
	18 h	19	5	26.4
	15 h	11	4	36.4

and moderate birefringence is contained in the Kitashirakawa granite (K-2). Figure 20 shows the variation of alpha track length distribution, and the ratios of long alpha track over 20 microns (Table 3) differ from each other. The ratio of long alpha track is small, when the total alpha track number is small. Probably, this owes radon or thoron leakage. Recently Giletti and Kulp[4] and Picciotto and Salvetti[5] assured the reports on the powdered rock on these subject. Elements of the post emanation group in the thorium uranium series eject the long range alpha particles, and loss of one atom of radon or thoron becomes the reduction of three alpha tracks from the photo plate. This loss appeares conspicuous, especially at the short time exposure.

The mineral grain of the granite (K-10 2) has its radioactivity as $T_\alpha = 20.82 \pm 2.03$, and it shows the metamict state with zirconic appearance including in quartz (Table 3). This example shows the same tendency of the track length (Fig. 21) that the total track number is smaller, the alpha track number longer than 20 microns is fewer.

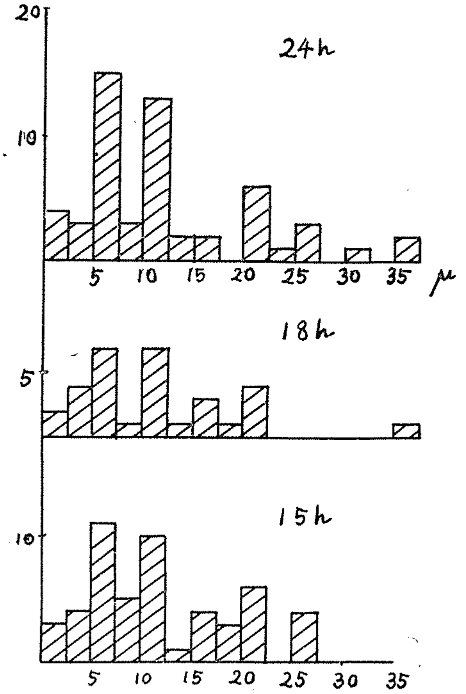


Figure 20 Histogram of the alpha track-length with the same mineral grain

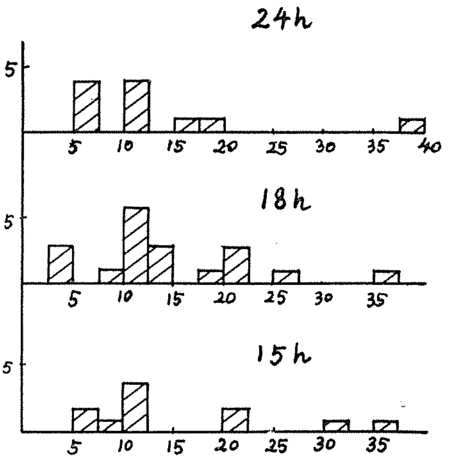


Figure 21 Histogram of the alpha track-length with the same mineral grain

Thorium uranium ratio measurement by means of absorber

1) Method In the above paragraphs, the length of alpha track was measured and then thorium uranium ratio was determined. This method is a very laborious task. When moderately large number of alpha track

Table 4

	Air (cm)	Range Emulsion (μ)	Silver foil (μ)
Th C′	8.57	54.6	23.8
Ra C′	6.91	44.0	19.2
Ac A	6.46	41.1	17.9
An	5.67	36.1	15.7
Th A	5.64	36.0	15.7
Ac C	5.36	34.1	14.9
Th C	4.73	30.1	13.1
Ra A	4.46	29.7	13.0

is obtained, long alpha tracks vertically ejected from the minute mineral can be selected. On the three natural radioactive series, long range alpha particles show Table 4. To catch only the vertically long alpha track, metalic foil acts as an absorber by the insertion between the thin section of rock and photo plate. In case of insertion of absorber equivalently thick to the alpha range of Ra C', photo plate catches only the residual track of Th C'. Thorium uranium ratio can be measured by the ratio of this residual track number and total track number.

2) Material for metalic foil. Metalic foil used as absorber must have the following natures, (a) thickness of the foil must be even, (b) known constant for the conversion factor of the alpha particle, (c) unaffection to the nuclear emulsion during the long time exposure, (d) unaffection of the metalic foil itself even during the long time exposure and (e) obtainable easily for the various thickness of the foil. Silver foil fits to this purpose, but aluminium foil does not.

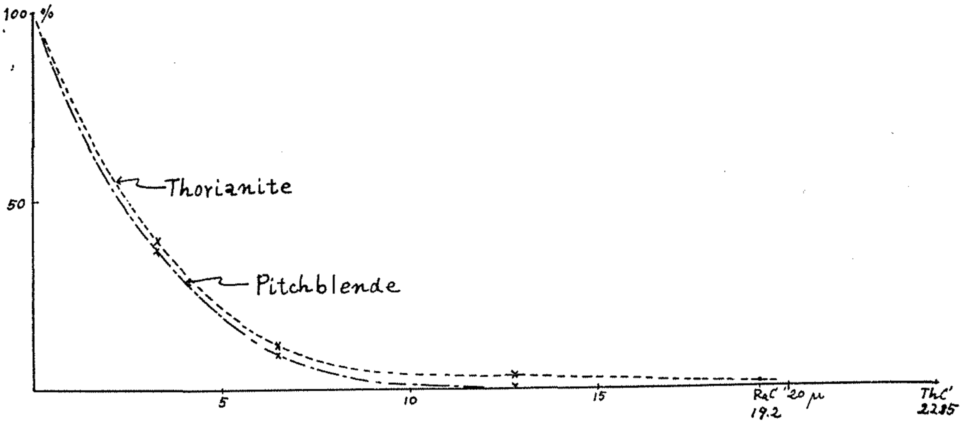


Figure 22 The relation of the thickness of silver foil with the number of residual alpha tracks

3) Thickness of silver foil and the number of residual alpha tracks.

Various thickness of silver foil are used for pitchblende from Joachimustal and thorianite from Ceylon. Figure 22 shows the relation of the thickness of silver foil with the number of residual alpha tracks. From this figure, comparatively many residual alpha tracks can be obtained with about 7 microns silver foil, but can not be distinguished whether they are uranium or thorium minerals. The ratio of residual alpha tracks and total alpha tracks varies almost linearly according to the thorium uranium ratio. Theoretically, 19.2 microns silver foil (RaC' alpha range equivalant) produces only ThC' residual alpha tracks, and then thorium measurement is expected possible. Experimentally, only 0.3 percent of this ratio was obtained even in case of Ceylon thorianite. This is not practical for the long time exposure.

4) Microscopic observation of high angle, short residual alpha track

Figure 23 shows the geometrical relation of the residual alpha tracks absorbed by silver foil. These alpha tracks in the emulsion is not easy to count even under the high power microscopic observation. Only under the high power immersion method, the counting or scanning of alpha tracks is

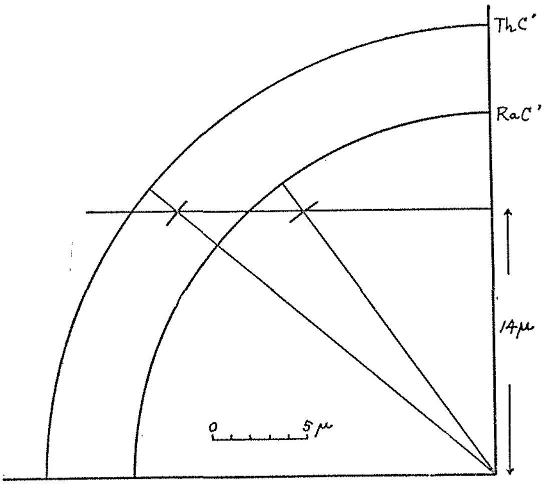


Figure 23 The geometrical relation of the residual alpha tracks absorbed by the silver foil (14 microns in thickness)

Least discernible alpha track: 5 microns in the ET-2E nuclear emulsion (2.14 microns in the silver foil)

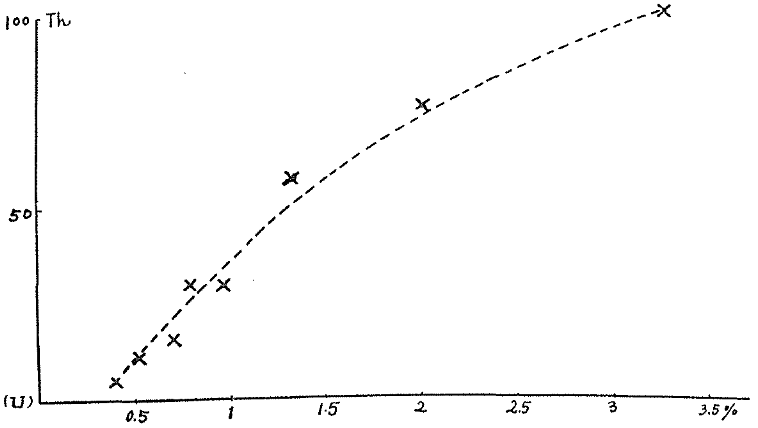


Figure 24 The ratio of the residual alpha track with silver foil of 14 microns in thickness, using the minerals of known contents of radio-elements.

Ordinate—Thorium-Uranium ratio

Abscissa—The ratio of residual alpha track easily performed.

Example of the known content of several radioactive minerals

Table 5 shows the radioactive minerals already known uranium and thorium content. These minerals may be heterogeneous distribution of radioactivity and even in radioactive unequilibrium condition. These theme will be touched in the following report. The ratio of the number of residual alpha tracks is 0.4 percent for uranium-rich euxenite, 2.34 percent for thorium-rich monazite, with the silver foil of 13.94 ± 0.14 microns thickness. This result shows that the exposure time for the residual alpha track must be increased about 100 times to obtain the same order of normal alpha track number. Figure 24 shows the tendency of these relations, and applies for any radioactive equilibriated minerals of unknowen quantity of thorium and uranium in them.

Some application to the thin section autoradiography

One minute monazite crystal included in biotite in the Tanakamiyama granite (T–750) ejected the 616 alpha tracks for 7 days exposure, and the 78 residual alpha tracks with the 14 microns silver absorber for 114 days exposure. The ratio of the two alpha tracks number is 3.11 percent and this grain shows almost thorium as a radioactive source from Figure 24. Zircon grain of the same thin section of rock shows the radioactivity $T_\alpha =$ 2.40, and the track number ratio is 2.23 percent. This mineral may be fairly rich in thorium. Minute grain with brownish stain and with pleochroic halo may be thorium rich mineral from the ratio as 2.64 percent.

At the present stage of this study, the curve of Figure 24 does not exactly quantitative due to the heterogeneous distribution of the standard radioactive minerals. This method is now proceeding to detect the heterogeneity, radioactive inclusion and no equilibrium conditions of the radioactive mineral. These data will soon be published.

Conclusion

1) Thorium uranium ratio of the mineral can be measured by the alpha track length distribution.
2) Care must be taken to the leakage of radon or thoron for the short

time exposure.

3) Thorium uranium ratio can easily be measured with the insertion of the silver foil (14 microns in thickness) between photo plate and the thin section of rock.

4) The distribution of radioactive material in the minute mineral can be detected exactly by means of silver foil absorber.

5) The minute radioactive minerals have often heterogeneous distribution of thorium uranium ratio in one grain.

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