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The Radioactivity of Rocks and Minerals Studied with Nuclear Emulsion VII

Radioactivity of Granitic Sphene

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Abstract

Irrespective of their different rock types, granitic sphenes contained in four of Japanese and Korean rocks vary in their radioactive order from T_α=0.000n to 0.1165. Activity varies according as the color of sphene and its position in the thin section of rock differ. The feeble radioactivity found around the sphene must not be neglected, for it seems to have been aboriginal already in the rock itself rather than contaminated, afterwards, during the polishing of a thin section. The *Th-U* ratio seems to be about 1 in sphene. A heterogenetic variation is seen even in a single crystal of sphene.

Introduction

Sphene is often contained as an accessory mineral in plutonic rocks, especially in alkali hornblende granite, granodiorite and quartz diorite.

As A. HOLMES reported already in 1918 on hornblende gneiss of Mozambique[1)], in case of acid plutonic rocks containing hornblende, sphene furnishes those rocks more weight in radioactivity than zircon and others do. In Mozambique gneiss, rich in sphene and scanty in zircon, its radium content was 2.13×10^{-12} g/g. HUTTON[2)] says that the sphene of NW Nelson, New Zealand, contains 0.28 percent of thoria. The radium contents of pegmatitic sphene were, according to R. J. STRUTT[3)], 51×10^{-12} g/g Ra and according to N. SAITO[4)], 28.15×10^{-12} g/g and 16.47×10^{-12} g/g Ra. These values, if calculated in term of uranium content in radioactive equilibrium, are 15×10^{-5} g/g, 8.3×10^{-5} g/g and 4.8×10^{-5} g/g uranium respectively. Autoradiographic method was carried out on this mineral by R. COPPENS[5)] and his alpha radioactive data were T_α=0.015 or 8×10^{-5} g/g uranium equivalents.

The existence of pleochroic haloes around the sphene presupposes slight radioactivity. Its colored host mineral is mostly green hornblende, and the halo, as a rule, is very faint. As often repeated by the present author, sphene greatly varies in its radioactivity—even in one thin section of rock—this radioactive diversity is evident already from the fact that, in the same thin section, some sphenes are surrounded by haloes, while others are lacking them.

Method

The radioactive order of such sphene was studied by means of autoradiography from their thin sections upon the nuclear emulsion (Fuji Photo Film Co. Ltd.) For the determination of their radioactive order, however, a long duration of exposure is required, because of their extremely feeble activity. Their order, nevertheless, is detectable because of their much larger grain size than zircon and other radioactive minute minerals. The emulsion always records and presents an integrated picture of the incident radiation, and this is of great importance in the study of feeble radioactive minerals. The exposure may extend 8 weeks, and even 12 weeks are never too long, and in one case 311 days' exposure was in use. Special care must be taken, so that the long duration of the exposure may not cause the fading or fogging of the photo plate.

For the alpha track counting under the microscope, recommendable is the already mentioned procedure[6]. For this purpose, however, the sphene sample must neither include inside itself any radioactivie zircon and other minute minerals nor lie so near as 0.2 millimeter to any of them.

Examples of the several rock samples

The author's samples are of alkali granite (Korea), hornblend granite (Katanoyama, Osaka Prefecture, and Kinbusan, Yamanashi Prefecture), and quartz diorite (Kurama, Kyoto City), in which hornblende and coarse grained sphene often make an intergrowth. In case of such an intergrowth, generally, the larger the radioactive content of the sphene is and the older the geologic age of the rock is, the darker the pleochroic haloes appear in the hornblende, though mostly quite faint. Even in the same thin section sphenes show diverse degrees of halo blackness, and this indicates that they are much different in radioactive content. The radioactivity of each sphene is given in Table 1 (its shape and size are shown in Fig. 1) and Table 2. The result of Table 1 was obtained from the 8 or 12 weeks exposed photo plates, and that of Table 2 was derived from the 311 days' exposure, for extremely feeble radioactive grains can not be measured by 8 or 12 weeks' exposure. The grains A–M, N–P, Q–S and T–Y in Fig. 1 belong respectively to that of the alkali granite in Korea, the Kinbusan hornblende granite of Tertiary age, the Katanoyama hornblende granite and the quartz diorite of Kurama, Kyoto City. Thesegranitic rocks have usually green hornblende.

The datum of "O" in Table 1 and Fig. 1 (Kinbusan granite) shows the radioactivity of the sphene with zircon-like inclusion.

Fig. 2 shows the relation between the grain size of sphene and its radioactivity listed in Tables 1 and 2 of the present author and also in that of COPPENS[5]. In

Table 1 Radioactivity of granitic sphene

Sample	Size (0.01 mm²)	Radioactivity (T_α)
A	2.42	0.0785
B	3.03	0.0434
C	2.25	0.0426
D	2.25	0.0339
E	1.24	0.0289
F	1.61	0.0289
G	2.58	0.0277
H	7.70	0.0190
I	1.68	0.0186
J	4.23	0.0157
K	10.80	0.0095
L	12.40	0.0087
M	6.34	0.0079
N	1.22	0.0752
O	(9.06)	(0.0378)
P	3.30	0.0149
Q	2.65	0.0794
R	11.92	0.0289
S	14.02	0.0252
T	1.45	0.1165
U	3.70	0.0843
V	4.13	0.0351
W	4.95	0.0277
X	9.76	0.0153
Y	14.60	0.0107

A—M Ham bug Gyōng Sōng Gun Zyu nan mōn, Korea
N—P Kinbusan, Yamanashi Prefecture, Japan
Q—S Katanoyama, Oosaka Prefecture, Japan
T—Y Kurama, Kyoto City, Japan

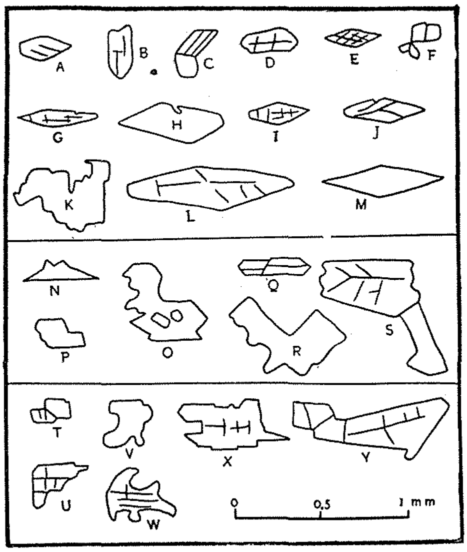
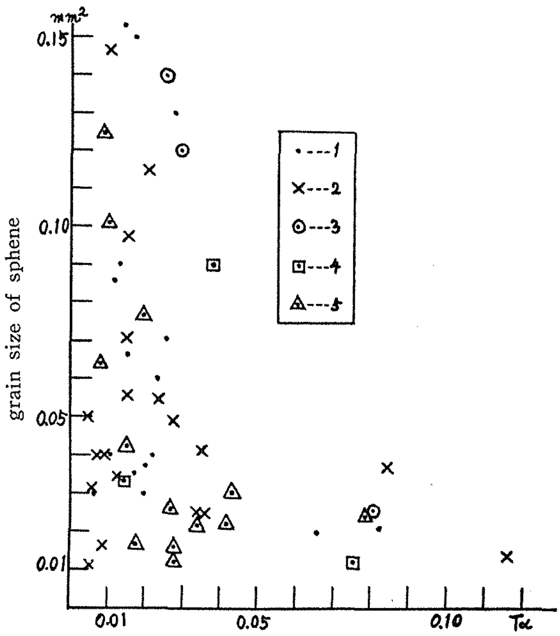


Fig. 1.



Radioactivity
1···Coppens' data 4···Kinbusan
2···Kurama 5···Korea
3···Katanoyama

Fig. 2.

that figure, however, it must be remembered that sphene of feebler radioactivity than $T_\alpha=0.01$ required 311 days' exposure, and that such sphenes must be far more in number, though various inconviniences disturbed their radioactive measurement. The grain larger than 0.15 mm square is eliminated from the data. Notwithstanding all these, Coppens' and the author's data fairly agree with each other.

It seems that the radioactivity of granitic sphene does not much differ by their rock composition, namely, whether they are basic or acid rocks. The sphenes of fairly high radioactivity (over $T_\alpha=0.05$) exist always as smaller grains—in granitic rocks of three different countries of the world. But all the minute crystals of sphene are not necessarily highly radioactive.

This significant fact which Fig. 2 suggests is that there lies in sphene regular and inverse relation between the grain size and the radioactivity; roughly speaking, that a certain amount of radioactive element was deposited in each sphene of most granitic rocks, and this is true of all the samples the author studied.

Table 2 Radioactivity of sphene (Kurama quartz diorite)

Sample	Size (0.01 mm²)	Radio-activity (T_α)	Color
Kurama 7	2.25	0.0354	
2	2.44 *	0.0341	
8	5.57	0.0235	brownish
7	11.50	0.0202	pale brown
6	5.68	0.0158	
6	7.16	0.0153	
6	3.41	0.0120	
8	3.97	0.0091	pale blue
6	1.60	0.0087	
6	3.92	0.0077	
2	3.17 *	0.0055	
2	1.71 *	0.0050	
8	5.08	0.0045	
6	large	0.0227–0.0260	
1	"	0.0078	pale yellow
6	"	0.0093	
6	"	0.0279	brown
6	" **	0.0435	brown part
	**	0.0016	intermediate part
	**	0.000n	transparent part

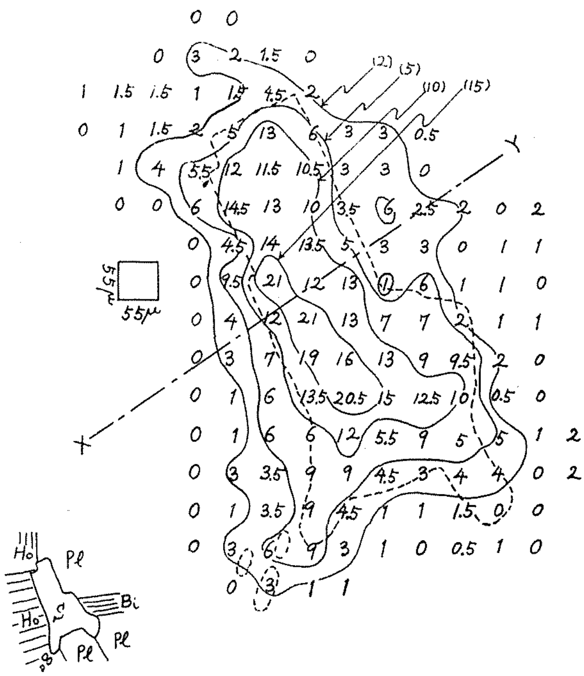
* sphenes of Figure 5
** in the same crystal

The accessory sphenes show wide variation in their radioactive order. (This was already told in a former paper[7], especially on zircon of Tanakamiyama granite). This tendency seems to be universal when rare elements like uranium and thorium are deposited as accessory minor constituents in plutonic rocks in general. Big as the ionic size of uranium and thorium may be, they are concentrated, accompanied by iron or rare earths[8], into sphene. It is improbable that, in the crystal lattices, these accessory constituents are regularly and homogeneously distributed all over the grain.

The radioactive order of a sphene is sometimes known by its color; for instance, brown one excels the pale bluish and the transparent, as is very clearly seen in Table 2, (see **) and especially in its lowest line which treats the color-and-radioactivity relation in differently colored parts of a sphene crystal. This relation leads us to the idea that, in their deposition into these grains, a quantitative parallelism is quite probable between rare earths and radioactive elements.

Fig. 3 shows the radioactive distribution in a sphene (the 4th one from the top in

Table 2) contained in Kurama quartz diorite, Kyoto City. To get this distribution map, the alpha track counting was carried on in every 55 microns square, and this procedure was done in the best possible adjustment of the photo plate on the thin section. In this map the sphene is outlined with the dotted line and well agrees with the radioactive distribution. In the mineral there is no special radioactive center nor remarkable heterogeneity whatever. In a sphene from Misasa granite (coarse biotite granite No. 248) the radioactivity is $T_{\alpha}=0.0115$ (not listed in the tables).



S···sphene Bi···biotite Pl···plagioclase Ho···hornblende

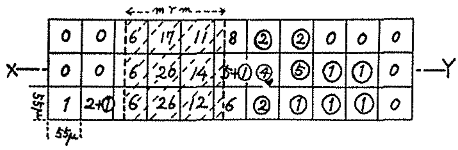
The number shows the alpha track number in 55 microns square after the 311 days' exposure.

Fig. 3. Distribution of alpha tracks from sphene (Kurama Quartz diorite)

Radioactivity around the granitic sphene

Feeble activity is often seen around the minute radioactive minerals in the thin section of granitic rock. It seems improbable that this feeble activity results from air gaps of groove around the minute mineral, although the author was once of that opinion[9]. The exact microscopic coincidence of alpha track pattern with the minute radioactive mineral clearly indicates that these feeble activity is ejected from the neighboring rock-forming mineral. Here the direction and the dip of each alpha track and its distance from the source mineral must be considered.

To test the mechanical contamination of radioactivity during the polishing, a thin section of placer monazite was used. The high angle alpha tracks alone are registered in the photo plate, if a silver foil of about 14 microns in thickness is inserted between the thin section of mineral and the photo plate. Thus the alpha track pattern precisely coinsides with the outline of monazite. In this case the radioactivity ejected from the gypsum around the monazite was as feeble as only 4.5 percent of that which comes from the monazite itself. A simmilar result was obtained concerning the thin section of urano-thorite from Naegi, its ratio was 5.4 percent without silver foil. In any way about 5 percent of radioactivity is seen

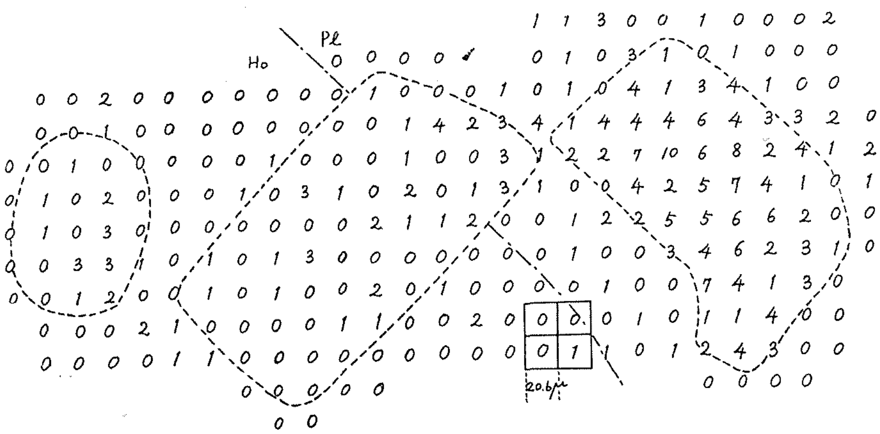


The enclosed numbers mean the track number ejected from outside of the grain per 55 microns square.
The "mrm" in the figure shows sphene area.

Fig. 4. Remeasurement of alpha tracks along the X-Y line in Figure 3.

around the radioactive mineral. But around the sphene, (see Fig. 3), fairly many alpha tracks are seen. To be more exact, the alpha track number was remeasured along the X–Y line with special reference to the direction and the dip angle of the tracks. The enclosed number in Fig. 4 indicates the alpha track number originated from the non-radioactive mineral around the sphene, and these tracks are usually high angled ones. The right-hand side grain corresponds to fresh zonal plagioclase, and radioactive contamination through the mechanical polishing is improbable.

Fig. 5 shows the alpha track distribution of 3 grains of sphene of the Kurama quartz diorite. In radioactivity each grain differs from another: the right-hand



dotted line···shape of sphene
Pl···sericitized plagioclase
Ho···hornblende
The number shows the alpha track number in 20.6 microns square after the 311 days exposure.

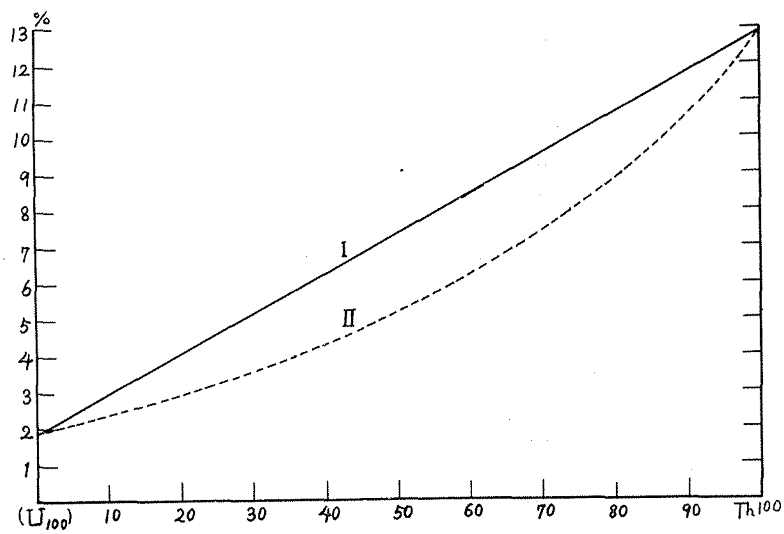
Fig. 5. Alpha track distribution of three sphene grains.

side one was included in the sericitized plagioclase and is the highest, and the left-hand side one was contained in green hornblende and is the feeblest (see Table 2, 2nd and 12th samples). It must be noticed that the plagioclase is higher in radioactivity than the hornblende shown in Fig. 5. The middle one is partly contacted with sericitized plagioclase and partly surrounded with hornblende. The activity of the third grain is fairly high on the plagioclase side, but low at the hornblende side (see Table 2, 11th sample). It is undeniable, therefore. that the radioactivity of such a grain ever stands in the closest relation with the quality of the rock-forming mineral surrounding or adjacent to it.

Th-U ratio of granitic sphene

The alpha permeability of sphene is 5.42.[10] According to Yagoda, the number of alpha particles escaping per second from 1 cm^2 of a uranium mineral amounts to $Tu=25.73\ U\Psi$. Here U is the uranium content in 1 gram of mineral, and Ψ is the alpha permeability of that mineral. If the above said three values (15×10^{-5} g/g[3], 8.3×10^{-5} g/g[4], 4.8×10^{-5} g/g[4], uranium equivalent) are applied to this formula, "*Tu*" must be $T_\alpha \doteqdot 0.0209$, 0.0116 and 0.0066 respectively. These values, which are of course irrespective of the thorium content in sphene, are given here for their comparison with the author's values shown in Table 1 and 2. It is generally believed that thorium is the sole or main radioactive content in sphene[2].

Thorium and uranium series differ in their range of alpha particles, and the longer alpha tracks are ejected from thorium. The number of long range alpha tracks per total alpha track number shows the *Th-U* ratio. According to the present author's experiments, the ratio of the longer track number over 20 microns to the total track number on the nuclear emulsion of 15 microns in thickness is the *Th-U* ratio as given in Fig. 6.



solid line I···*Th-U* activity ratio
dotted curve II···*Th-U* Weight ratio

Ordinate: Number of longer tracks over 20 microns/Number of total alpha tracks
Abscissa: *Th-U* ratio

Fig. 6.

In VON BUTTLAR and HOUTERMANS' terminology, J_u is the number of alpha tracks longer than a certain length ejected solely from the uranium (both 235 and 238) content in unit time and area, while that of thorium is represented with J_{Th}. The J_u/J_{Th} ratio, they say, is 2.40, if that length is over 2.31 cm; but 1.88, if it is 3.50 cm. Track length over 20 microns, as is the case with the author's experiments, correspond to 2.55 cm in the air range. Consequently the J_u/J_{Th} is 2.29, if the length be over 2.55 cm in the air.

In Fig. 6 the *Th-U* ratio of their radioactivity is expressed with the straight line I, and the weight ratio of *Th-U* with the dotted curve II.

Now the alpha tracks over 20 microns occupy 5.34 percent of the total track number in a grain (Kurama 8 or the 3rd crystal in Table 2) whose T_α was 0.0235; and from this ratio (5.34 percent) we know that the *Th/U* is 1.08. The *Th-U* ratio was 0.92 in another grain (the 8th crystal in Table 2) whose T_α was 0.0091. This result contradicts the general belief that thorium was the sole or principal radioactive source in sphene.

Summary and conclusion

1. The radioactivity of sphene was measured on some granitic rocks of Japan and Korea.
2. The radioactive variation of sphene was conspicuous even in the same thin section of rock.
3. Nearly the same amount of radioactive materials was probably deposited in each of such grains, densely concentrated in smaller crystals, but dilutely or heterogeneously scattered in larger ones.
4. The radioactivity of sphene greatly depends upon its color, and upon the quality of its neighboring mineral.
5. Hence the feeble radioactivity around the sphene in the granite must not be neglected.
6. The content ratio of thorium and uranium in sphene are nearly the same.

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