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The Radioactivity of Rocks and Minerals Studied with Nuclear Emulsion III

Radioactivity of Biotite of the Tanakamiyama Granite, Shiga Pref., Japan.

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Abstract

Tanakamiyama granite was studied by means of nuclear emulsion to determine the order of radioactivity contained in its biotite constituent, which was found strong in the biotite belonging to the later stage or having suffered a deuteric action, and consequently in its finer flakes situated interstitially between quartz and feldspar. The biotite itself is rather feebler in radioactivity when some minute radioactive inclusions are existent therein than when they are absent.

Introduction

What has been noticed concerning the distribution of radioactive elements in granite is that they are concentrated mainly in its minute accessory minerals. But little investigation has been made as to their distribution in its essential minerals. Hurley[(1)], for instance, merely says of pulverized granite that its radioactive elements are clustered chiefly upon the surface of the grain. As a rule, among the essential minerals, quartz and feldspar are less radioactive than biotite.[(2)] But in this case, the radioactivity in question means not only the radioac ive elements implied exclusively in quartz and feldspar themselves, but also those involved in the very minute accessory minerals which the quartz and feldspar contain.

Now, the author's autoradiographic detailed studies were applied to the thin sections of granite to ascertain the radioactivity of biotite itself. And if the author's experiment should be exposed to any uncertainty, it is due firstly to the existence of ultra-microscopic inclusions, and secondly to the contamination of the fragments of minute accessory minerals and biotite flakes that might occurred in the process of polishing the thin section. Since such contamination can be avoided by washing the thin section, the existence of those inclusions, as his experiment shows, is negligibly small. Now what was true of zircons(3) is also true of biotites : minerals of the same sort, but of divergent radioactivity, are often contained in one thin section of rock. The degree of the radioactivity stands in close relation with the stage of biotite crystalization : the later the stage of biotite is, the richer it is in radioactive contents.

Method

The autoradiographic method, indeed, characterizes itself by the fact that it renders the micro-distribution of radioactive elements detectable, however feeble their radioactivity may be ; but for the determination of an activity as feeble as biotite proper furnishes, a special care must be taken for exposure : firstly, a countable track number cannot be availed before three months' exposure.* To enable such long exposure, the photo plate must be kept cool or in dried nitrogen gas, lest any fading should occur. Secondly, the thin section must be as smooth as possible that it may be kept tightly together with the emulsion during the exposure. For in favourable circumstances the original mineral in the thin section is exactly knowable, under a microscope, from which the alpha tracks were coming, because the minute radioactive mineral, here in this case lying inside or near the biotite, projects the tracks in a radial form. If, on the contrary, there is a smallest room between the thin section and the emulsion, the area of the track distribution can not be localized with a great precision even under a microscope : some fractuation is unavoidable between the area of the tracks scattered upon the photo plate and the position of biotite in the thin section which is projecting the tracks upon the emulsion. The track number upon the emulsion is the largest in the center of their radiation and becomes smaller as it gets remoter from the center. The author's experiment shows that in the distance of one hundred microns it is reduced into a negligible value of less than one per cent.

To be strict, however, the pale brownish earthy materials contained on biotite and emitting alpha tracks, must be taken into consideration, as shown in the next paragraph. During the microscopic counting of alpha tracks, it also must be remembered that biotite, because of its large size, projects its tracks rather wider upon the emulsion than the minute accessory minerals do.

The radioactivity of biotite

The samples treated here were chiefly the granite containing coarse grained biotite and obtained at Shishitobi, Shiga Prefecture, relatively near the contact margin of the Tanakamiyama granite. The biotite involved in that granite

* Probably more than six months' exposure is required for the granite that contains less than three grammes uranium per ton.

shows the following varieties:

1) Green biotite with dark brown pleochroism and scattered between crystals of quartz or of feldspar; author's microscopic observation and alpha track counting ascribed both its blackness and pleochroic halo to a similar origin. Generally the fine grains of this biotite are found, together with albitic or potash feldspar, interstitially between quartz or feldspar crystals (Fig. 1); in the same thin section of rock its radioactive order varies, according as the position it occupies there differs, from $T\alpha = 0.03$ to 0.11; inside this biotite no radioactive inclusion is found; nor does the track distribution on the autoradiographic plate suggest any such inclusion. (Table 1)

Table 1 Radioactivity of dark green biotite

Sample	$T\alpha$	Size ($0.01 mm^2$)	U equiv. (%)	
668	0.110	1.61	0.082	Stripe
	0 097	3.78	0.072	between two Qu.
896 (2)	0.080	0.36	0.060	〃
	0.070	0.70	0.052	〃
896 (3)	0.062	4.46	0.046	Stripe
896 (2)	0.059	3.75	0.044	between two Qu.
658	0.058	3.60	0.043	
896 (2)	0.053	2.69	0.040	between two Qu.
895 (2)	0.051	1.08	0.039	Stripe
896 (1)	0.050	3.36	0.037	〃
	0.050	0.56	0.037	〃
770 (1)	0.048	1.08	0.036	〃
668	0.045	18.5	0.034	
895 (2)	0.044	0.22	0.033	Stripe
668	0.038	1.08	0.028	between two Qu.
896 (1)	0.028	6.90	0.021	Stripe
846 (1)	0.028	0.63	0.021	
	0.028	1.08	0.021	
	0.028	1.92	0.021	with inclusions

In a thin section of rock oriented in the direction perpendicular to the 001 plane, this sort of biotite furnishes pleochroism of pale green and dark green. Radioactivity stands high at such especially dark colored parts. Like the pleochroic haloes often found in biotite, this intensely dark pleochroism has been blackened by the alpha particles; but in case of these parts, radioactive elements are scattered much wider in the biotite, and the alpha effect is much wider than in case of haloes.

2) Green biotite in parallel intergrowth with muscovite. (Fig. 2) This laminar structure has been derived from brown or green biotite which has undergone a deuteric action, and which is always found in contact with

albitic or potash feldspar. The white stripe in this structure lies in the same optical orientation with the dark green and is linked up with muscovite in potash feldspar. The occasionally existent pleochroic haloes (Fig. 6) and the extraordinary smallness of 2V identify the white stripes with phengite rich in Fe". In radioactivity, however, such muscovite is as feeble as less

Table 2 Radioactivity of greenish brown biotite with smoky stain

Sample	$T\alpha$	Size (0.01 mm^2)	U equiv. (%)	
770 (1)	0.023	0.84	0.017	
896 (2)	0.019	23.1	0.014	
668	0.019	2.5	0.014	
770 (1)	0.014	2.0	0.010	
	0.012	1.8	0.009	with inclusions
770 (1)	0.058	0.47	0.043	Black stain only
896 (1)	0.040	3.61	0.030	Brown stain only
896 (3)	0.039	2.61	0.029	〃
	0.036	3.32	0.027	〃
668	0.031	3.15	0.023	〃
895 (4)	0.029	1.90	0.022	〃
896 (1)	0.020	7.60	0.015	〃

than $T\alpha=0.0005$ (Table 4) (Fig. 6), and the dark brown stain amounts to $T\alpha=0.020$—0.058 (Table 2), while the dusty part and the brownish stained portion are the strongest in this sort of green biotite. Inclusions of radioactive minute minerals are extremely rare and, if there are any, very feebly radioactive is the biotite itself.

3) Brown biotite. (Fig. 3) This is the commonest in Tanakamiyama granite and often furnished with myrmekite structure on its fringe. Minute radioactive minerals, together with remarkably developed pleochroic haloes, are concentrated especially in its brownish parts, while the biotite itself is feebly radioactive: $T\alpha=0.001$—0.007. (Table 3) Hence brown biotite containing minute radioactive minerals is naturally higher in activity than that which does not contain them. Owing to its comparatively large size, however, the radioactivity is not uniform in all its parts. In its protrusion into potash or albitic feldspar $T\alpha=0.010$—0.012. (Table 3) (Fig. 4)

4) Green biotite. This exists either as a myrmekite structure on the fringe of brown biotite or as a detached pale greenish biotite lying side by side with the former. In radioactivity, however, if pale, not dusty in color, it is as feeble as $T\alpha=0.0006$—0.002, but if slightly smoky then nearly $T\alpha=0.007$. (Table 4) And this stain has been produced by the radioactive elements which a deuteric action brought into the green biotite.

Table 3 Radioactivity of greenish brown biotite

Sample	Tα	Size (0.01mm²)	U equiv (%)	
668	0.0080	3.24	0.0060	
	(0.0127	2.16	0.0095)	*
	0.0080	22.0	0.0060	
	0.0079	3.46	0.0059	
	(0.0100	3.60	0.0075)	*
	(0.0116	5.41	0.0087)	*
846 (3)	0.0072	23.0	0.0054	
668	0.0070	5.75	0.0052	
896 (1)	0.0070	12.6	0.0052	with inclusions
843 (3)	0.0062	10.8	0.0046	
846 (1)	0.0062	7.0	0.0046	
	0.0062	2.16	0.0046	
846 (1)	0.0058	33.8	0.0043	
895 (4)	0.0054	72.0	0.0040	with inclusions
846 (3)	0.0050	15.6	0.0037	
	0.0034	12.6	0.0025	with inclusions
896 (2)	0.0030	16.6	0.0022	〃
846 (1)	0.0026	7.9	0.0019	
895 (2)	0.0025	23.0	0.0019	fresh biotite
896 (2)	0.0016	large	0.0012	with inclusions
895 (2)	0.0015	32.2	0.0011	
770 (2)	0.0012	15.3	0.0009	
846 (2)	0.0011	45.0	0.0008	
770 (1)	0.0010	16.7	0.0007	with inclusions

* Biotite protrusion part into orthoclase

Table 4 Radioactivity of pale green biotite and muscovite

Sample	Tα	Size (0.01mm²)	U equiv. (%)	
846 (2)	Under 0.00032	51.0	Under 0.0002	* Anhedral
	0.000 3	22.4	0.0004	* 〃
770 (1)	0.0070	15.7	0.0052	Dusty
895 (4)	0.0056	35.0	0.0042	Anhedral
770 (2)	0.0030	13.4	0.0022	Peripheric part of Brown biotite
846 (2)	0.0025	23.0	0.0019	Anhedral
	0.0016	19.0	0.0012	〃
895 (4)	0.0012	10.4	0.0009	
895 (2)	0.0012	20.3	0.0009	
	0.0007	17.9	0.0005	with inclusions
	0 0006	19.0	0.0004	Anhedral
895 (4)	0.0006	64.0	0.0004	

* Muscovites

The heterogeneity of radioactivity distribution in biotite

It is natural that, if the inclusions of minute radioactive minerals are clearly perceivable in biotite, the radioactivity is heterogeneously distributed in it. What, then, is the activity of the biotite itself (if that of those inclusions is excluded)? In transparent and fresh biotite, whether brown or green or pale green in color, its radioactivity is homogeneously distributed in it. If heterogeneously, on the contrary, some dusty stains brought about by a deuteric action are always perceivable therein.

The laminar intergrowth of biotite with muscovite is found always in conjunction with potash or albitic feldspar, and the brown parts still remaining in quartz often suggest that this laminar intergrowth has been transformed from fresh brown biotite. The dark green laminae (Fig. 5,6) are heterogeneously radioactive; and the more radioactive the biotite is, the more remarkable this heterogeneity appears; and in such a biotite as shown in Fig. 6 this tendency is the most obvious. Besides, here in this sample, heterogeneous distribution of radioactivity is seen, not only among similar pieces of biotite, but also on the two sides of the same biotite piece. Moreover, the radioactivity is concentrated especially on its sharp edges. Biotite kept free from the immersion of residual solution, probably because of its being included either in quartz or in oligoclase, remains fresh, brown and homogeneous in activity. Biotite alone causes majority of its radioactivity, and the heterogeneous distribution of radioactive elements in it is also due to the uranium deposition which a residual solusion offered to it after the biotite had been formed.

Some petrological observations

It is characteristic of Tanakamiyama granite that minute radioactive minerals have produced many remarkable haloes mainly inside brown biotite. As given in Fig. 3, sometimes the minute mineral may be monazite. Having been influenced by an albitic or potash feldspar solution, the laminar intergrowth of phengite muscovite with dark green biotite has been made from brown biotite. Changeable into a laminar intergrowth were, firstly, the boundaries of brown biotite, secondary, pale green biotite of later formation than that of the brown biotite and, thirdly, as shown in Fig. 6, even the part most abundant in minute radioactive minerals, which is supposed to have once been the very core of the brown biotite. Before the transformation the brown biotite must have been just as it is demonstrated in Fig. 3. The formation of various sorts of biotite happened probably in the following order:

brown biotite→myrmekite green biotite→pale green biotite→Fe phengite
└→dark green biotite

The uranium deposition occurred either during or after the formation of the biotite of the last sort, and much later than the pegmatitic stage of radioactive element deposition. The heterogeneity of such an uranium deposition indicates that it is due to a hydrothermal solution.

Noteworthy are the facts that minute radioactive minerals, though rarely, are included even in green biotite, that those minute minerals are not homogeneous in activity, and that radioactive elements are deposited far more on the surface than in the core of green biotite(3). In short, green biotite has been twice and in two different stages deposited with radioactive elements; and in the hydrothermal stage, however, the deposition was solely on the surface of the biotite. The laminar intergrowth of phengite with dark green biotite is seen only near the place in which granite and Palaeozoic formation are in contact, and where the deuteric action was the most prominent.

Conclusion

The radioactivity of biotite obtained at Shishitobi, Tanakamiyama, is due to the above said two different depositions. As regards the radioactivity of biotite proper, that which contains minute radioactive inclusions stands generally rather lower than that which does not imply them; fresh and transparent biotite is feebler in activity than smoky one or that which has suffered some affection.

	Biotite of earlier stage	Biotite of later stage
1	Brown	Green——Pale green——Dark green ——White
2	Inclusions are many and highly radioactive. Radioactive element deposition is of pegmatitic stage. When first crystalized, almost homogeneous in activity.	Inclusions are rare and rather feebly radioactive. Radioactive element deposition is of hydrothermal stage. Elements crowded around the grain are heterogeneous in activity.
3	Mostly large sized. The biotite proper is feebly, but almost homogoneously, radioactive.	Generally small sized. If unaffected, the biotite proper is feebly radioactive. If partly affected, the radioactive heterogeneity is more remarkable.

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Plate VII

Explanation of Plate VII

Fig. 1. Dark green biotite between quartz crystals, $T\alpha = 0.028 \sim 0.11$.

Fig. 2. Dark greenish biotite stripes in the pale green biotite. Surrounding part: albitic plagioclase. Striped part $T\alpha = 0.02 \sim 0.58$.

Fig. 3. Brown biotite with peripheric pale green myrmekitic zone. B: Brown part $T\alpha = 0.001 \sim 0.008$; G: Green part $T\alpha = 0.0006 \sim 0.006$

Fig. 4. Dark green biotite after brown biotite. D: Dark green biotite; M: Muscovite; P: Potash feldspar; B: Brown biotite.

Fig. 5. Altered biotite by potash feldspar. G: Green biotite; M: Muscovite; P: Potash feldspar.

Fig. 6. Pleochroic haloes (ThC' halo) in Phengitic muscovite. D: Dark green biotite; F: Phengite; M: Monazite.

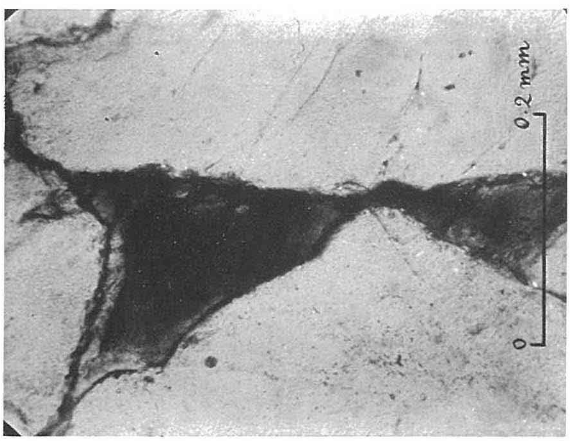

Fig. 1.

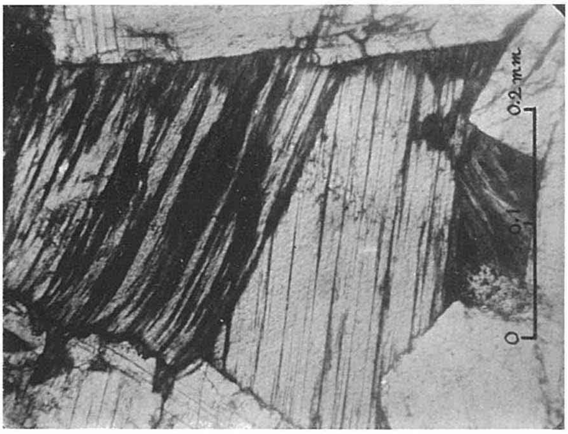

Fig. 2.

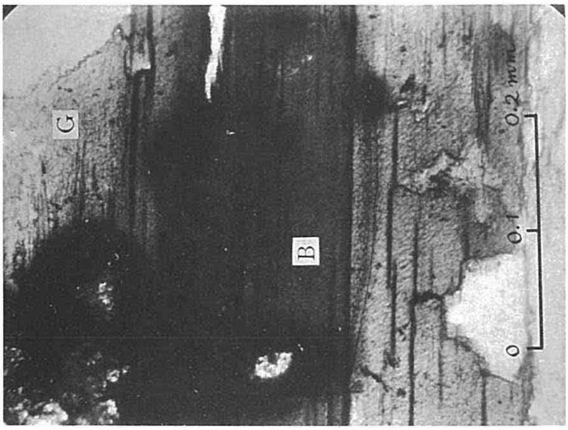

Fig. 3.

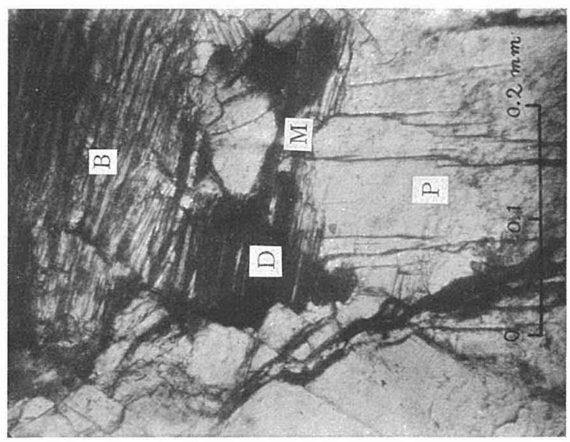

Fig. 4.

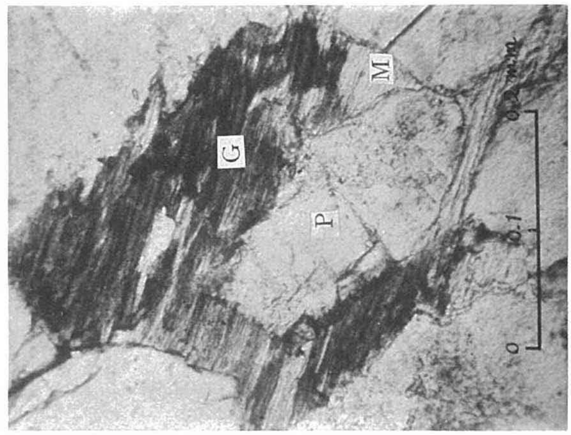

Fig. 5.

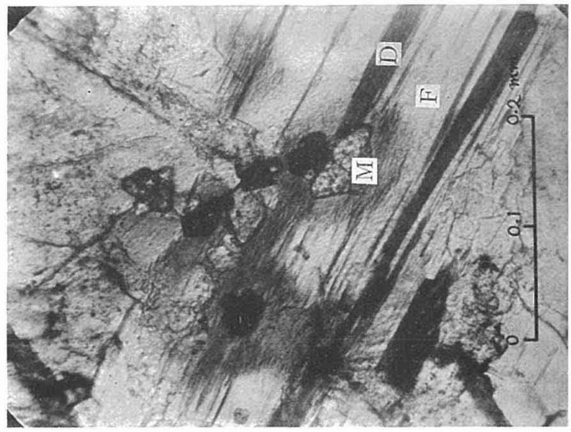

Fig. 6.