研究報文

TWO CONTRASTIVE TRENDS OF EVOLUTION OF
BIOTITE IN GRANITIC ROCKS

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INTRODUCTION

As is known well, the ratio of iron to magnesium in comagmatic granitic rocks usually tends to become higher as differentiation progresses. A similar relation is recognized also in cogenetic granitic rock series; namely, the more the rock is acidic, the higher becomes the ratio. The dominant ferromagnesian minerals in granitic rocks are usually biotite and hornblende, so the ratio of iron to magnesium of the rock is decided largely by the chemical composition of these minerals. According to the writer's investigation, however, the ratio of iron to magnesium of biotite and hornblende is not always proportional to that of rock. This implies that the chemistry of granitic rocks is connected not only with biotite and hornblende but also with other constituents such as magnetite, the formation of which could be facilitated under high oxygen partial pressure. In this respect, the trends of evolution of biotite must be important in clarifying the physico-chemical conditions during the evolution of granitic magma.

BIOTITES IN THE LATE MESOZOIC TO EARLY TERTIARY
GRANITIC ROCKS OF SOUTHWEST JAPAN

The late Mesozoic to early Tertiary granitic rocks are represented by the large batholithic intrusion from northern Kyushu to Chubu, which has an extension of about 500 km or more with a maximum width 80 km (Fig. 1). After Kawano and Ueda (1966, 1967) the granites are separated into two groups, one is 75 to 95 million years and the other is 50 to 65 million years,

Fig. 1. Map showing the distribution of late Mesozoic to early Tertiary granites and Palaeogene cauldrons in Southwest Japan.

according to the data of isotope dating. Regardless of the difference in age, the granites of the two groups have a close affinity in petrographic characters and field occurrences. They intrude the Palaeozoic to Mesozoic formations with discordant contact relations. As emphasized by Kojima (1954), these granites are assigned to a shallow facies from their texture, contact effect, properties of the constituent minerals and other features. Accordingly, it can be safely said that these granites bear resemblance to the so-called post-orogenic granites in the orogenic zones, although they differ from the latter in the mode of emplacement depending on the character of the tectonic movement. The main component rock types are biotite granite (adamellite) of comparatively uniform chemical composition (Murakami, 1960) to hornblende-biotite granodiorite with minor amount of quartz diorite to quartz gabbro. It is noted that these granitic rocks have no transitional relation to quartz porphyry nor to rhyolite, though they often become porphyritic or fine-grained at the margin. Eleven specimens of granites and one specimen of granodiorite were collected from various districts and chemically analysed, with the result as given in Table 1. The analysed granites are coarse- to medium-grained, occasionally accompanied by aplitic and pegmatitic veins. The main constituents are subhedral plagioclase of albitic to sodic andesinic composition (An 15–40), orthoclase to microcline (Or 69–95) with conspicuous perthitic texture, quartz and biotite. The orthoclase to microcline tends to be potassic in bulk composition with increasing optic axial angle and trilinearity (Murakami *et al.*, 1964). Commonly, it is present interstitially, often porphyroblastically. Quartz occurs

Table 1. Chemical compositions of the late Mesozoic to early Tertiary granitic rocks

No.	1	2	3	4	5	6	7	8	9	10	11	12
SiO ₂	63.54	77.31	74.33	73.74	72.06	72.85	75.62	76.26	74.61	74.45	75.34	71.41
TiO ₂	0.55	tr.	0.23	0.13	0.20	0.14	0.07	0.04	0.14	0.19	0.18	0.21
Al ₂ O ₃	15.64	12.28	13.53	13.97	13.66	14.70	12.41	12.45	12.66	12.34	12.64	14.79
Fe ₂ O ₃	1.01	0.50	0.51	0.39	0.43	0.61	0.58	0.69	0.21	0.37	0.68	0.72
FeO	4.18	0.43	1.10	1.77	1.68	1.18	1.70	0.62	1.38	1.73	1.61	1.95
MnO	0.11	0.02	0.04	0.04	0.11	0.08	0.06	0.05	0.06	0.02	0.05	0.07
MgO	1.88	0.04	0.17	0.16	0.72	0.20	0.14	0.06	0.27	0.13	0.17	0.43
CaO	4.48	1.19	0.95	1.41	2.22	2.03	1.33	1.14	1.89	1.38	1.72	2.97
Na ₂ O	4.66	3.75	4.18	3.98	4.08	3.59	4.26	3.91	3.95	4.18	4.04	3.77
K ₂ O	2.29	4.24	3.98	3.73	3.41	3.28	3.22	4.10	3.84	4.10	2.61	2.77
P ₂ O ₅	0.07	0.02	0.08	0.03	0.06	0.07	0.05	0.01	0.03	0.04	0.03	0.03
H ₂ O(+)	1.26	0.52	0.37	0.46	1.14	0.89	0.39	0.27	0.50	0.47	0.20	1.23
H ₂ O(-)	0.25	0.08	0.16	0.14	0.05	0.20	0.13	0.08	0.06	0.20	0.20	0.03
T.	99.92	100.38	99.63	99.95	99.82	99.82	99.96	99.68	99.60	99.60	99.47	100.38
SI	13.4	0.4	1.5	1.1	7.0	2.3	1.4	0.6	2.8	1.2	1.9	4.5
(FeO+Fe ₂ O ₃)/ (FeO+Fe ₂ O ₃ +MgO)	0.73	0.96	0.91	0.95	0.75	0.90	0.94	0.96	0.85	0.94	0.93	0.86

1 : Granodiorite. 2-12 : Biotite granite.

Analyst : N. Murakami

1 : Takamizu, Yamaguchi Pref.

2 : Iwaki Islet, Ehime Pref.

3 : Ennotani, Tottori Pref.

4 : Mannari, Okayama Pref.

5 : Masago, Shimane Pref.

6 : Narikawa Yamaguchi Pref.

7 : Kagawa, Yamaguchi Pref.

8 : Onojiri, Yamaguchi Pref.

9 : Shiratori, Kagawa Pref.

10 : Kure, Hiroshima Pref.

11 : Kake, Hiroshima Pref.

12 : Kuga, Yamaguchi Pref.

Table 2. Chemical compositions of the biotites in the

No.	1 B	2 B	3 B	4 B	5 B
SiO ₂	37.37	33.95	34.96	34.62	35.31
TiO ₂	2.46	2.10	5.28	3.48	4.04
Al ₂ O ₃	14.89	13.79	14.29	13.22	14.53
Fe ₂ O ₃	4.33	4.33	4.98	5.76	1.58
FeO	18.79	29.03	17.91	27.00	21.54
MnO	0.26	0.69	1.91	0.63	0.89
MgO	8.94	2.83	8.06	2.98	7.93
CaO	0.38	0.51	0.65	0.90	1.34
Na ₂ O	0.60	1.19	1.02	0.49	0.84
K ₂ O	8.64	6.86	6.82	7.55	7.54
H ₂ O (+)	2.91	4.00	3.94	3.37	3.72
H ₂ O (-)	0.09	0.20	0.40	0.05	0.60
Total	99.66	99.48	100.02	100.05	99.86
Si	5.788	5.475	5.360	5.561	5.488
Al ^{IV}	2.212	2.525	2.583	2.439	2.512
Al ^{VI}	0.507	0.096	—	0.065	0.156
Ti	0.287	0.255	0.609	0.420	0.473
Fe ⁺³	0.505	0.526	0.575	0.696	0.185
Fe ⁺²	2.434	3.915	2.296	3.627	2.806
Mg	2.064	0.680	1.842	0.713	1.841
Mn	0.034	0.094	0.248	0.086	0.117
Ca	0.063	0.088	0.107	0.155	0.224
Na	0.138	0.372	0.303	0.153	0.254
K	1.696	1.411	1.334	1.547	1.499
OH	3.007	4.303	4.029	3.611	3.866
Z	8.00	8.00	7.94	8.00	8.00
Y	5.83	5.67	5.57	5.71	5.57
Z	1.90	1.87	1.74	1.86	1.98
Fe ⁺³ /(Fe ⁺² +Fe ⁺³)	0.17	0.13	0.12	0.16	0.06
(FeO+Fe ₂ O ₃)/ (FeO+Fe ₂ O ₃ +MgO)	0.72	0.92	0.74	0.92	0.74
γ	1.656	1.667	1.660	1.666	1.661

Numbers (1-12) correspond to those in Table 1.

13B : Biotite granite, Iwakura, Yamaguchi Prefecture

* Includes P₂O₅ 0.23.

late Mesozoic to early Tertiary granitic rocks

6 B	7 B	8 B	9 B	10 B	11 B	12 B	13 B
38.03	37.85	33.33	35.96	37.00	34.63	33.71	33.68
1.85	1.97	2.84	2.54	2.31	2.58	3.06	2.28
15.28	16.14	13.26	14.41	13.75	13.84	15.85	17.18
1.74	2.15	6.34	4.05	5.40	5.27	4.81	2.42
22.46	21.69	27.83	24.07	23.57	25.24	20.92	25.60
0.96	1.31	0.92	0.82	0.67	0.80	0.78	1.66
5.22	4.89	1.92	4.49	4.28	4.27	7.59	4.46
0.39	0.48	0.76	0.91	0.95	1.00	1.73	0.80
1.04	1.33	1.09	0.59	0.76	0.37	0.70	0.48
8.24	7.97	7.32	7.79	7.55	7.58	6.50	7.37
3.87	3.64	3.98	3.85	3.33	3.56	4.12	3.50
0.33	0.12	0.18	0.27	0.26	0.57	0.12	0.15
99.41	99.54	99.77	99.75	99.83	99.71	99.89	99.81*
5.897	5.853	5.395	5.655	5.836	5.546	5.207	5.351
2.103	2.147	2.530	2.345	2.164	2.454	2.793	2.649
0.689	0.795	—	0.326	0.382	0.159	0.094	0.569
0.216	0.229	0.346	0.300	0.274	0.311	0.356	0.272
0.203	0.250	0.772	0.479	0.641	0.635	0.559	0.289
2.912	2.805	3.767	3.166	3.099	3.381	2.703	3.401
1.206	1.127	0.463	1.052	1.066	1.019	1.747	1.056
0.126	0.172	0.126	0.109	0.089	0.019	0.102	0.223
0.065	0.080	0.132	0.153	0.163	0.172	0.286	0.136
0.313	0.399	0.342	0.165	0.231	0.115	0.210	0.148
1.630	1.573	1.512	1.563	1.519	1.549	1.281	1.494
4.003	3.755	4.297	4.039	3.503	3.803	4.245	3.709
8.00	8.00	7.97	8.00	8.00	8.00	8.00	8.00
5.35	5.38	5.47	5.43	5.49	5.61	5.56	5.81
2.01	2.05	2.00	1.88	1.91	1.84	1.78	1.78
0.07	0.08	0.17	0.13	0.17	0.16	0.17	0.08
0.82	0.83	0.95	0.86	0.87	0.88	0.77	0.86
1.661	1.662	1.668	1.664	1.663	1.663	1.660	—

Analyst : N. Murakami

(Tsuboi *et al.*, 1935)

usually as interstitial crystals, rarely porphyroblastically. The minor accessories are magnetite, allanite, epidote, apatite and zircon. The granodiorite is medium-grained and consists of oligoclasic to labradoritic plagioclase (An 23–57), orthoclase (Or 85), quartz, green hornblende and biotite, with subordinate magnetite, titanite, epidote, apatite and zircon.

Description of biotite

Biotite in the analysed granites occurs commonly as large subhedral crystals. Fine flakes included in the felsic minerals are also found. Small aggregates of this mineral arranged randomly are rarely detected in the granites, despite of its evenly distributed appearance. It often suffers chloritization along cleavage and periphery. Minute inclusions of magnetite, zircon and apatite are scattered. By volumetry the amount of biotite is estimated at 0.1 to 1.0 per cent. The refractive index (γ) is characteristically high, usually 1.660 to 1.664, attaining to 1.668. It is strongly pleochroic: X=light yellow, Y=Z=deep brown to dark brown, often with a greenish or reddish tinge.

Biotite in the granodiorite occurs as isolated subhedral crystals, or as intergrowth with amphibole, frequently replacing the latter. $\gamma=1.654$, X=light yellow, Y=Z=brown.

The above-cited biotites were taken out of powdered rock specimens by

Fig. 2. Relations between biotites and rocks in the $(\text{FeO} + \text{Fe}_2\text{O}_3) / (\text{FeO} + \text{Fe}_2\text{O}_3 + \text{MgO})$ ratio and SI of the late Mesozoic to early Tertiary granites. The data on the Ichinohe alkali plutonic rocks (Onuki and Tiba, 1964) are also plotted.

means of magnetic separator and heavy liquid. Their analytical values are listed in Table 2.

Mutual relations in chemistry between biotites and rocks

Fig. 2 shows the relation between the solidification index (SI) (Kuno, 1954) of rocks and the $(\text{FeO} + \text{Fe}_2\text{O}_3)/(\text{FeO} + \text{Fe}_2\text{O}_3 + \text{MgO})$ ratio of rocks and biotites. From this figure, the followings are inferred:

- a) The rocks and the biotites have similar values of the ratio.
- b) The values are more or less lower in the biotites.
- c) With decreasing SI, both rocks and biotites become gradually iron-rich and magnesium-poor.

These mean that a large percentage of the iron content of the granites is contained in the biotites, and only a small amount in other constituents such as magnetite. Modal analyses reveal that the iron ore content does not exceed 1.5 per cent in these granites. A small discrepancy in the ratio is observed between rocks and biotites from SI 1 to 5. In these granites the crystallization of magnetite must be slightly advanced.

BIOTITES IN THE PALAEOGENE TAMAGAWA GRANITES AND RELATED PLUTONIC ROCKS

The Tamagawa granites occur in the large cauldron area, which occupies an area of about 80 square kilometers in the northeastern margin of Yamaguchi Prefecture, Southwest Japan. This cauldron named Tamagawa Cauldron by Murakami (1968) is presumed to be Oligocene or Eocene in age from isotope data and geological evidences (Murakami and Hase, 1967). It is represented by huge downfaulted blocks of volcanics and associated plutonic intrusives, bounded by a ring-shaped shear zone with a width of about 20–300 m (Fig. 3). The volcanics within the cauldron are andesite lava, dacite tuff and tuff breccia (partly welded), rhyodacite to rhyolite tuff breccia, and andesite dikes in the order of extrusion and intrusion. They belong to the calc-alkaline rock series (Fig. 4). The cauldron is considered to have been formed by collapse resulting from the withdrawal of magma just after the eruption of dacitic pyroclastics. The total volume of the collapsed volcanics is estimated at 50 to 120 km³ or more. The plutonics intrude the dacite tuff and tuff breccia and are invaded by the rhyodacite tuff breccia which is regarded as the derivatives from residual granite magma. They are divided into two groups, the central granitic rocks and the marginal basic complexes, although besides the main mass, the granites belonging to the former group crops out as small stock-like masses in close association with the latter group in the northeastern area outside the cauldron. The central granitic rocks are composed of biotite

Fig. 3. Geological map of the Tamagawa Cauldron area.

Fig. 4. $\text{MaO}-(\text{FeO}+\text{Fe}_2\text{O}_3)-(\text{Na}_2\text{O}+\text{K}_2\text{O})$ triangular diagram of the Tamagawa plutonic rocks (solid circle) and related volcanic rocks (open circle).

granite with minor amounts of hornblende-biotite granite, two-mica granite, aplite and granite porphyry. The pegmatitic rock is nearly absent.

Both biotite granite and hornblende-biotite granite are characterized by the conspicuous semiporphyritic texture with ovoid plagioclase phenocrysts. Other constituents are alkali feldspar, quartz, biotite, hornblende and magnetite, with subordinate quantities of titanite, allanite, epidote, tourmaline, apatite, zircon and others. Plagioclase occurs as subangular to rounded phenocrystic crystals ranging from An 16 to 39 in composition. In some aplites, usually equigranular, this mineral is almost lacking. It commonly suffers alkali feldspathization along periphery and cleavage, and perfectly mantled crystals are often observable (Fig. 5). Alkali feldspar and quartz constitute the groundmass mainly as interstitial fillings. They frequently show granophyric intergrowth, occurring as phenocryst in a few cases. Of these, the alkali feldspar has about 49 to 60 Or composition and replaces plagioclase as described before. It is marked by a moiré appearance with abundant dusty inclusions. In some granites, however, the overgrowth composed of clear potassic core and turbid sodic shell is developed (Fig. 6). Hornblende is not of common occurrence. Where present, it occurs as discrete subhedral crystals with biotite. $a=1.650$, $\beta=1.664$, $\gamma=1.673$. $2Vx=80-81^\circ$. X=pale yellow, Y=pale yellowish green, Z=yellowish green. Occasionally pale green fibrous amphibole ($2Vx=72^\circ$), probably actinolite, replaces this mineral. As given in

Fig. 5. Sketch showing the plagioclase mantled by alkali feldspar (antirapakivi). Pl: plagioclase. Kf: Alkali feldspar. Q: Quartz.

Fig. 6. Outgrowth of alkali feldspar. The core is relatively clear and potassic ($2Vx=47-55^\circ$, $\gamma=1.525-1.526$), whereas the shell is turbid and sodic ($2Vx=66^\circ$, $\gamma=1.526-1.528$). Pl: Plagioclase. Kf: Alkali feldspar. Q=Quartz. Bi: Biotite. Mt. Magnetite.

Fig. 7. Sketch showing the occurrence of magnetites. (I). They are largely of discrete euhedral to rounded grains. Pl: Plagioclase. Kf: Alkali feldspar. Q: Quartz. Bi: Biotite. Mt: Magnetite. Ap: Apatite. Zr: Zircon.

Fig. 8. Sketch showing the occurrence of magnetites. (II). They occur as interstitial fillings. The mantling of plagioclase by alkali feldspar is remarkable around magnetite. Fine flakes of magnesium-rich biotite are accompanied.

Table 5, high magnesium and low aluminium content is a characteristic feature of this mineral. In comparison with the late Mesozoic to early Tertiary granites, the Tamagawa granites have a larger amount of magnetite amounting 1 to 4 per cent by volume. Two modes of occurrence of magnetite are contrastive. One is euhedral to rounded minute grains arranged along crystal boundaries or enclosed in other crystals (Fig. 7) and the other is interstitial fillings with poikilitically included minerals such as plagioclase and quartz (Fig. 8). Besides those there are also fine grained replacement products after biotites along cleavage and periphery (Fig. 9). It is noted that the highly magnesium-rich biotites are intimately associated with these interstitial magnetites and mantled plagioclases. Hydrothermal alteration products such as chlorite, epidote, sericite, carbonate

Fig. 9. Sketch showing the occurrence of magnetites. (III). They occur as fine replacement products along cleavage and periphery of the biotite.

Table 3. Chemical compositions of the

No.	1	2	3	4	5	6
SiO ₂	49.22	52.36	56.09	56.96	59.08	60.97
TiO ₂	0.76	0.62	0.86	0.92	0.30	0.65
Al ₂ O ₃	17.52	18.60	16.60	16.76	16.09	16.64
Fe ₂ O ₃	2.99	1.27	2.84	1.93	3.30	2.28
FeO	6.47	5.86	3.41	4.77	4.11	4.27
MnO	0.19	0.14	0.14	0.12	0.13	0.14
MgO	7.55	5.35	4.39	4.72	3.84	2.66
CaO	9.97	9.82	6.82	6.97	6.79	5.40
Na ₂ O	2.54	3.58	4.56	3.89	4.09	3.64
K ₂ O	0.22	0.84	1.23	1.20	1.18	1.14
P ₂ O ₅	0.08	0.25	0.19	0.17	0.24	0.13
H ₂ O(+)	2.03	1.16	2.31	1.44	0.67	1.44
H ₂ O(-)	0.05	0.17	0.13	0.09	0.00	0.10
T.	99.59	100.02	99.57	99.94	99.82	99.46
SI	38.2	31.7	28.5	28.6	23.2	19.0
(FeO+Fe ₂ O ₃)/ (FeO+Fe ₂ O ₃ +MgO)	0.56	0.57	0.59	0.59	0.66	0.71
No.	15	16	17	18	19	20
SiO ₂	71.96	72.12	72.47	72.48	73.08	73.15
TiO ₂	0.21	0.25	0.31	0.18	0.23	0.26
Al ₂ O ₃	13.90	13.56	13.89	13.86	13.74	13.59
Fe ₂ O ₃	0.94	0.94	2.56	1.23	1.09	1.55
FeO	1.36	1.01	1.21	0.92	0.83	0.55
MnO	0.02	0.04	0.04	0.05	0.05	0.06
MgO	0.48	0.32	0.33	0.43	0.33	0.35
CaO	1.62	2.39	1.84	1.58	2.34	1.87
Na ₂ O	4.95	4.50	4.65	4.58	5.04	4.45
K ₂ O	3.79	2.87	2.45	3.73	2.20	2.77
P ₂ O ₅	0.05	0.04	0.04	0.08	0.02	0.06
H ₂ O(+)	1.17	1.01	0.51	0.41	0.49	0.69
H ₂ O(-)	0.28	0.42	0.05	0.07	0.11	0.19
T.	100.73	99.47	100.35	99.60	99.55	99.54
SI	4.2	3.3	2.9	3.9	3.5	3.6
(FeO+Fe ₂ O ₃)/ (FeO+Fe ₂ O ₃ +MgO)	0.83	0.86	0.92	0.83	0.85	0.85

1 : Augite gabbro. 2 : Biotite-hornblende-two pyroxene gabbro.
mica granite. 15-19 : Biotite granite. 20 : Hornblende-biotite
granite. 23 : Aplite. 24 : Granite porphyry. 25-26 : Aplite.
1-9, 11-13, 23, 26 : Marginal plutonic rocks. 10, 14-22, 24-25,
(Kumogi Cauldron of the same age as Tamagawa Cauldron).

Tamagawa plutonic rocks

7	8	9	10	11	12	13	14
61.31	64.84	65.25	65.78	66.67	67.17	67.79	70.97
0.90	0.23	0.44	0.80	0.44	0.52	0.37	0.38
15.35	15.54	14.88	14.98	15.09	14.98	15.28	13.23
1.79	0.24	2.74	2.08	0.88	0.86	0.86	0.89
4.04	3.94	2.54	2.33	2.73	2.86	2.84	0.84
0.09	0.12	0.22	0.10	0.06	0.13	0.07	0.07
3.15	1.34	1.20	2.44	1.54	1.72	1.47	0.88
5.67	5.58	4.03	2.71	3.61	3.97	3.31	2.06
3.63	4.20	5.29	4.98	4.51	3.44	3.78	4.11
1.63	1.68	2.11	1.89	2.35	2.95	2.71	3.61
0.19	0.12	0.15	0.15	0.14	0.09	0.23	0.03
2.04	2.12	0.68	1.36	1.57	1.31	0.83	2.78
0.15	0.07	0.07	0.04	0.09	0.20	0.10	0.38
99.94	100.02	99.60	99.64	99.68	100.20	99.64	100.23
22.1	11.8	8.6	17.8	12.8	14.5	12.6	8.5
0.65	0.75	0.81	0.64	0.70	0.68	0.72	0.66
21	22	23	24	25	26	27	28
73.72	73.89	74.59	76.02	76.14	76.17	76.52	74.96
0.19	0.15	0.08	0.03	0.16	0.07	0.12	0.10
13.30	13.29	12.60	12.34	12.81	11.56	13.03	13.29
0.46	1.10	0.17	1.05	0.66	1.80	0.64	1.15
0.92	0.37	0.79	0.31	0.30	0.39	0.45	0.92
0.03	0.00	0.01	0.03	0.01	0.01	0.01	0.06
0.80	0.13	0.37	0.57	0.08	0.09	0.07	0.39
1.47	0.74	1.35	0.91	0.80	0.66	0.90	1.60
4.29	3.91	4.85	4.70	3.89	3.57	3.64	4.32
2.35	4.70	3.73	3.01	4.15	4.52	3.73	2.90
0.03	0.06	0.04	0.04	0.01	0.01	0.00	0.10
2.07	1.27	0.92	1.23	0.42	0.56	0.56	0.59
0.29	0.28	0.14	0.09	0.00	0.05	0.14	0.06
99.92	99.89	99.64	100.33	99.43	99.46	99.81	100.44
9.1	1.3	8.0	5.9	0.9	0.9	0.8	4.0
0.63	0.92	0.72	0.70	0.92	0.96	0.94	0.84

Analyst : N. Murakami

3-7 : Quartz diorite. 8-13 : Granodiorite. 14 : Two-

granite. 21 : Albitized granite. 22 : Alkali feldspathized

27 : Granite porphyry. 28 : Biotite granite.

27 : Central granitic rocks. 28 : Kumogi, Shimane Pref.

and albite, are carried subordinately, though often abundantly.

The marginal plutonic complex crops out in the basement rocks close to the cauldron and along the marginal shear zone. It consists mainly of granodiorite and quartz diorite veined by aplite dikes, and comprises subordinate amounts of gabbroic rocks in which two types are distinguished, augite gabbro and biotite-hornblende-two pyroxene gabbro. They are present in contact with quartz diorite. The distribution of the marginal plutonic complex is characteristic. It occurs as discontinuous several tens of masses, which are arranged annual as a whole, surrounding the cauldron. The greater part of the component rocks exhibits semiporphyritic appearance owing to the presence of ovoid plagioclases similar to those in the granites, especially prominent in the granodiorite. Plagioclase (An 17–87), green hornblende and biotite essentially make up the phenocrystic minerals set in the groundmass consisting of plagioclase (An 20–56), alkali feldspar (Or 63–73), quartz, hornblende, biotite, magnetite and others. Of these, the alkali feldspar and quartz are in small quantities or absent in the quartz diorite and gabbroic rocks. Besides those there are augite ($\text{Ca}_{42}\text{Mg}_{40}\text{Fe}_{18}\text{—Ca}_{42}\text{Mg}_{31}\text{Fe}_{27}$) and orthopyroxene ($\text{Mg}_{60}\text{—Mg}_{67}$) in the gabbroic rocks and quartz diorite, rarely also in the granodiorite as relics enclosed by amphibole. Alteration is frequent, such as chloritization of amphibole and biotite and albitization of plagioclase. Some remarks are added to above descriptions.

a) Plagioclase occurs as subangular to rounded phenocrystic crystals, while in the groundmass it appears as subhedral to anhedral grains with alkali feldspar and quartz. Its composition varies with rock type, ranging from An 34 to 87 in gabbroic rocks, 23 to 68 in quartz diorite, 17 to 54 in granodiorite.

b) Alkali feldspar usually occurs interstitially, often intergrown with quartz. It is more potassic than that in the granites. Optic axial angle ($2V_x$) ranges from $48\text{--}54^\circ$ in clear crystals to $62\text{--}73^\circ$ in turbid ones, varying with rock type and rock specimen or even in one section. Some crystals have clear core with turbid shell.

c) Hornblende is present generally as discrete subhedral to anhedral crystals, often replacing pyroxene. Optical properities are briefly summed up below.

In biotite-hornblende-two pyroxene gabbro: $\alpha=1.652$, $\beta=1.665$, $\gamma=1.672$, $2V_x=75^\circ$, X=pale yellow, Y=yellowish green, Z=deep yellowish green.

In quartz diorite: $\alpha=1.657\text{--}1.660$, $\beta=1.670\text{--}1.673$, $\gamma=1.678\text{--}1.681$, $2V_x=78\text{--}82^\circ$, X=pale yellow, Y=yellowish green, Z=deep yellowish green to deep green.

In granodiorite: $\alpha=1.648\text{--}1.658$, $\beta=1.660\text{--}1.673$, $\gamma=1.667\text{--}1.680$, $2V_x=64\text{--}74^\circ$, X=pale yellow, Y=yellowish green to green, Z=deep green to deep

green with brownish tinge.

It must be noted that there is a noticeable difference in optic axial angle between hornblende in the quartz diorite and that in the granodiorite. Actinolitic amphibole with pale greenish pleochroism frequently replaces this mineral.

d) Augite is ferrous in the granodiorite ($\beta=1.698$, $2V_z=53^\circ$), while it is magnesian in the quartz diorite and gabbroic rocks ($\beta=1.692-1.696$, $2V_z=46-52^\circ$).

e) Orthopyroxene is rare except for gabbroic rocks. It consists of ferrous shell and magnesian core.

f) Magnetite occurs usually as isolated minute grains or granular aggregates. It is also included abundantly in the chloritized biotite and hornblende, and uralitized pyroxene. Interstitial fillings characteristic in the gabbroites are not detected.

Description of biotites

The biotite in the granites is present as discrete subhedral crystals, scaly aggregates and often fine flakes replaced by magnetite, while in the granodiorite to gabbroic rocks it is accompanied by intergrowing and overgrowing amphibole. Minute grains of magnetite are included, especially predominant in the biotite of the granites, and scattering along cleavage in most cases. The optical properties are summarized in the following.

Biotite in the granodiorite to gabbroic rocks: $\gamma=1.642-1.656$ (~ 1.663 in aplites), $X=\text{pale yellow}$, $Y=Z=\text{brown to reddish brown}$.

Biotite in the granite: $\gamma=1.617-1.634$, $X=\text{pale yellow}$, $Y=Z=\text{brown with reddish tinge to reddish brown}$.

In general, the lower the refractive index, the more reddish becomes the pleochroism of biotite.

Six biotites were separated together with three associated hornblendes, and were chemically analysed. The result is listed in Table 4 and 5. On the basis of this result, the refractive index of biotite is considered to be related mainly to the iron/magnesium ratio, whereas the ferric/ferrous ratio appears to lower the index.

Mutual relations in chemistry between biotites and rocks.

In comparison with the late Mesozoic to early Tertiary granites, the Tamagawa granites have a different tendency in the chemistry of biotites. The following two figures are indicative of this tendency. Fig. 10 illustrates the variation of γ -index of biotites with decreasing SI, and Fig. 11 shows the relations between biotites and rocks in the $(\text{FeO}+\text{Fe}_2\text{O}_3)/(\text{FeO}+\text{Fe}_2\text{O}_3+\text{MgO})$ ratio and SI. The data of hornblendes are also plotted in Fig. 11. From these figures, it is evident that the biotites as well as hornblendes become gradually

Table 4. Chemical compositions of the biotites in the Tamagawa plutonic rocks

No.	2 B	13 B	17 B	18 B	20 B	28 B
SiO ₂	36.12	36.31	36.84	37.60	36.30	37.54
TiO ₂	2.99	2.38	3.61	2.71	3.56	3.71
Al ₂ O ₃	15.43	13.67	15.45	13.76	13.37	13.93
Fe ₂ O ₃	3.00	1.33	2.24	1.48	9.84	4.23
FeO	16.22	23.64	11.14	16.58	7.25	12.31
MnO	0.20	0.64	0.23	0.39	0.08	0.87
MgO	13.78	10.24	16.76	12.51	16.83	12.35
CaO	0.36	0.36	0.56	0.36	0.44	1.65
Na ₂ O	0.59	0.67	1.23	1.83	0.50	0.86
K ₂ O	7.45	7.17	6.86	8.00	8.04	7.96
H ₂ O(+)	3.60	3.30	3.84	4.32	2.81	4.29
H ₂ O(-)	0.16	0.18	0.44	0.32	0.50	0.32
T.	99.90	99.89	99.70	99.86	99.52	100.02
Si	5.546	5.657	5.433	5.645	5.466	5.579
Al ^{IV}	2.454	2.343	2.567	2.355	2.373	2.421
Al ^{VI}	0.159	0.167	0.119	0.080	—	0.019
Ti	0.811	0.279	0.400	0.306	0.403	0.415
Fe ⁺³	0.635	0.156	0.249	0.167	1.115	0.473
Fe ⁺²	3.381	3.080	1.374	2.082	0.913	1.530
Mg	1.019	0.084	3.684	0.062	0.010	2.735
Mn	0.109	2.377	0.029	2.799	3.777	0.110
Ca	0.172	0.060	0.088	0.058	0.071	0.263
Na	0.115	0.202	0.352	0.533	0.146	0.248
K	1.549	1.425	1.385	1.532	1.545	1.509
OH	3.803	3.429	3.778	4.326	2.823	4.253
Z	8.00	8.00	8.00	8.00	8.00	8.00
Y	5.61	6.14	5.86	5.50	6.06	5.28
X	1.84	1.69	1.83	2.12	1.76	2.02
Fe ⁺³ /(Fe ⁺³ +Fe ⁺²)	0.14	0.05	0.15	0.07	0.55	0.24
(FeO+Fe ₂ O ₃)/(FeO+Fe ₂ O ₃ +MgO)	0.56	0.71	0.44	0.59	0.50	0.57
γ	1.644	1.656	1.626	1.630	1.618	1.642

Numbers correspond to those in Table 3. Analyst: N. Murakami

Table 5. Chemical compositions of the hornblendes in the Tamagawa plutonic rocks

No.	2 H	13 H	20 H
SiO ₂	51.30	49.48	52.00
TiO ₂	0.59	0.95	1.28
Al ₂ O ₃	3.08	4.86	4.69
Fe ₂ O ₃	2.40	1.16	3.89
FeO	12.61	17.25	7.34
MnO	0.34	0.93	0.73
MgO	13.96	10.90	14.41
CaO	10.56	9.51	11.23
Na ₂ O	2.29	1.19	1.68
K ₂ O	0.59	0.60	0.53
H ₂ O(+)	2.13	2.99	1.77
H ₂ O(-)	0.10	0.16	0.11
T.	99.95	99.98	99.66
Si	7.494	7.265	7.485
Al ^{IV}	0.506	0.735	0.515
Al ^{VI}	0.024	0.106	0.281
Ti	0.065	0.105	0.138
Fe ⁺³	0.264	0.128	0.421
Fe ⁺²	1.541	2.118	0.884
Mg	3.039	2.385	3.091
Mn	0.042	0.116	0.089
Ca	1.653	1.496	1.732
Na	0.649	0.339	0.469
K	0.110	0.112	0.097
OH	2.076	2.929	1.699
Z	8.00	8.00	8.00
Y	4.97	4.96	4.90
X	2.41	1.95	2.30
Fe ⁺³ /(Fe ⁺³ +Fe ⁺²)	0.15	0.06	0.32
(FeO+Fe ₂ O ₃)/(FeO+Fe ₂ O ₃ +MgO)	0.52	0.63	0.44
<i>a</i>	1.652	1.650	1.650
<i>β</i>	1.665	1.663	1.664
<i>γ</i>	1.672	1.670	1.673
2Vx	75°	73°	81°

Numbers correspond to those in Table 3.

Analyst : N. Murakami

iron-rich from gabbroic rocks to granodiorite, then become magnesium rich rapidly with decreasing SI, excluding the biotites in the aplite veins in the granodiorite and quartz diorite, whose refractive indices are higher than those in the latter rocks

Attention is focused on the large discrepancy between granites and associated biotites in the $(\text{FeO} + \text{Fe}_2\text{O}_3)/(\text{FeO} + \text{Fe}_2\text{O}_3 + \text{MgO})$ ratio which

Fig. 10. Relation between γ -index and SI in the Tamagawa plutonic rocks.

Fig. 11. Relations between biotites and rocks in the $(\text{FeO} + \text{Fe}_2\text{O}_3)/(\text{FeO} + \text{Fe}_2\text{O}_3 + \text{MgO})$ ratio and SI of the Tamagawa plutonic rocks. The data of the hornblendes are also plotted.

Fig. 12. Relation between the volume content of magnetites and the $(\text{FeO} + \text{Fe}_2\text{O}_3)/(\text{FeO} + \text{Fe}_2\text{O}_3 + \text{MgO})$ ratio of associated biotites.

shows reversed trends in the latter. Namely, the granites become iron-rich as differentiation progresses, while the biotites tend to be magnesium-rich. As the dominant ferromagnesian mineral in the granites is biotite, this discrepancy must be attributed to the formation of a fairly large amount of magnetite. In this respect, the relation is elucidated well by Fig. 12 in which the volume content of magnetite in the granites is inversely proportional to the $(\text{FeO} + \text{Fe}_2\text{O}_3)/(\text{FeO} + \text{Fe}_2\text{O}_3 + \text{MgO})$ ratio of associated biotites.

DISCUSSION

In the preceding chapter the writer described the variation of the iron/magnesium ratios of granites and associated biotites in connection with crystallization of magnetite, as summarized below.

1) Late Mesozoic to early Tertiary granites: With decreasing SI the iron/magnesium ratio increases gradually in the granites as well as in the biotites. The magnetite separation is generally low.

2) Palaeogene Tamagawa granites: The iron/magnesium ratio in the biotites drops rapidly during crystallization of granite magmas in spite of the increasing tendency of the ratio in the rocks. The magnetite separation is much advanced.

Fig. 13 shows the difference in the amount of magnetite in the above-cited two granite series.

The results similar to the Tamagawa granites have been obtained also from Ben Nevis Pluton, Scotland (Haslam, 1968) and Finnmarka Pluton, Norway (Czamanske, 1965), both of which occur in the area associated with cauldron. Comparing Figures 10 and 11 with Figures 15 and 16, their close affinity with the Tamagawa granites is evident.

The equilibrium relation between biotite and magnetite in the biotite

Fig. 15. Relations between biotites and rocks in the $(\text{FeO} + \text{Fe}_2\text{O}_3)/(\text{FeO} + \text{Fe}_2\text{O}_3 + \text{MgO})$ ratio and SI of the Ben Nevis Plutonics, Scotland. The data of the hornblendes are also plotted. Solid circle: Biotite. Half solid circle: Hornblende. Open circle: Rock.

temperature, an increase in oxygen partial pressure will result in the magnesium-rich biotite and a fairly large amount of magnetite. On the contrary, under decreasing oxygen partial pressure the iron-rich biotite and very little magnetite will coexist. The dependence of the amount of magnetite on the chemical composition of the ferromagnesian minerals in the volcanic rocks has been emphasized by Taneda (1949, 1950).

Based on experimental results, Wones and Eugster (1965) considered two contrastive trends of crystalliza-

tion of biotite. Trend I represents a magma in which oxygen fugacity remains constant or somewhat increase, whereas in Trend II oxygen fugacity decreases. It would seem possible that trend I applies to the Tamagawa granites together with Ben Nevis Pluton and Finnmarka Pluton. Fig. 12 is an attempt to indicate that the equilibrium corresponding to the above-cited equation was roughly established between biotite and magnetite. The late Mesozoic to early

Fig. 16. Relation between γ -index of biotites and SI in the Finnmarka Plutonics, Norway.

Tertiary granites might be referred to trend II, although other ferromagnesian minerals than biotite, such as pyroxene and olivine, have not been found. A similar result has been obtained for magnesioferrite by Muan and Osborn (1956).

As to the crystallization of biotite in the Tamagawa granites, the following processes are considered. From gabbroic rocks to granodiorite the biotite was crystallized under the conditions of slightly decreasing oxygen partial pressure, and thereafter the oxygen partial pressure began to increase gradually, forming the magnesium-rich biotite and euhedral magnetite (Fig. 7). In some cases the magnesium-rich hornblende was crystallized. As differentiation progressed and the magma reached to a granitic composition, the oxygen partial pressure increased rapidly, accompanied by the crystallization of highly magnesium-rich biotite and abundant magnetites. Under this condition the already-crystallized biotites were absorbed (Fig. 9), and produced more magnesium-rich biotite and interstitial magnetite (Fig. 8). Through similar process the magnesium-rich hornblende was transformed into more magnesium-rich one. The liquid of the feldspar composition in the granite magma which coexisted with two feldspars, plagioclase and alkali feldspar, rapidly varied from sodic to potassic in composition as a result of the dissociation of biotites. Thence, the equilibrium relation between two feldspars and liquid was not maintained and only one alkali feldspar and liquid could coexist (Stewart and Roseboom, 1962). This might have lead to the formation of some aplites almost devoid of plagioclase. Through these processes the liquid would react with already-crystallized plagioclase to make more sodic alkali feldspar. The final product would be the alkali feldspar mantling the plagioclase, so-called "antirapakivi" and overgrowth of alkali feldspar described already. This

Fig. 17. Reaction series for the constituent minerals in the Tamagawa plutonic rocks.

problem will be discussed in detail in the other paper.

Deduced from the above description, the reaction relations of component minerals in the Tamagawa granites are diagrammatized in Fig. 17.

In most calc-alkaline rock series, trend II by Wones and Eugster (1965) seems to be the case. The data by Heinrich (1946), Nockolds (1965), Tsuboi *et al.* (1938), Shibata *et al.* (1966), Tatekawa (1964), and Tatekawa and Fujimoto (1967) all suggest this case.

Osborn (1959, 1962) stressed the role played by oxygen partial pressure in the course of crystallization of magma in which the oxygen is assumed to result from the dissociation of water. However, this is likely to be the case for deep-seated magmas. Some other external source should be considered for the Tamagawa granites, because they are regarded as the intrusives at a too shallow depth* to retain a high water pressure, judging from their porphyritic texture, occurrence of aluminium-poor hornblende and other data. Haslam (1968) considered that a high level of oxygen pressure was possibly derived in part from the atmosphere through ring fracture and other fissures. This assumption may be valid for the Tamagawa granites, but the atmosphere kept within the bottom space of the cauldron may be another source of oxygen. It must be emphasized that the plutonic rocks with a reversed trend of the iron/magnesium ratio in biotite, occur mostly in the area associated with cauldron subsidence as exemplified by the Tamagawa, Ben Nevis and Finnmarka Plutonics.

SUMMARY

Two markedly different trends of evolution of biotites were found in the granitic rocks of Southwest Japan.

(1) Late Mesozoic to early Tertiary granites: The iron/magnesium ratios in both rocks and biotites increase with decreasing solidification index (SI). The difference of the ratio between rocks and biotites is small. The magnetite content in the granities is below 1.5 per cent usually.

(2) Palaeogene Tamagawa plutonic rocks: As the differentiation progresses, the iron/magnesium ratio has a similar trend as that in (1) from gabbro to granodiorite in both biotites and rocks, whereas from granodiorite to granite the ratio becomes reversed in the biotites due to their turning into magnesium-rich compositions. The content of magnetites is high, amounting to 4 per cent or more. They replace biotites and form interstitial fillings.

On the basis of physico-chemical aspects, it is assigned that the oxygen

* In general, the depth of cover of the plutonics is considered to be extremely shallow in the cauldron area; for example, the depth to the plutonics of the Georgetown Inlier, Australia is estimated at 2000 to 3000 ft., in part only 500 to 1000 ft. (Branch, 1967).

partial pressure during the evolution of biotites has been under slightly decreasing condition in the late Mesozoic to early Tertiary granites, while almost constant or somewhat increased in the Tamagawa plutonic rocks in which at least a part of the oxygen are presumed to have been derived from atmosphere, although its invading process is quite questionable. The latter case is common in the plutonics of the cauldron subsidence area.

The reaction series of the constituent minerals for the Tamagawa plutonic rocks is conspicuously differed from ordinary types of granite belonging to calc-alkaline rock series as shown in Fig. 17.

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花崗岩類中の黒雲母に見られる 2 つの異なった進化経路

村 上 允 英

摘 要

西南日本産花崗岩類中、それぞれ起源を共通する 2 時期のものについて全岩および含有黒雲母の化学組成を検討した結果、次のとおりいちじるしい差異が見いだされた。

(1) 後期中生代—初期第三紀花崗岩類：岩石の固化指数 (SI) の減少に伴って全岩・黒雲母ともに Fe/Mg 比が増加し、しかもその値の差が小さい。含有磁鉄鉱量も小さく通常 1.5% 以下である。

(2) 古第三紀田万川深成岩類：全岩・黒雲母の Fe/Mg 比は岩石が斑禰岩から花崗閃緑岩に進化する過程では増加する。しかし、花崗閃緑岩から花崗岩への進化過程では黒雲母の Fe/Mg 比はいちじるしく減少し、全岩の同比と逆の傾向を示す。含有磁鉄鉱量はいちじるしく多く(通常 1~4%, ときに以上), この磁鉄鉱は黒雲母を交代し、さらに間隙充填を示すものが普遍的に存在する。

以上の諸性状をもとにて考察した結果、(1)の花崗岩類は酸素分圧のやや減少する条件下で形成されたもので、(2)の深成岩類は酸素分圧がほとんど一定か、やや増加する条件下の性成と推定される。(2)のような深成岩類は一般に鍋状陥没地の深成岩類に特有で、酸素の少なくとも一部は大気中より何等かの経路を経て供給されたものであろう。このような深成岩類では黒雲母の分解に伴ってアルカリ長石の組成も急変し、いわゆる逆ラバキビ組織の形成をみる。したがって田万川深成岩類中の含有鉱物の反応系列 (Fig. 17) は一般の花崗岩類におけるものと可成り異なっている。