CHEMICAL FEATURES OF BIOTITES FROM METAMORPHIC AND GRANITIC ROCKS OF THE YANAI DISTRICT IN THE RYOKE BELT, JAPAN

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INTRODUCTION

Composition of biotites in metamorphic rocks has been discussed by many authors in terms of bulk composition and metamorphic grade of host rocks (*e.g.* Miyashiro, 1956; Engel and Engel, 1960; Ōki, 1961; Hayama, 1964; Wenk, 1970). It has been shown that Fe in biotite increases in general with increasing metamorphic grade, whereas Ti, Mg and Al decrease. However, the behaviours of these elements in metamorphic processes are not known in detail.

Biotites from some Caledonian calc-alkaline plutonic rocks follow a well-defined trend in the FeO-MgO-Al₂O₃ diagram (Nockolds and Mitchel, 1948). The ratio Al₂O₃: (MgO+total Fe as FeO) in biotites from calc-alkaline igneous rocks is determined by their paragenesis (Nockolds, 1947) and there is a general increase in the Al content of biotites with increasing acidity of the host rocks. However, some intrusions show a reverse trend and no agreement has been reached about the precise bearings of the Al content of biotites on magmatic processes.

Shibata *et al.* (1966) reported chemical analyses of biotites from metamorphic and granitic rocks of Japan and suggested that

the high Al contents of biotites from some Ryōke granitic rocks are due to contamination by pelitic sediments.

Majority of reported analyses of biotites from igneous rocks shows a general decrease in the Mg/Fe ratio with increasing acidity of the host rocks. However, reverse is the case with many intrusions (*e.g.* see Murakami, 1969; Kanisawa, 1972). Wones and Eugster (1965) determined experimentally the composition of biotites co-existing with sanidine and magnetite under varying conditions of f_{H_2O} , f_{O_2} and temperature and suggested that biotites crystallizing from a magma may follow either iron-rich or magnesium-rich trend, depending upon f_{O_2} during cooling.

Thirty-one biotites from metamorphic and granitic rocks of the Yanai district in the Ryōke belt, Southwest Honshū, Japan, have been chemically analysed as a part of the geochemical, mainly oxygen isotopic, study of the rocks. All the samples are believed to have been formed under roughly a uniform P-T condition and, hence, differences in their chemical compositions and parageneses would reflect those in the chemistry of their host rocks. In the present paper, this will be shown to be the case. Condition of formation of the meta-

morphic and granitic rocks of this district will also be discussed from the results. This paper is complementary to the previous one that described the major element chemistry of these rocks (Honma, 1974).

GEOLOGICAL SETTINGS

An outline of the geology of the area, a brief description of the host rocks of the most biotites of the present study and a geological map showing sampling localities are available in the previous paper (Honma, 1974).

The granitic rocks including migmatites are grouped into the syntectic layered (Kojima and Okamura, 1968) and discordant rock types. The former (the Ōbatake and Gamano layered granodiorites and the Okiura layered granite) are intimately associated with amphibolite and sedimentary gneiss. Though it may be granted that the main facies of the Ōbatake and Gamano layered granodiorites originated from a magma of the equivalent composition, petrological and structural evidence shows that "granitization" in a broad sense took place in the formation of these rocks (Okamura, 1957). However, such "granitization" including metasomatic formation of some minor facies of these layered rocks could occur as a result of intense interaction between the granitic magma and surrounding sedimentary gneiss or amphibolite (Honma, 1974).

The host rocks of the biotites of the present study are from the migmatite zone of Nureki (1960) with two exceptions (YN28 and YN38, from the transitional zone) and include amphibolites, pelitic and siliceous gneisses and granitic rocks. Mineral paragenesis within each kind of these metamorphic rocks is rather uniform and

the layered granitic rocks are intimately associated with metamorphic rocks, both forming thin layers intercalated by each other. Oxygen isotopic equilibrium temperatures of several mineral pairs from these rocks fall in a narrow range from 600° to 700°C (Honma, 1974, unpublished). These facts appear to suggest that most biotites studied were formed under roughly a constant P-T condition.

YN1-1, YN8-1 and YN24 are from rocks which were not previously studied. The first two are from siliceous banded gneisses of chert origin and are much the same as OS2-4 in occurrence and petrography. YN24 is a K-feldspar porphyritic two-mica granite named as the Kibe granite, one of the younger (discordant) Ryōke granites (Okamura, 1957). The chemical composition of the Kibe granite was given by Shibata *et al.* (1960). In the previous paper, YN15, an intrusive and garnet-bearing biotite leucogranite, was included into the Ōbatake and Gamano layered granodiorites. But it is similar in petrography and chemistry to the Okiura layered granite and is treated in this paper as a member of this granite.

EXPERIMENTAL

Separation of biotites was made by panning and repeated passing in a Frantz magnetic separator. The biotite samples analysed were better than 97 per cent in purity.

Analytical procedures are based on the method proposed by Ōki *et al.* (1962) using ion exchange columns. Alkalies were separated from each other and determined volumetrically. Weight loss by ignition corrected for oxidation of ferrous iron was taken as H_2O^+ . Replicate partial analyses

Table 1

No.	Composition of							
	1	2	3	4	5	6	7	8
Specimen	OS 2-4	YN 1-1	YN 8-1	YN 38	YN 28	OS 2-1	OS 8-2	OS 7-1
SiO ₂	34.17	33.99	34.70	34.67	33.03	34.03	34.40	34.08
TiO ₂	2.31	3.46	2.76	3.30	3.28	2.66	3.79	3.97
Al ₂ O ₃	18.34	18.37	18.86	19.33	18.86	19.49	14.42	14.96
Fe ₂ O ₃	3.73	2.80	2.46	1.81	0.44	3.17	2.18	2.28
FeO	15.00	14.90	16.17	19.53	18.96	17.73	19.37	20.19
MnO	0.58	0.34	0.51	0.11	0.21	0.50	0.29	0.16
MgO	9.15	9.66	8.40	6.82	9.41	6.99	8.64	8.85
CaO	0.11	0.37	0.33	0.58	0.75	0.39	0.95	0.54
Na ₂ O	0.46	0.32	0.64	0.34	0.64	0.48	0.50	0.22
K ₂ O	9.55	8.90	9.94	8.62	9.16	9.06	9.21	9.37
H ₂ O (+)	4.96	5.43	4.18	4.30	3.98	4.34	3.86	4.34
H ₂ O (-)	0.62	0.93	0.69	0.72	0.48	0.78	1.32	0.97
P ₂ O ₅	0.01	0.01	0.00	0.00	0.01	0.00	0.01	0.01
Total	98.99	99.48	99.64	100.13	99.21	99.62	98.94	99.94
Number of Atoms on Basis								
Si	5.17	5.20	5.28	5.26	5.11	5.20	5.38	5.28
Al ^{IV}	2.83	2.80	2.72	2.74	2.89	2.80	2.62	2.72
Al ^{VI}	0.44	0.51	0.66	0.72	0.55	0.71	0.03	0.03
Ti	0.26	0.40	0.32	0.37	0.38	0.31	0.44	0.46
Fe ³⁺	0.42	0.32	0.28	0.20	0.05	0.36	0.25	0.26
Fe ²⁺	1.90	1.91	2.06	2.48	2.46	2.27	2.53	2.62
Mn	0.07	0.04	0.07	0.01	0.03	0.06	0.04	0.02
Mg	2.06	2.20	1.90	1.54	2.17	1.59	2.01	2.04
Ca	0.02	0.06	0.05	0.09	0.12	0.06	0.16	0.09
Na	0.13	0.09	0.19	0.10	0.19	0.14	0.15	0.07
K	1.84	1.74	1.93	1.67	1.81	1.76	1.83	1.85
OH	5.01	5.55	4.25	4.36	4.11	4.42	4.02	4.49

1-3 : From siliceous gneiss 4-5 : From pelitic gneiss 6: From pelitic gneiss, partly migmatitic 7-9: From migmatitic facies of the Ōbatake and Gamano layered granodiorites associated with amphibolite 15-16 : granodiorites

were made for a biotite sample from a very leucocratic granite (YN6-1).

RESULTS AND DISCUSSION

Biotite analyses and atomic ratios of

cations and hydroxyl ion calculated on the basis of 24 (O, OH) are presented in Table 1. In most biotites, the sum of twelve-coordinated large cations, Ca, Na and K, is close to the ideal number of 2.00. Some

Biotites

Analyst : K. Ikehara and H. Fukui

9	10	11	12	13	14	15	16
OS 11	YN 3-1	OS 1	OS 8-1	YN 10	YN 3-2	OS10-2	YN 30
35.22	35.12	33.84	34.48	35.44	33.38	33.94	35.01
3.60	3.90	3.70	3.79	3.86	3.80	4.26	2.80
14.56	14.79	14.84	15.20	14.01	16.27	16.10	14.71
1.70	2.01	4.51	2.81	1.94	2.33	2.74	2.35
17.66	18.06	19.85	19.05	19.59	21.06	20.55	22.44
0.23	0.22	0.40	0.45	0.40	0.41	0.34	0.56
11.26	10.05	6.78	8.14	8.31	6.81	7.94	7.32
0.77	0.44	0.67	0.17	1.19	0.29	0.34	0.57
0.81	0.37	0.28	0.34	0.02	0.01	0.21	0.35
8.91	8.80	8.92	9.31	9.16	9.10	9.06	9.36
3.67	4.83	4.49	4.18	3.70	4.72	4.10	3.98
0.85	0.97	1.46	1.56	1.47	1.21	0.73	0.49
0.00	0.01	0.02	0.01	0.00	0.00	0.00	0.01
99.24	99.57	99.76	99.49	99.09	99.39	100.31	99.95
(O,OH) = 24							
5.45	5.35	5.29	5.38	5.58	5.20	5.24	5.48
2.55	2.65	2.71	2.62	2.42	2.80	2.76	2.52
0.11	0.00	0.03	0.18	0.18	0.19	0.17	0.19
0.42	0.45	0.43	0.44	0.46	0.45	0.49	0.33
0.33	0.23	0.53	0.33	0.23	0.27	0.32	0.28
2.29	2.30	2.60	2.49	2.58	2.74	2.66	2.94
0.03	0.03	0.05	0.06	0.05	0.05	0.04	0.07
2.60	2.28	1.58	1.89	1.95	1.58	1.83	1.71
0.13	0.07	0.11	0.03	0.20	0.05	0.06	0.10
0.24	0.11	0.08	0.10	0.01	0.00	0.06	0.11
1.76	1.71	1.78	1.85	1.84	1.81	1.78	1.87
3.79	4.91	4.68	4.35	3.89	4.91	4.23	4.15

amphibolite in agmatitic mass 10 : From amphibolite in small xenolith-like block 11-14; From coarse-grained granodiorite, the main facies of the Ōbatake and Gamano layered

biotites (OS2-4, YN1-1, YN3-1, YN10, YN3-2, YN3-3, OS4 and OS2-2) are higher in H_2O but normal in K_2O . The K_2O contents and microscopic inspection favour that they are free from weathering. This also is supported by their oxygen isotopic

ratios which are sensitive to weathering (Honma, 1974, unpublished).

In all the biotites analysed, the total number of trivalent cations in octahedral site is less than 1.5 (the limit of the number for natural trioctahedral biotites according

Table 1
Composition of

No.	17	18	19	20	21	22	23
Specimen	YN 9-1	YN29-1	YN 4-1	YN 4-2	YN 3-3	YN 8-3	OS 2-5
SiO ₂	35.22	34.90	32.98	34.15	32.65	33.83	33.01
TiO ₂	3.60	3.50	3.50	3.60	3.73	3.16	3.66
Al ₂ O ₃	14.73	14.37	14.68	15.08	16.81	19.07	16.03
Fe ₂ O ₃	1.74	2.42	4.72	3.21	2.76	1.83	2.75
FeO	20.11	22.62	20.80	22.15	20.72	20.54	19.96
MnO	0.42	0.34	0.42	0.39	0.50	0.47	0.53
MgO	8.52	6.84	6.03	6.10	6.35	5.92	6.35
CaO	0.40	0.62	0.49	0.65	0.18	0.42	0.67
Na ₂ O	0.32	0.47	0.33	0.30	0.59	0.35	0.74
K ₂ O	9.05	8.84	8.64	8.95	9.54	9.01	9.89
H ₂ O (+)	4.49	3.83	4.32	4.27	4.97	4.54	4.30
H ₂ O (-)	0.85	0.44	2.28	0.88	1.23	0.47	1.44
P ₂ O ₅	0.00	0.00	0.00	0.01	0.01	0.00	0.02
Total	99.45	99.19	99.19	99.74	100.04	99.61	99.35
Number of Atoms on							
Si	5.44	5.50	5.28	5.35	5.07	5.19	5.22
Al ^{IV}	2.56	2.50	2.72	2.65	2.93	2.91	2.78
Al ^{VI}	0.12	0.17	0.05	0.14	0.14	0.54	0.20
Ti	0.42	0.42	0.42	0.42	0.44	0.36	0.43
Fe ³⁺	0.20	0.29	0.57	0.38	0.32	0.21	0.33
Fe ²⁺	2.60	2.98	2.78	2.90	2.69	2.63	2.64
Mn	0.05	0.05	0.06	0.05	0.07	0.06	0.07
Mg	1.96	1.61	1.44	1.42	1.47	1.35	1.49
Ca	0.07	0.11	0.08	0.11	0.03	0.07	0.11
Na	0.10	0.14	0.10	0.09	0.18	0.10	0.23
K	1.78	1.78	1.76	1.79	1.89	1.76	1.99
OH	4.62	4.02	4.61	4.47	5.15	4.65	4.53

17-19 : From coarse-grained granodiorite, the main facies of the Ōbatake and Gamano layered migmatitic facies in YN3-2 22-25 : From the Okiura layered granite 26-27 : From migmatitic granite 29 : From the Kibe granite 30 : From basic facies of the Tōwa granite 31 : From fine-

to Deer *et al.*, 1962). However, in some of them, the number of cations in octahedral site is significantly less than the maximum number of six per formula unit. The strict layer charge relations, defined by

Foster (1960) and shown graphically by Deer *et al.* (1962), are much disturbed in these biotites.

Paragenesis of biotites and their Al₂O₃ contents

(Continued)

Biotites

Analyst : K. Ikehara and H. Fukui

24	25	26	27	28	29	30	31
OS 4	YN 15	OS 2-2	OS 2-3	YN 11	YN 24	OS 6	YN 6-1
34.50	34.62	33.89	34.06	34.31	34.60	33.03	n.d.
2.66	2.00	2.40	3.00	2.30	2.60	3.57	2.65
17.41	17.44	19.12	19.14	16.02	18.07	14.25	18.85
3.10	1.51	2.65	2.44	2.57	2.44	3.78	}(27.33)
21.69	22.66	16.53	16.95	23.59	19.71	24.27	
0.42	0.49	0.59	0.51	0.45	0.47	0.45	0.97
5.36	4.80	8.95	7.48	4.87	6.27	4.99	4.79
0.43	1.09	0.39	0.43	1.53	0.74	0.33	0.12
0.73	0.73	0.51	0.41	0.64	0.57	0.32	0.48
8.02	8.73	9.61	9.03	9.10	9.42	9.11	9.48
4.78	4.49	4.89	4.43	4.02	4.09	4.32	n.d.
0.41	0.72	0.70	1.49	0.26	0.78	1.25	n.d.
0.00	0.00	0.00	0.00	0.00	0.00	0.01	0.01
99.51	99.28	100.23	99.37	99.66	99.76	99.68	
Basis (O,OH) = 24							
5.30	5.40	5.10	5.22	5.40	5.35	5.27	
2.70	2.60	2.90	2.78	2.60	2.65	2.68	
0.45	0.60	0.49	0.68	0.37	0.64	0.00	
0.31	0.23	0.27	0.35	0.27	0.31	0.43	
0.36	0.17	0.30	0.28	0.30	0.28	0.45	
2.78	2.95	2.08	2.17	3.11	2.55	3.24	
0.05	0.06	0.08	0.07	0.06	0.06	0.06	
1.22	1.12	2.01	1.17	1.14	1.44	1.19	
0.07	0.18	0.06	0.07	0.26	0.12	0.06	
0.22	0.22	0.15	0.12	0.19	0.17	0.10	
1.57	1.74	1.85	1.76	1.83	1.86	1.85	
4.89	4.66	4.91	4.53	4.22	4.21	4.59	

granodiorites 20 : From biotite-rich schlieren in YN4-1 21 : From K-feldspar porphyroblastic facies of the Okiura layered granite associated with siliceous gneiss 28 : From the Murotsu grained, biotite granite

The biotites of the present study may be grouped into three types by their paragenesis, that is, biotites associated with hornblende, those with muscovite and those that lack both of them. Biotites

from amphibolite are in intimate association with hornblende and those from sedimentary gneiss are with muscovite, whereas in granitic rocks, biotites occur in the three types of paragenesis.

Fig. 1. The MgO-total iron as FeO-Al₂O₃ (weight per cent) diagram for biotites classified by paragenesis (a) and by type of the host rock (b). C: field for Caledonian plutonic biotites (Nockolds and Mitchell, 1948); M: trend of biotites from acid members of Murrumbidgee Batholith (Joyce, 1973); I, II and III are the fields defined by Nockolds (1947) for biotites from calc-alkaline igneous rocks associated with muscovite, without muscovite or hornblende and with hornblende respectively. Y: YN; S: OS

As is shown in Fig. 1a, the three types of biotites are distinct from each other in their relative Al₂O₃ content, each confining themselves in the three narrow fields defined by Nockolds (1947) for the corresponding biotites from calc-alkaline igneous rocks.

The distribution of Al between octahedral and tetrahedral sites in biotite is illustrated in Fig. 2. Both Al^{IV} and Al^{VI}

Fig. 2. The distribution of aluminium between octahedral and tetrahedral sites in biotite. Grouping of biotites by their paragenesis.

show only limited ranges of variation within each group of paragenesis. Inspection of Fig. 2 reveals that in biotites associated with muscovite, aluminium has a stronger site preference to octahedral site than in other two groups.

As is shown in Fig. 3, the muscovite-bearing rocks (sedimentary gneiss, migmatitic facies of the Okiura layered granite and many of the discordant granites) have excess aluminium to form normative corundum, whereas other two rocks, especially, those of hornblende-biotite paragenesis, are deficient in aluminium over calcium, showing normative wollastonite. These facts strongly indicate that the paragenesis of the biotites, their Al₂O₃ contents and the partition of Al between octahedral and tetrahedral sites are, in most cases, controlled by the chemistry of their host rocks.

Fig. 3. Variation in Al_2O_3 content of biotite plotted as a function of normative w_o or C of the host rock. Grouping of biotites by their paragenesis.

Fig. 4. Variation in the MgO/FeO ratio in biotite vs. the content of alkalis of the host rock. Grouping of biotites by type of their host rocks. Dashed arrows show possible variation trends of granite during assimilation.

The MgO/FeO ratio in biotites

Fig. 1b indicates that MgO/FeO ratios vary rather widely within each group,

biotites from the metamorphic rocks having higher ratios than those from the granitic rocks.

Fig. 4 shows the variation in the MgO/FeO ratio of biotites with solidification of magma or "granitization" of metamorphic rock. In this diagram, the $\text{MgO}/(\text{MgO}+\text{FeO})$ ratio of biotite is plotted against the alkalis/(alkalis+ $\text{MgO}+\text{FeO}$) ratio of the host rock. The SiO_2 content cannot be used as an index of "granitization," because "granitization" of siliceous gneiss causes complementary compositional changes including an increase in alkalis and a decrease in SiO_2 and because K_2O , Na_2O , MgO and CaO in granoblastic amphibolite vary markedly regardless of the SiO_2 content (Honma, 1974).

The MgO/FeO ratio of biotites in this district shows a general trend of decrease with increasing contents of alkalis in the host rocks (Fig. 4). The ratio is similar to and varies in parallel with the host rocks (Fig. 5).

The latter trend is interpreted by the facts that the granitic rocks as well as the

Fig. 5. Relationship of the MgO/FeO ratio in biotite to that in the host rock.

Fig. 6. Relationship of the MgO/FeO ratio in biotite to that in coexisting hornblende (*unpublished data of the present author).

metamorphic rocks of the present study almost lack magnetite and that, as is shown in Fig. 6, hornblende, the other major ferro-magnesian mineral, has almost the same Mg/Fe ratio as biotite. Cordierite or garnet occurs only as a minor constituent in sedimentary gneisses and does not affect the Mg/Fe ratio of biotite.

According to Kanisawa (1972), the

Fig. 7. The Fe^{3+} - Fe^{2+} - Mg^{2+} (cation per cent) diagram for biotites. Dashed lines represent compositions of "buffered" biotites in the ternary system $\text{KFe}_3^{+3}\text{AlSi}_2\text{O}_{13}\text{H}_{-1}$ - $\text{KFe}_2^{+2}_3\text{AlSi}_3\text{O}_{10}(\text{OH})_2$ - $\text{KMg}_3\text{AlSi}_2\text{O}_{10}(\text{OH})_2$ by Wones and Eugster (1965). Grouping of biotites by type of the host rock.

Ryōke granitic rocks of the Takatō district, Central Japan, are distinctly higher in $(\text{Mg}/\text{Fe})_{\text{biotite}}/(\text{Mg}/\text{Fe})_{\text{hornblende}}$ than other granitic rocks. The high value of this ratio (close to 1) may be one of the characteristic features of the Ryōke granitic rocks. *Temperature and oxygen fugacity during the formation of biotites*

In the Fe^{3+} - Fe^{2+} - Mg^{2+} diagram shown in Fig. 7, many biotites studied are near Ni-NiO-buffered biotites estimated by Wones and Eugster (1965). According to them (Fig. 2 and Fig. 3), Ni-NiO-buffered biotites with $\text{Fe}/(\text{Fe}+\text{Mg})$ of 0.75 are stable below 680°C at a total pressure of 1035 bars and below 710°C at 2070 bars. Oxygen isotopic equilibrium temperatures of quartz-biotite pairs from the granitic rocks fall in a narrow range between 600° and 700°C (Honma, 1974, unpublished). These facts strongly suggest that the biotites were formed at a temperature between 600 and 700°C and under a low oxygen partial pressure close to the Ni-NiO-buffered system.

Tsue and Ishihara (1974) mentioned that contents of opaque minerals of the Ryōke granitic rocks are low, reflecting a low oxygen fugacity during their formation. Observed evidence for intimate association and interaction of the layered granitic rocks with sedimentary gneiss suggests that a large amount of carbonaceous matter in the pelitic rocks should be responsible for the low oxygen fugacity during the formation of these granitic rocks and, hence, for the absence of magnetite in most of them. Progressive reduction of oxygen fugacity with increasing metamorphic grade has been suggested to occur owing to the removal of oxygen by graphite in pelitic rocks (Miyashiro, 1974).

Biotites from the three types of granitic rocks

As is seen from Fig. 1b, biotites from metamorphic rocks (amphibolite, pelitic gneiss and siliceous gneiss) are higher in MgO/FeO than those from granitic rocks. Biotites from amphibolite are much poorer in Al_2O_3 than those from sedimentary gneisses.

Biotites from coarse-grained granodiorite, the main facies of the Ōbatake and Gamano layered granodiorites, vary only in MgO/FeO and follow the variation trend of the biotites from amphibolite. Almost all biotites from migmatitic facies of these granodiorites follow this trend, although a biotite from porphyroblastic K-feldspar-bearing migmatitic granite (YN3-3, one of minor facies of the Ōbatake and Gamano layered granodiorites) as well as one from migmatitic granodiorite (YN3-2) associated with this granite (YN3-3) deviate from the trend. YN3-3 is chemically equivalent to the main facies of the Okiura layered granite (Honma, 1974) and might have been formed from YN3-2 through metasomatic enrichment of normative Q and or .

Biotites from the main facies of the Okiura layered granite (OS2-5, OS4 and YN15) follow a general trend characteristic of the biotites from acid members of Caledonian plutonic rocks (Nockolds and Mitchel, 1948) or Murrumbidgee Batholith (Joyce, 1973). The difference in the variation trend between the main facies of the Ōbatake and Gamano layered granodiorites and the Okiura layered granite given above may reflect difference in the mode of formation between these rocks.

Points for biotites from the discordant granites are much scattered and, except for those from the Kibe granite (YN 24), they are characteristically low in MgO/FeO .

Biotites from migmatitic facies of the Okiura layered granite (OS2-2 and OS2-3) which forms thin bands in siliceous gneiss (OS2-4) differ much from those of the main facies of this granite. The siliceous gneiss should be responsible for distinctly high MgO/FeO and Al_2O_3 in them. The high Al_2O_3 content of biotite from garnet-bearing two-mica granite (YN8-3) alternating with siliceous gneiss may also reflect a contribution of alumina from the sedimentary gneisses.

A material balance calculation indicates that MgO/FeO ratios of more mafic rocks which existed prior to the "granitization" must have been changed only slightly by addition of granitic materials. This is confirmed by Fig. 4 where only small differences in $\text{MgO}/(\text{MgO}+\text{FeO})$ in biotites are recognized between the paired rocks: coarse-grained leucosome in agmatitic mass (OS8-1) and associated amphibolite (OS8-2); porphyroblastic K-feldspar-bearing migmatitic granite (YN3-3) and associated medium-grained migmatitic granodiorite (YN3-2). The biotites from the siliceous gneiss-migmatitic granite association (OS2-4, OS2-2 and OS2-3) show a similar trend as is shown in Fig. 4. This also is interpreted by the fact that the siliceous gneiss has a higher content of $(\text{MgO}+\text{FeO})$ relative to alkalis than the granitic magma.

On the other hand, the $\text{MgO}/(\text{MgO}+\text{FeO})$ ratio of granite must have changed markedly by assimilation of rocks which are high in $(\text{MgO}+\text{FeO})$ relative to alkalis. The trend of biotites from the Okiura layered granite may reflect that the sedimentary gneiss played a significant role in controlling the chemistry of some of this type of granites.

The trend of biotites from the Ōbatake

and Gamano layered granodiorites indicates that MgO of the magma was used up by biotite and hornblende in earlier stages of crystallization. The low MgO/FeO ratio in biotites from the discordant granites which are the later intrusives may similarly be interpreted.

CONCLUDING REMARKS

The relationship between composition and paragenesis defined by Nockolds (1947) for biotites from calc-alkaline igneous rocks hold for those from various kinds of rocks from highly metamorphic zone of the Yanai district including pelitic and siliceous gneisses, amphibolite, two types of syntectitic layered granitic rocks and the discordant granites. The main difference in composition among the biotites of each paragenesis is in the content of Al_2O_3 . Both Al^{IV} and Al^{VI} in the biotites of each paragenesis vary within limited ranges. It is found that both the Al_2O_3 content and paragenesis of these biotites are controlled by the chemistry of their host rocks. The similar relation is seen in the MgO/FeO ratio between biotite and the host rock. These facts are in accord with the geological postulation that all the biotites were formed at roughly an equal P-T condition.

There is a marked difference in the content of Al_2O_3 in biotite between amphibolite and sedimentary gneiss. The biotites from the main facies of the Ōbatake and Gamano layered granodiorites follow a well-defined trend of biotites from the amphibolite without any significant variation in the relative content of Al_2O_3 . This strongly supports the view that these layered granodiorites are magmatic in origin or, at least, sedimentary gneiss played no important role in the formation of the

main facies of these granodiorites. On the other hand, unusually high contents of Al_2O_3 of biotites from some facies of the Okiura layered granite suggest a contribution from sedimentary gneiss to this granite.

Variation in MgO/FeO ratio in the rocks of this district is distinctly reflected in the ratio in biotites; the ratios in biotites and their host rocks are similar to and vary in parallel with each other, decreasing with increasing alkalis in the host rocks. This may be due to: (1) interaction or exchange of materials between granitic magma and rocks which existed prior to its intrusion and (2) predominance of hornblende and biotite over other ferromagnesian minerals, the nearly equal partitioning of MgO and FeO between the two minerals and using up of MgO by biotite and hornblende in earlier stages of crystallization.

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柳井地方領家帯の変成岩類と花崗岩類中の黒雲母の化学的特徴

本 間 弘 次

高変成域の岩石中の黒雲母 31 試料を分析した。泥質および珪質片麻岩、角閃岩、2種の層状花崗岩類、非調和性花崗岩類など多様な岩石でも、白雲母や角閃石との共生関係と、黒雲母の Mg-Fe-Al 組成との間に規則的な関係が認められる。黒雲母の Al 含量は、母岩の化学組成に支配される。Mg/Fe 比は母岩のそれと一致し、酸素分圧による磁鉄鉱の欠除、黒雲母と角閃石間に Mg・Fe 分別の殆どないことによる。黒雲母の Al 含量は、母岩の型ごとに一定で、花崗岩類の一部の黒雲母の Al 含量が高いのは、片麻岩の影響によるものと考えられる。黒雲母の Mg/Fe 比も、各岩型ごとに特徴的な変化をするが、母岩中のアルカリの相対含量の増加と共に規則的に減少する。これは、マダマと変成岩との物質交換、角閃石、黒雲母による Mg の消費などによるものと考えられる。