CHEMICAL CHARACTERISTICS OF BIOTITES AND HORNBLENDES FROM METAMORPHIC ROCKS AROUND LÜTZOW-HOLMBUKTA, EAST ANTARCTICA

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Abstract: Chemical compositions of biotites and hornblendes in metamorphic rocks around Lützow-Holmbukta show that these minerals have the characteristics of granulite facies ones. Some low-Ti biotites and hornblendes may be due to low TiO_2 content of host rocks. Chemical natures of biotites and hornblendes reflect those of their host rocks, that is, biotites from metabasites are phlogopite and Na_2O-rich ones, while those from paragneisses are Fe-rich and Na_2O-poor ones. The replacement of R^{2+} by R^{3+} ions in octahedral sites in biotites increases from metabasites through charnockites to paragneisses. Hornblendes from metabasites are richer in Na_2O than K_2O, but the proportion is reversed in hornblendes from charnockites and hornblende gneisses. The values of log (f_{H_2O}/f_{HF}) of fluid phase coexisting with biotites during crystallization are 4.3 in metabasites and 3.8 in paragneisses, respectively. From the available data of the F content of hydrous minerals in various metamorphic terrains, it can be presumed that the ratio of f_{H_2O}/f_{HF} decreases during progressive metamorphism.

1. Introduction

Mineralogical and petrological investigations on metamorphic rocks around Lützow-Holmbukta have been carried out by BANNO *et al.* (1964 a, b) and SUWA (1966). According to BANNO *et al.*, the metamorphic rocks of the district were formed under the condition of hornblende-granulite facies metamorphism. Further, YOSHIDA (1979) has concluded from his structural and petrological study that the metamorphic rocks of the district are the products of polymetamorphism, and that three stages of metamorphism can be distinguished.

In this paper, chemical characteristics of biotites and hornblendes from the metamorphic rocks of the district will be described and the behavior of fluorine in these minerals during progressive metamorphism will be discussed.

2. Chemical Composition of Biotites and Hornblendes

Most of the metamorphic rocks of the district are composed of metabasites (ultrabasic and basic granulites, pyroxenites, hornblendites, amphibolites and eclogites), charnockites (pyroxene gneisses), paragneisses (hornblende gneisses, garnet-biotite gneisses and biotite gneisses). Eleven biotites and seven hornblendes were

Table 1. Chemical analyses of biotites from metamorphic rocks around Lützow-Holmbukta.

	1	3	4	5	6	7	8	9	10	11	12
SiO_2	38.68	38.39	38.71	37.98	38.36	36.43	38.11	35.82	37.07	37.41	36.39
TiO_2	1.07	2.74	0.46	3.37	3.16	5.70	5.03	6.36	5.88	5.15	5.65
Al_2O_3	16.04	16.69	16.66	15.61	15.28	14.17	13.92	16.07	15.70	16.19	14.59
Fe_2O_3	1.29	2.26	0.95	2.19	0.67	0.51	2.41	3.13	1.07	1.91	2.61
FeO	10.36	9.77	10.27	8.30	8.64	18.60	15.34	14.37	15.67	16.72	15.60
MnO	0.06	0.05	0.04	0.03	0.03	0.06	0.19	0.03	0.08	0.03	0.07
MgO	18.21	17.40	18.70	19.63	19.97	10.88	12.75	11.59	12.07	9.86	11.52
CaO	0.42	0.19	0.60	0.61	0.29	0.19	0.15	0.14	tr	0.25	tr
Na_2O	1.33	0.51	0.55	0.51	0.57	0.33	0.14	0.31	0.15	0.44	0.42
K_2O	9.23	8.81	8.62	8.82	9.03	9.47	9.36	9.30	9.53	8.94	9.92
H_2O+	2.35	2.57	4.25	3.09	3.45	3.29	2.37	2.95	2.30	2.98	2.26
H_2O-	tr	0.08	0.10	0.05	0.25	0.14	0.05	0.12	0.06	0.13	0.04
F	0.85	0.71	0.73	0.32	n.d.	0.66	0.72	0.44	1.27	n.d.	0.51
	99.89	100.17	100.64	100.51	—	100.43	100.54	100.63	100.85	—	99.58
F=O	0.36	0.30	0.31	0.13	—	0.28	0.30	0.19	0.53	—	0.21
Total	99.53	99.87	100.33	100.38	99.70	100.15	100.24	100.44	100.32	100.01	99.37
	Anhydrous basis of O=22										
Si	5.589	5.506	5.610	5.424	5.521	5.512	5.613	5.296	5.463	5.545	5.446
Al^{IV}	2.411	2.494	2.390	2.576	2.479	2.488	2.387	2.704	2.537	2.455	2.554
Z	8.000	8.000	8.000	8.000	8.000	8.000	8.000	8.000	8.000	8.000	8.000
Al^{VI}	0.321	0.326	0.455	0.050	0.113	0.038	0.029	0.095	0.203	0.372	0.018
Ti	0.116	0.295	0.050	0.362	0.342	0.648	0.557	0.707	0.651	0.574	0.635
Fe^{3+}	0.141	0.245	0.104	0.235	0.073	0.058	0.267	0.348	0.119	0.214	0.295
Fe^{2+}	1.251	1.171	1.245	0.991	1.040	2.352	1.889	1.776	1.913	2.072	1.952
Mn	0.008	0.006	0.005	0.003	0.003	0.008	0.031	0.004	0.010	0.004	0.009
Mg	3.919	3.718	4.037	4.176	4.282	2.452	2.799	2.553	2.650	2.177	2.568
Y	5.756	5.761	5.896	5.817	5.853	5.556	5.572	5.483	5.546	5.413	5.477
Ca	0.065	0.029	0.093	0.093	0.045	0.031	0.024	0.022	—	0.040	—
Na	0.373	0.141	0.155	0.141	0.156	0.096	0.041	0.089	0.043	0.126	0.122
K	1.701	1.611	1.593	1.606	1.667	1.826	1.759	1.753	1.791	1.690	1.919
X	2.139	1.781	1.841	1.840	1.868	1.953	1.824	1.864	1.834	1.856	2.041
mg	0.74	0.72	0.75	0.77	0.79	0.50	0.56	0.55	0.57	0.49	0.53
$6-\sum R^{2+}$	0.82	1.11	0.71	0.83	0.68	1.19	1.28	1.67	1.43	1.75	1.47
Oct. R^{3+}	0.58	0.87	0.61	0.65	0.53	0.74	0.86	1.15	0.97	1.16	0.95
Tet. R^{3+}	2.41	2.49	2.30	2.58	2.48	2.49	2.39	2.70	2.54	2.46	2.55

Analyst: H. ONUKI. F determination by S. KANISAWA.

separated and analysed chemically. The results are shown in Tables 1 and 2. Mineral assemblages of the host rocks are as follows.

1) Metabasites

1. 68091201-1. Biotite-hornblende eclogite (Loc. East Ongul). Ga-ho-opx-cpx-pl-bi.
2. 68021509. Hornblende eclogite (Loc. West Ongul). Opx-cpx-ho-ga-pl.
3. 68040105. Biotite-garnet amphibolite (Loc. West Ongul). Bi-ho-ga-pl-ore.
4. 68091201-2 Biotite-orthopyroxene-hornblende plagioclase rock (Loc. East Ongul). Bi-opx-ho-pl-(ore).
5. 68051904. Orthopyroxene hornblendite (Loc. Langhovde). Opx-ho-(bi).

Table 2. Chemical analyses of hornblendes from metamorphic rocks around Lützow-Holmbukta.

	1	2	3	4	5	7	8
SiO_2	40.85	40.56	40.32	39.81	40.72	40.22	40.05
TiO_2	0.62	1.52	1.47	0.31	1.53	2.53	2.02
Al_2O_3	17.43	14.79	16.88	16.93	14.81	13.01	11.51
Fe_2O_3	2.46	5.42	3.17	1.86	1.24	5.02	6.75
FeO	9.83	10.19	9.23	10.19	10.16	13.22	12.18
MnO	0.06	0.06	0.17	0.10	0.16	0.20	0.30
MgO	12.22	10.70	12.29	12.67	14.02	9.51	10.29
CaO	11.17	11.16	10.50	11.30	12.12	11.28	10.77
Na_2O	1.79	2.11	3.01	2.23	2.14	1.54	1.71
K_2O	1.39	1.74	1.03	2.21	1.44	1.85	1.74
H_2O+	2.27	1.66	1.81	1.90	1.69	1.44	2.28
H_2O-	0.01	0.08	0.03	tr	0.03	0.03	0.04
F	0.26	0.10	0.36	0.29	0.19	0.34	0.38
	100.40	100.09	100.27	99.80	100.15	100.19	100.02
F≡0	0.11	0.04	0.15	0.12	0.08	0.14	0.16
Total	100.29	100.05	100.12	99.68	100.07	100.05	99.86
Anhydrous basis of O=23							
Si	5.963	5.893	5.896	5.897	5.976	5.998	6.093
Al^{IV}	2.037	2.107	2.104	2.103	2.024	2.002	1.907
Z	8.000	8.000	8.000	8.000	8.000	8.000	8.000
Al^{VI}	0.967	0.495	0.800	0.853	0.537	0.284	0.156
Ti	0.068	0.081	0.161	0.035	0.169	0.284	0.231
Fe^{3+}	0.270	0.608	0.347	0.208	0.137	0.562	0.773
Fe^{2+}	1.197	1.271	1.125	1.262	1.234	1.648	1.549
Mn	0.008	0.008	0.021	0.012	0.020	0.025	0.038
Mg	2.653	2.380	2.696	2.795	3.064	2.113	2.332
Y	5.163	4.843	5.150	5.165	5.161	4.916	5.079
Ca	1.744	1.784	1.645	1.793	1.905	1.801	1.755
Na	0.506	0.610	0.851	0.641	0.618	0.444	0.504
K	0.259	0.332	0.193	0.418	0.270	0.351	0.338
X	2.509	2.726	2.689	2.852	2.483	2.596	2.597
mg	0.64	0.56	0.64	0.65	0.69	0.49	0.50
Na+K	0.765	0.942	1.044	1.059	0.878	0.795	0.842
$Fe^{3+}+Ti+Al^{VI}$	1.305	1.184	1.308	1.096	0.843	1.130	1.160

Analyst: H. ONUKI. F determination by S. KANISAWA.

6. 68032704. Pyroxenite (Loc. East Ongul). Opx-cpx-bi.

2) Charnockite

7. 68032310. Orthopyroxene-biotite-hornblende gneiss (Loc. West Ongul). Opx-bi-qz-pl-(K. fel)-(ore).

3) Paragneisses

8. 68022607. Biotite-hornblende gneiss (Loc. West Ongul). Bi-ho-qz-K. fel-ore.
9. 68090706. Biotite-garnet gneiss (Loc. Ongulkalven). Bi-ga-pl-(K. fel).
10. 68051908. Garnet-biotite gneiss (Loc. Langhovde). Ga-bi-qz-pl-(ore).
11. 68032313. Garnet-biotite gneiss (Loc. West Ongul). Ga-bi-qz-pl-K. fel-(ore).
12. 68022002. Biotite gneiss (Loc. West Ongul). Bi-qz-pl-K. fel.

Minerals in parentheses are "present but small in amount".

Biotites from metabasites are rich in Mg, with mg-values ranging from 0.72 to 0.79, and belong to phlogopite according to the classification of DEER *et al.* (1963). The Al_2O_3 content of the biotites is rather high, being about 16%, and the TiO_2 content is rather low showing 0.46–3.37%. Si in formula unit is about 5.5 (anhydrous basis of O=22) in all biotites. However, biotites from charnockite and paragneisses have mg-values of 0.49 to 0.57 and are rich in Fe. They are titanobiotites having TiO_2 over 5%. These characteristics of biotites are also shown in the data by BANNO *et al.* (1964 a, b). The MnO content is very low in all biotites. Such TiO_2-rich and MnO-poor biotites were frequently reported from other granulitic terrains such as Kondapalli, India (LEELANANDAM, 1970). Rich TiO_2 and poor MnO are characteristic of the charnockitic biotites (BANNO *et al.*, 1964 b). The Na_2O content is the highest in biotite from eclogite (68091201–1), the lowest in those from paragneisses, and intermediate in those from other matabasites. Biotites rich in Na_2O are also often found in ultrabasic to basic granulites in Kondapalli (LEELANANDAM, 1970). The $Fe^{3+}/(Fe^{2+}+Fe^{3+})$ ratios are all smaller than 0.2, mostly less than 0.1. The number of R^{2+} cations displaced, which is represented as $6-\Sigma R^{2+}$, is plotted against the number of proxying octahedral R^{3+} cations of biotites of the district (Fig. 1), according to FOSTER (1960). It is obvious that most biotites fall on the line which represents the proxy ratio $R^{3+}:R^{2+}=0.67:1$, and consequently there is a general decrease in octahedral occupancy from 5.90 to 5.41 (Table 1) with increasing replacement of divarent ions by trivarent ones in the octahedral group. The ratio of replacement of R^{3+} is smaller in biotites from metabasites, larger in those from paragneisses, and intermediate in those from charnockites. The amounts of both octahedral R^{3+} and tetrahedral R^{3+} ions are slightly larger in biotites from paragneisses and charnockites than in those from metabasites (Fig. 2).

The chemical composition of analysed hornblendes shows a range of mg-value from 0.69 to 0.49, and hornblendes from metabasites are richer in Mg and Al, especially in Al^{VI}, and poorer in Ti than those from charnockites and hornblende gneisses. All hornblendes are rich in (Na+K) and Al^{IV} and have nearly constant values of them,

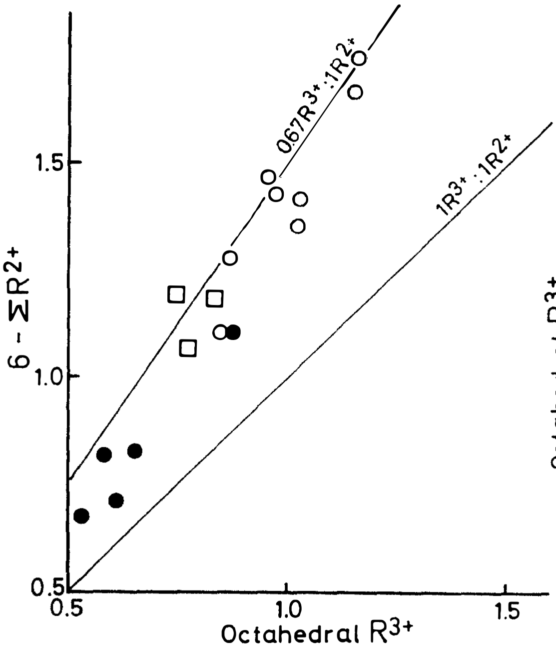

Fig. 1. *Relationship between the R^{2+} cations displaced and the proxying R^{3+} octahedral cations in Lützow-Holmbukta biotites. The data presented by* Banno *et al. (1964 b, Nos. 31–35) are adopted in addition to the present analyses. Solid circle: Biotites from metabasites. Open square: Biotites from charnockites. Open circle: Biotites from paragneisses.*

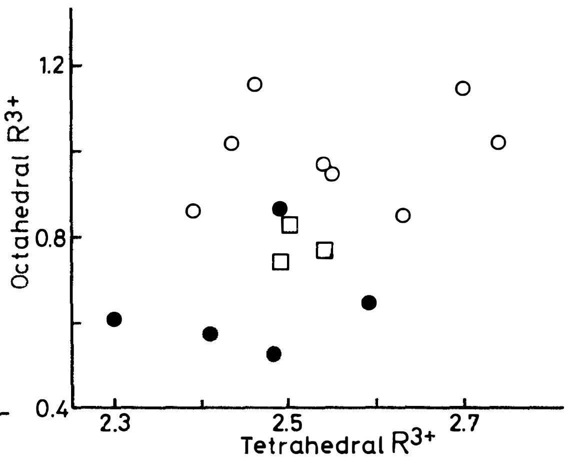

Fig. 2. *Relationship between the octahedral and the tetrahedral R^{3+} cations in Lützow-Holmbukta biotites. Symbols are the same as those in Fig. 1.*

thus they belong to pargasite in the diagrams of Deer *et al.* (1963) as shown in Fig. 3. Al^{IV} is slightly higher in hornblendes from metabasites than those from charnockites or hornblende gneisses. Ti in formula unit (anhydrous basis of O=23) varies over a wide range from 0.035 to 0.284, which represents the values from the greenschist-amphibolite transition facies to the hornblende-granulite facies in the diagram of Raase (1974) as shown in Fig. 4. Especially, hornblendes from eclogite (68091201–1) and metabasite (68091201–2) have low TiO_2 content of 0.62 and 0.31 %, respectively and they coexist with low-Ti biotites and are free from any other Ti-bearing phases, thus Ti in hornblendes depends largely on the TiO_2 content in host rocks and the modal ratios of hornblende and biotite in addition to the metamorphic grade. The H_2O+ content in hornblendes of the district is generally low, whereas the F content is rather high. The $Fe^{3+}/(Fe^{2+}+Fe^{3+})$ ratio is variable from 0.10 to 0.33. Such Al^{IV}-, (Na+K)- and F-rich and H_2O+ poor pargasitic hornblendes have been reported also from the other upper amphibolite to the granulite facies

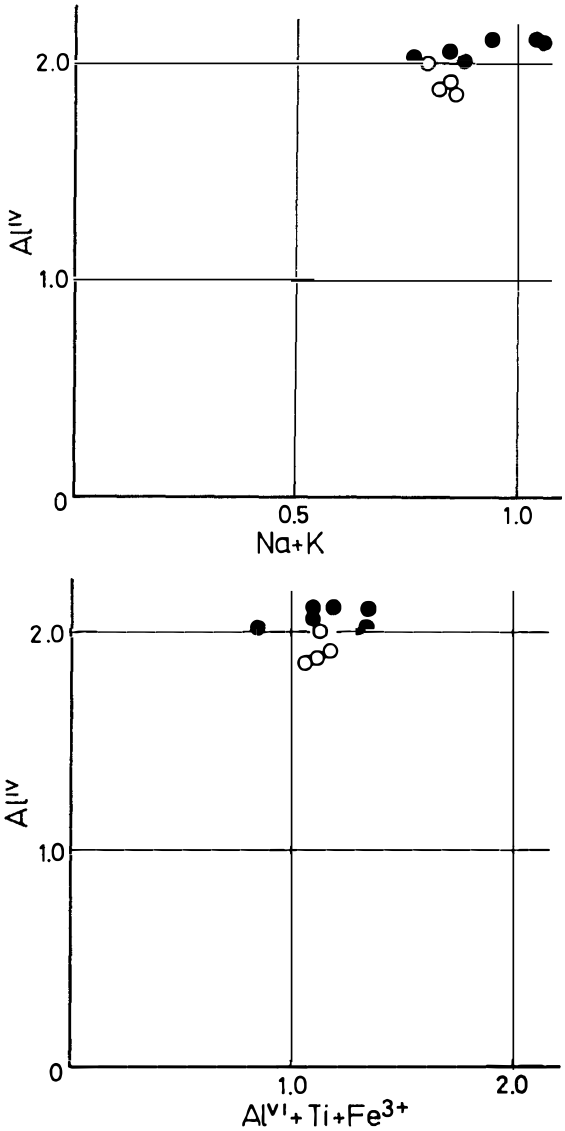

Fig. 3. *Relationship between the* Al^{IV} *and* $(Na+K)$ *atoms and the* Al^{IV} *and* $(Al^{VI}+Ti+Fe^{3+})$ *atoms in Lützow-Holmbukta hornblendes. The data presented by* BANNO *et al.* (*1964 b, Nos. 22–24*) *are also plotted in the figure. Solid circle: Hornblendes from metabasites. Open circle: Hornblendes from charnockites and hornblende gneisses.*

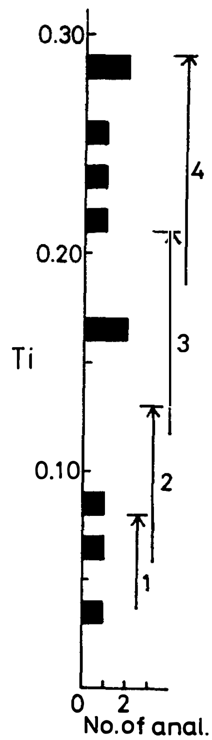

Fig. 4. *Histogram of Ti contents of Lützow-Holmbukta hornblendes. Arrowes 1, 2, 3 and 4 indicate the upper limits and the ranges of Ti in hornblendes in various metamorphic facies after* RAASE (*1974*). *1; Greenschist-amphibolite transition facies. 2: Lower-grade amphibolite facies. 3: Higher-grade amphibolite facies. 4: Hornblende granulite facies.*

metamorphic terrains such as the Adirondack Mountains (ENGEL and ENGEL, 1962), Kondapalli (LEELANANDAM, 1970) and Madras (HOWIE, 1955). In Lützow-Holmbukta, hornblendes from metabasites are all richer in Na_2O than K_2O, but the proportion is reversed in those from charnockites and hornblende gneisses. Hornblendes from ultrabasic granulites have higher Na_2O than K_2O, but the proportion is reversed in those from basic granulites in Kondapalli and Madras (LEELANANDAM, 1970).

3. Behavior of Fluorine in Biotites and Hornblendes

Biotites and hornblendes in rocks of Lützow-Holmbukta are rather rich in F.

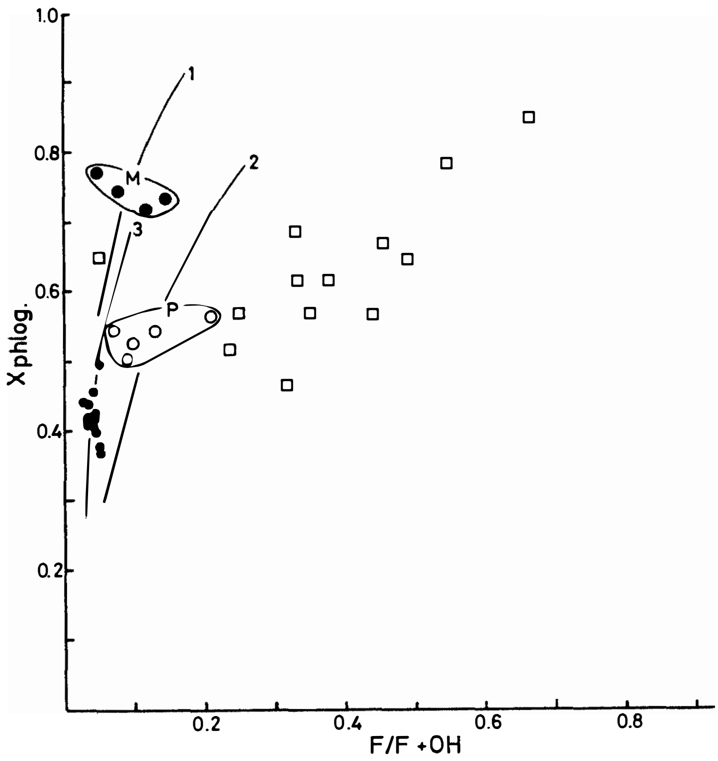

Fig. 5. Relationship between $X_{phlog.}$ and $F/(F+OH)$ ratios of biotites in metamorphic rocks from various districts. Solid circle: Biotites from metabasites in the Lützow-Holmbukta region. Open circle: Biotites from charnockites and paragneisses in the region. Biotites from the Lützow-Holmbukta region are enclosed by curves M and P, respectively. Open square: Biotites from Kondapalli granulites (LEELANANDAM, 1970). Small solid circle: Biotites from pelitic gneisses in the Komagane district, central Japan (unpublished data). The mg-values of biotites are regarded as $X_{phlog.}$. Each curve is calculated from the revised equations of LUDINGTON and MUNOZ (1975) for micas having intermediate Mg/Fe ratios in the following conditions. Curve 1: 680°, 5.5 kb (YOSHIDA, 1979) and $\log(f_{H_2O}/f_{HF})=4.3$. Curve 2: The same P-T conditions and log $(f_{H_2O}/f_{HF})=3.8$. Curve 3: 550°, 4 kb (P-T condition near sillimanite isograd of the Komagane district estimated by ONO (1977)), and $\log(f_{H_2O}/f_{HF})=4.5$.

It is considered that F replacing OH ion may play an important role during metamorphism in hydrous minerals such as biotite and hornblende. Fluorine-hydroxyl exchange between biotite and coexisting fluid phase has been investigated experimentally by MUNOZ and LUDINGTON (1974), and the f_{H_2O}/f_{HF} ratio of fluid phase coexisting with biotite can be estimated from the F content and the Mg/(Mg+Fe) ratio of a given biotite. Moreover, Mg-rich biotites can be more exchangeable of fluorine than Fe-rich ones in the same f_{H_2O}/f_{HF} ratio. The estimation of f_{H_2O}/f_{HF} of fluid phase under the condition of biotite formation of the district was carried out from the revised equations by LUDINGTON and MUNOZ (1975). According to YOSHIDA (1979), metamorphism of the Lützow-Holmbukta region can be divided into three stages and P–T conditions at the last stage were about 680°C and 5.5 kb. The relation between $X_{phlog.}$ (=mg value) and F/(F+OH) of biotites is shown in Fig. 5. It is obvious that the biotites are divided into two groups, *i. e.*, the one from metabasites and the other from charnockites and paragneisses, and the values of log (f_{H_2O}/f_{HF}) are 4.3 in the former and 3.8 in the latter, respectively. It is expected that fluorine may concentrate more into biotites of Mg-rich composition from metabasites than into those of Mg-poor ones from charnockite and paragneisses under the same f_{H_2O}/f_{HF} condition in all rocks, because F has a strong affinity for Mg than for Fe (RAMBERG, 1952; ROSENBERG and FOIT, 1977). However, the difference of f_{H_2O}/f_{HF} between metabasites and charnockites and paragneisses depends upon the difference of H_2O and F contents in the host rocks and upon that of mobility of H_2O and F during metamorphism. The value of f_{H_2O}/f_{HF} during metamorphism in paragneisses of the Lützow-Holmbukta region is smaller about one order than that in the Ryoke metamorphic rocks of the Komagane district, Japan (unpublished data) (Fig. 5). Moreover, charnockitic biotites from Kondapalli indicate far lower f_{H_2O}/f_{HF} ratios during metamorphic process than those in Lützow-Holmbukta, and $X_{phlog.}$ corresponds well to F/(F+OH).

Fluorine in amphiboles is also expected to have a strong affinity for Mg (CAMERON and GIBBS, 1973). The relation between the F content and the mg-value of hornblendes from various metamorphic terrains is shown in Fig. 6. From this figure, the F–Mg relation is not so clear, but hornblendes in each metamorphic terrain tend to have their respective ranges of the F content. Generally, hornblendes from the granulite facies rocks have higher F content than those from the amphibolite facies rocks, and those from the Lützow-Holmbukta district are similar to those from the hornblende-granulite facies rocks of the Colton area in the northwestern Adirondack Mountains (ENGEL and ENGEL, 1962). Typical granulites from Kondapalli and Madras have hornblendes richer in F than rocks from Lützow-Holmbukta and attain 1.5% of F. In the Adirondack Mountains, hornblendes of rocks from the Colton area belonging to the hornblende-granulite facies have higher F content than those from the Emeryville area of the amphibolite facies. It can be presumed from these facts, on the dehydration process of hydrous minerals during progressive metamor-

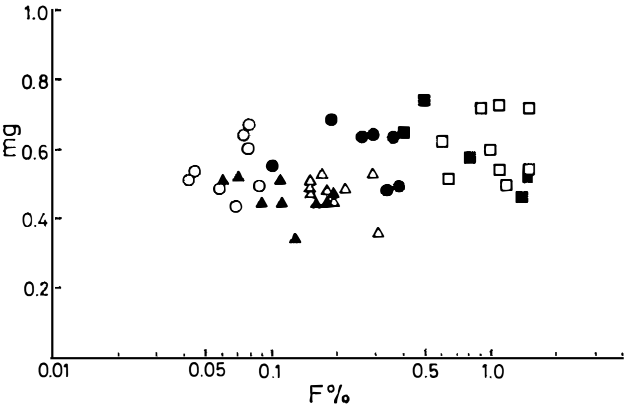

Fig. 6. *Relationship between the F content and the mg-value of hornblendes from various metamorphic terrains. Solid circle: Hornblendes from the Lützow-Holmbukta region. Solid square: Hornblendes from Madras* (HOWIE, *1955*). *Open square: Hornblendes from Kondapalli* (LEELANANDAM, *1970*). *Solid triangle and open triangle: Hornblendes from the Emeryville and Colton areas, respectively* (ENGEL *and* ENGEL, *1962*). *Open circle: Hornblendes from the Abukuma Plateau* (*unpublished data*).

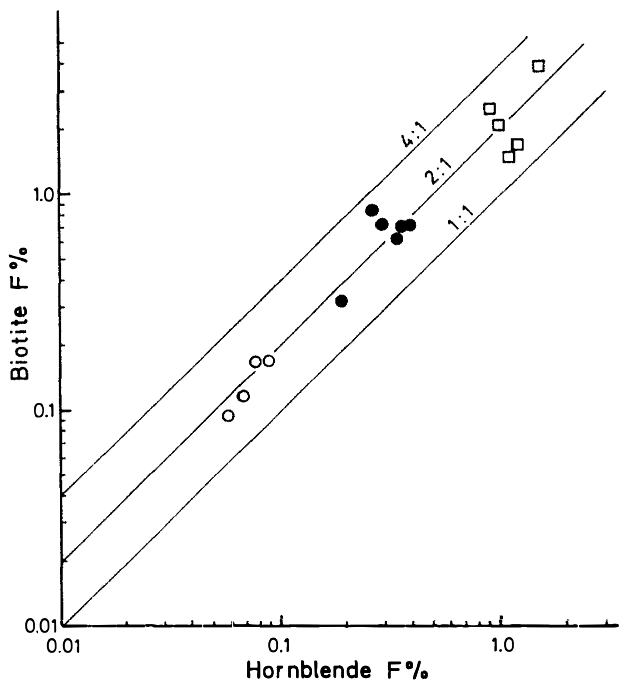

Fig. 7. *Distribution of F between coexisting biotites and hornblendes in metamorphic rocks. Symbols are the same as those in Fig. 6.*

phism, that F tends to remain selectively in crystal structure of hydrous minerals compared with hydroxyl ion, and concentrates into residual hydrous minerals such as biotite and hornblende. The fact that the replacement of OH by F in amphibolites significantly raises the upper stability limit (TROLL and GILBERT, 1972) is correlative with stable existence of F-rich hornblende in the granulite facies rocks.

EKSTRÖM (1972) found that the distribution coefficient of F for coexisting biotite and Ca-amphibole varies with increasing metamorphic grade and Ti content in Ca-amphibole, and that samples carrying hematite have normally a higher content of F in hydrous minerals due to the increasing activity of F during oxidation. The distribution of F between coexisting biotite and hornblende pairs from various metamorphic terrains studied here is all about 2: 1 by weight as shown in Fig. 7, and the distribution coefficient K_D, which is expresses as (F/OH bio)/(F/OH ho), is nearly 1, similar to the values for Japanese Cretaceous granites (KANISAWA, 1979; KANISAWA *et al.*, 1979), and does not show any difference by metamorphic grade or equilibrium temperature. Two pairs (Nos. 68091201-1 and 68091201-2) having slightly larger K_D values than the others are both low-Ti hornblende-low-Ti biotite pairs. More detailed data from various districts are required to clarify the distribution of F between biotite-hornblende pairs.

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References

BANNO, S., TATSUMI, T., KUNO, H. and KATSURA, T. (1964 a): Mineralogy of granulite facies rocks in the area around Lützow-Holm Bay, Antarctica. JARE Sci. Rep., Ser. C (Earth Sci.), **1**, 1-12.

BANNO, S., TATSUMI, T., OGURA, Y. and KATSURA, T. (1964 b): Petrographic studies on the rocks from the area around Lützow-Holmbukta. Antarctic Geology, ed. by R. J. ADIE, Amsterdam, North-Holland, 405-414.

CAMERON, M. and GIBBS, G. V. (1973): The crystal structure and bonding of fluor-tremolite; A comparison with hydroxyl tremolite. Am. Mineral., **58**, 879-888.

DEER, W. A., HOWIE, R. A. and ZUSSMAN, J. (1963): Rock Forming Minerals, **2**; Chain Silicates, and **3**; Sheet Silicates. London, Longmans, 379 p. and 270 p.

EKSTRÖM, T. K. (1972): The distribution of fluorine among some coexisting minerals. Contr. Mineral. Petrol., **34**, 192-200.

ENGEL, A. E. J. and ENGEL, C. G. (1962): Hornblendes formed during progressive metamorphism of amphibolites, Northwest Adirondack Mountains, New York. Geol. Soc. Am. Bull., **73**, 1499-1514.

FOSTER, M. D. (1960): Interpretation of the composition of trioctahedral micas. U. S. Geol. Surv., Prof. Pap., **354-B**, 11-49.

HOWIE, R. A. (1955): The geochemistry of the charnockite series of Madras, India. Trans. R. Soc. Edinburgh, **62**, 725–768.

KANISAWA, S. (1979): Content and behavior of fluorine in granitic rocks, Kitakami Mountains, northeast Japan. Chem. Geol., **24**, 57–67.

KANISAWA, S., TANAKA, H. and NAKAI, Y. (1979): Behavior of fluorine in granitic rocks from the Abukuma Plateau, the Ryoke Zone, and the San-in Zone, Japan. J. Geol. Soc. Jpn., **85**, 123–134.

LEELANANDAM, C. (1970): Chemical mineralogy of hornblendes and biotites from charnockitic rocks of Kondapalli, India. J. Petrol., **11**, 475–505.

LUDINGTON, S. and MUNOZ, J. L. (1975): Application of fluor-hydroxyl exchange data to natural micas. Geol. Soc. Am. Program Abstr., **7**, 1179.

MUNOZ, J. L. and LUDINGTON, S. (1974): Fluoride-hydroxyl exchange in biotite. Am. J. Sci., **274**, 396–413.

ONO, A. (1977): Takato-Shiojiri chihô no Ryoke henseigan no gansekigaku-teki kenkyû (Petrologic study of the Ryoke metamorphic rocks in the Takato-Shiojiri area, central Japan). Ganseki Kôbutsu Kôshô Gakkai-shi (J. Jpn. Assoc. Mineral. Petrol. Econ. Geol.), **72**, 453–468.

RAASE, P. (1974): Al and Ti contents of hornblende, indicators of pressure and temperature of regional metamorphism. Contr. Mineral. Petrol., **45**, 231–236.

RAMBERG, H. (1952): Chemical bonds and distribution of cations in silicates. J. Geol., **60**, 331–355.

ROSENBERG, P. E. and FOIT, F. F., Jr. (1977): Fe^{2+}-F avoidance in silicates. Geochim. Cosmochim. Acta, **41**, 345–346.

SUWA, K. (1966): On plagioclases in metamorphic rocks from Lützow-Holmbukta area, East Antarctica. Proc. Jpn. Acad., **42**, 1175–1180.

TROLL, G. and GILBERT, M. C. (1972): Fluorine-hydroxyl substitution in tremolite. Am. Mineral., **57**, 1386–1403.

YOSHIDA, M. (1979): Metamorphic conditions of the polymetamorphic Lützow-Holmbukta region, East Antarctica. J. Geosci., Osaka City Univ., **22**, 97–139.

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