ON THE CHEMISTRY ON COEXISTING GARNET AND BIOTITE IN PELITIC-PSAMMITIC METAMORPHIC ROCKS, CENTRAL ABUKUMA, JAPAN

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I. Introduction

Since the later 19th century, a number of Japanese geologists as KOTO (1893), SUGI (1935), HORAI (1944), and MIYASHIRO (1958) have engaged with the petrological studies in the Gozaisho-Takanuki district of central Abukuma. Among them the works of the last author (MIYASHIRO, 1958 and 1961) have made famous the name of Abukuma over the world. According to him, the Gozaisho-Takanuki metamorphic rocks were derived from the Upper Paleozoic and were metamorphosed under high temperature and low pressure accompanied by the granitic intrusion of Cretaceous time. Based on some geological and petrological evidences (KURODA, 1963; MINATO *et al.*, 1965; KANO and KURODA, 1968; RESEARCH GROUP OF ABUKUMA PLATEAU, 1969; KURODA and KANO, 1970), we have proposed to make a fundamental correction of MIYASHIRO's scheme. The metamorphic rocks, at least a part of the so-called Takanuki gneiss, may not be derivatives from the Upper Paleozoic but probably from the pre-Cambrian. The metamorphic history may consist of at least two episodes, the older and the younger, of which the former may be of the *Dalradian* type characterized by the occurrence of kyanite and staurolite in pre-Cambrian or early Paleozoic and the latter may be of the regional contact type associated with vast intrusion of granitic rocks in late Mesozoic.

On the other hand, MIYASHIRO (1953 and 1958) stated an important fact and made a valuable suggestion on the chemistry of coexisting garnet and biotite. His emphasis of statement was put on the following points.

1) With the increase of metamorphic grade, the composition of garnet changes from spessartinous to almandinous and the composition of

biotite becomes richer in Mg, though the bulk compositions of their host rocks are the same. This is an earlier contribution to the partition problem on coexisting minerals and was indeed an excellent idea at that time.

2) The Mn content of garnet in metamorphic pelitic-psammitic rocks depends on the character and grade of metamorphism. Under the low pressure condition it is high in lower grade part, while under the high pressure condition it is low even in lower grade part. In both cases, of course, it becomes lower with the increase of metamorphic grade.

After that the Chemistry of coexisting garnet and biotite in pelitic metamorphic rocks has been investigated from the standpoint of partition of Mg and Fe by many authors (FROST, 1962; PHINNEY, 1963; ALBEE, 1965; MATEJOVSKA, 1970; PERCHUK, 1970, *etc.*). All of these works show that the partition coefficient of Mg and Fe between garnet and biotite is an indicator of metamorphic grade, especially of temperature. Recent development in analytical instrument as the electron microprobe X-ray microanalyser has brought us many valuable informations on the chemistry of garnet, especially on its zonation. For example, the recent papers by EDMUNDS and ATHERTON (1971) and MÜLLER and SCHNEIDER (1971) are very interesting and instructive for our study.

In this study a number of coexisting garnet and biotite were investigated by means of an EPMA. The results will present a new positive evidence for the theory of polymetamorphism in the Abukuma Plateau which we and our collaborators have proposed for years.

II. General Geology and Petrography

A full explanation on geology and geologic structure of this district is given in *Geology of the Takanuki district* (Quadrangle series, scale 1 : 50 000) published by the Geological Survey of Japan (KANO, *et*

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Fig. 1. Localities of the studied specimens.

1 Gozaisho-Takanuki metamorphic rocks, 2 plutonic rocks (S: Samegawa mass, Is: Ishikawa mass, J: Jumonji mass, M: Miyamoto mass, R: Ronden mass, Y: Yoshimagawa mass, Ir: Iritono mass, T: Tabito mass and H: Hanawa mass), 3 Tertiary and Quaternary systems, 4 specimens from Area II (Nos. 40—82), 5 specimens from Area I (Nos. 1—24), 6 specimens from Area I' (Nos. 31—34), 7 boundary between Areas, 8 Gozaisho high way, 9 prefectural boundary between Fukushima and Ibaragi.

al., 1973). Therefore only an outline of metamorphic geology and petrology is described in this paper in order to help the understanding of the description on the chemistry of the coexisting garnet and biotite.

As shown in Fig. 1, geology of the present district is roughly divided into two groups, metamorphics and plutonics. Several plutons of granitic composition occur in this district, among which the Ishikawa, Samegawa, Miyamoto, Yoshimagawa, Iritono, and Tabito masses distribute arranging in this order from the west to the east of the metamorphic terrain. The Samegawa mass makes a

composite pluton composed of quartz-diorite to adamellite surrounded by migmatitic shell. Structurally the mass is concordant with the surrounding metamorphic rocks and is situated in the core of a large dome structure. The Miyamoto mass forms also a typical composite pluton consisting of adamellite, granodiorite, diorite, and gabbro. It is injected into the country rocks with generally concordant and locally forceful relation. The Ishikawa mass is a concordant pluton but it is not a distinct composite pluton. The other masses are more or less discordant plutons commonly associated with narrow but distinct contact aureoles, and are considered to be younger in age than the former three plutons.

Though the true ages of these plutons are still problematical, K-Ar dating of micas in main plutons shows 90—100 m.y. (KAWANO and UEDA, 1965). On the other hand, Rb-Sr isochron ages measured recently by MARUYAMA (1972) are 350—400 m.y. for the Ishikawa, Samegawa, and Miyamoto masses, and 150 m.y. for the Iritono and Tabito masses.

On the basis of the axial color of amphibole and the mineral assemblage, MIYASHIRO (1958) divided the metamorphic belt into three zones, A, B, and C. Zone A corresponds to greenschist facies, Zone B and C together to amphibolite facies. He emphasized the absence of metamorphic rocks of epidote-amphibolite facies. Differing from MIYASHIRO's scheme, we divided into two Areas, I and II, from the viewpoint of the zonation pattern of garnet as well as the structural behavior of metamorphic rocks. Generally speaking, Area I occupies the eastern half of the district which is composed of the major part of the Gozaisho metamorphic rocks; Area II does the western half including the Takanuki metamorphic rocks and a minor part of the Gozaisho metamorphic rocks.

The boundary line between Area I and Area II runs north-northwestwards along the eastern to northeastern side of the Miyamoto mass, and lies near the fault from Babadaira to Kaiya. It is also noted that minor intrusives of ultrabasic rocks are found sporadically along this boundary line.

Area I Among the metamorphic rocks of this area, basic rocks are dominant and pelitic-psammitic or siliceous rocks are subordinate. Structurally they are characterized with angular or *chevron* folds of which wave-length are 2—3 km, and the fold axes trend NNW to SSE plunging to either north or south with a low angle.

The grain size of recrystallized minerals is generally small. Equigranular quartz and feldspar are 0.02—0.04 mm, and elongated mica and amphibole are about 0.08—0.10 mm in length.

Garnet idioblast is 0.05—0.4 mm in diameter. It is noteworthy that there are observed various mineral assemblages of basic metamorphic rocks. The first is green hornblende+andesine. In this case some hornblendes have pale-colored cores and green-colored shells, and some plagioclase grains show reverse zoning with the cores of slightly lower An content than the shells. This is the standard assemblage of amphibolite facies. The second is green hornblende+albite or oligoclase (+epidote+chlorite), probably belonging to epidote-amphibolite facies. The third is actinolite+chlorite+albite of greenschist facies. However it is difficult to draw boundaries between these three facies areas because of the complicated geologic structure. Generally speaking the rocks of the amphibolite facies assemblage are the most widely distributed in this area.

The mineral assemblage of pelitic-psammitic metamorphic rocks is almost constantly biotite+muscovite + quartz + plagioclase ($\pm$ garnet) + opaque minerals. Garnet occurs in biotite-rich pelitic schist but more abundantly in biotite-poor quartzose schist. Chlorite is found in many specimens, though it is not distinct whether it is of primary or retrogressive in origin. Opaque minerals are chiefly manganiferous ilmenite or magnetite, sometimes accompanying pyrite and hematite. As is described later, all the garnets exhibit the normal type of zonation, that is, the core is richer in MnO and poorer in FeO than the rim.

Area II This area consists of mainly pelitic-psammitic and siliceous gneisses with minor amount of amphibolite, metamorphic peridotite and marble. Staurolite bearing rock with peculiar chemical composition (URUNO and KANISAWA, 1965) occurs in a narrow horizon in the vicinity of Takanuki. Structurally the rocks of Area II form a gently undulated open fold around the Samegawa plutonic mass, and the fold axis trends largely parallel to that of Area I. The stratigraphic relation between the rocks of Area I and Area II is not always conclusive but the latter may be overlain by the former without discontinuity.

The grain size is remarkably coarser than Area I. Equigranular minerals are 0.08—0.2 mm and elongated ones are 0.2—0.35 mm in length. Garnet crystals measure 0.2—3.0 mm in diameter. Thus the difference of grain size between Area I and Area II is fairly critical.

The main assemblage of basic metamorphic rocks is hornblende+andesine. Hornblende is green or brown, and occasionally bluish green variety. Plagioclase contains commonly 40—45% An and shows weak reverse zoning. Besides cum-

ingtonite and diopside are often found. Almandinous garnet-amphibolite and two pyroxene-amphibolite occur rarely.

The pelitic-psammitic metamorphic rocks are chiefly composed of muscovite, biotite, garnet, plagioclase, potassium feldspar and quartz. Among opaque minerals graphite and/or magnetite are most abundant, and the others are ilmenite, rutile, pyrite, chalcopyrite, pyrrhotite *etc.* Sillimanite occurs commonly, particularly around the Samegawa plutonic mass. Occurrence of andalusite and cordierite is limited only in a comparatively thin bed. As previously reported, kyanite occurs in normal pelitic gneiss as a relic mineral (KANO and KUODA, 1968). Zonation of garnet shows generally the reverse type contrary to that of Area I, that is, MnO is increased in the rim and decreased in the core and FeO *vice versa*.

In addition, contact metamorphic rocks are found in narrow aureoles around the Yoshimagawa and Iritono plutonic masses. The aureole is 500—1000 m in width at the contact of the Yoshimagawa mass and several 10 m around the Iritono mass. The contact rocks are apparently polymetamorphic hornfels derived from the rocks of Area I. Their mineral assemblage and the pattern of garnet zonation are the same as those of Area II. Therefore the aureole should be separated from Area I and defined as Area I'. The boundary between Area I and Area I' is gradual and transitional in a short distance.

III. Mineralogy and Chemistry of Host Rocks

In the Gozaisho-Takanuki district, metamorphic rocks including pyralspite garnet occur abundantly. The localities and mineral assemblages of the rocks of which garnets and biotites were studied in this study are shown in Table 1 and Fig. 1. There are remarkable differences in mineralogy between the rocks from Area I and those from Area II. In Area I the common opaque mineral is ilmenite with remarkable amount of MnO. Chlorite is fairly abundant. Orthoclase is generally absent with an exception of No. 9 in which K₂O amounts 4.07%. In Area II the opaque minerals are commonly magnetite and graphite. Ilmenite occurs rarely, but it is never manganiferous (Table 2). Sillimanite and orthoclase are common especially in the higher grade rocks.

Among the garnet bearing metamorphic rocks the chemical compositions of 9 specimens from Area I, 2 specimens from Area I' and 17 specimens from Area II were analysed and listed in Table 3. These are plotted together on the AKF diagram

Table 1. Sample number, locality and mineral assemblage of the host rock of garnet.

Area I

No.	Sample no.	Locality	Mineral assemblage
1	Y O700410	Sawatari mine	ga-bi-pl-qt-ilm-mg-py
2	HM66101405	Shinden	ga-mu-bi-chl-pl-qt-ilm
3	Y O69073002	"	ga-bi-chl-pl-qt-ilm
4	Y O69080301	Kumakura	ga-bi-pl-qt-ilm
5	HK69081906	"	ga-mu-bi-pl-qt-mg
6	HK69081904	"	ga-bi-mu-pl-qt (quartz-porphyry)
7	HK65073003	Tairakuchi	ga-bi-pl-qt-ilm
8	HK65073008	"	ga-bi-qt-pl-ilm
9	HK65072711	Gozaisho	ga-chl-mu-bi-pl-or-qt-mg
10	HK65072713	"	ga-chl-bi-pl-qt-ilm
11	Y K65-11	"	ga-chl-bi-pl-qt-mg
12	HK72041305	"	ga-bi-chl-pl-qt-ilm-mg
13	K I 67090807	Idosawa	ga-bi-chl-qt-pl-ilm
14	TM72041005	Kamiyama	ga-mu-bi-pl-qt-ilm
15	TM72041008	"	ga-bi-pl-qt-ilm
16	TM72041009	"	ga-mu-bi-pl-qt-ilm
17	U. T.-5	Nakanosawa	ga-mu-bi-pl-qt
18	HK72041006	Kaidomari-Tokusa	ga-mu-bi-pl-qt
19	K I 67091703	"	ga-mu-bi-pl-qt
20	TM72041412	Nabana	ga-mu-bi-ql-qt-ilm
21	TM72041418	"	ga-mu-bi-pl-qt-ilm
22	HK72041014	Yazukuri	ga-mu-bi-pl-qt-ilm
23	HK72041412	Takabo	ga-act-qt-hm
24	TM660728-132	Abuki	st-ad-ga-mu-bi-chl-pl-qt-mg-gr-tm

Area I'

31	Y O69091602	Horinouchi	ga-mu-bi-pl-qt-mg-gr
32	Y O69090103	Osogawa	ga-bi-pl-qt
33	Y O69090301	Shuku	ga-bi-pl-qt-gr
34	Y O69110403	Takano	ga-bi-pl-or-pt

Area II

40	U-N. 72041315	Kaiya	ga-bi-pl-qt-ilm-py
41	Y O69080903	Koshidai	ga-mu-bi-qt-pl-ilm-mt
42	Y O69082501	Kaiya-N	ga-mu-bi-pl-or-qt-mg-tm
43	HM66101104	Ishizumi	ga-mu-bi-pl-qt
44	HM66102308	Nita	ga-mu-bi-pl-qt-ilm-py-gr
45	HM66102309	"	ga-mu-bi-pl-qt
46	HK67072102	Kaidomari	ga-ho-bi-pl-qt
47	K I 67092602	Oinudaira	si-ga-co-mu-bi-pl-qt-gr
48	K I 670925 C2	"	ga-bi-pl-qt-gr
49	Y O700502	Hanazono	si-ga-co-mu-bi-pl-or-qt-gr
50	A F71092406	Hanawa	ga-co-mu-bi-pl-or-qt-gr
51	A F71092304	"	ga-co-mu-bi-pl-qt
52	MG014	Ide	si-ga-bi-pl-qt-or-gr
53	TM65072917 a	Nyudô	ga-bi-pl-or-qt-ilm
54	TM65072917 b	"	ga-bi-mu-pl-qt-gr
55	TM65072919	"	ga-mu-bi-pl-or-qt-rt-py
56	HM66102306	"	ga-bi-pl-qt-mg-gr
57	TM7112206	Onasawa	ad-si-ga-co-mu-bi-pl-or-qt-mg-gr
58	HM66102301	Huruuchi	si-ga-mu-bi-pl-or-qt
59	HM66103107	Baba	si-co-ga-mu-bi-pl-or-qt-gr

60	HM66082005	Daibara	si-co-ga-mu-bi-pl-or-qt-gr
61	HM66082002 a	Mappo	ga-mu-bi-pl-qt
62	HM66082002 b	"	ga-mu-bi-pl-qt
63	HM66082003	"	ga-mu-bi-pl-qt-gr
64	HM66100703	Maegi	ga-mu-bi-chl-pl-or-qt-mg
65	HM66090306	Yokogawa	si-ga-sp-mu-bi-mg
66	HM66100407	Furudono	ga-mu-chl-bi-pl-qt-py
67	HM66090309	Furudono-Tokusa	ga-mu-bi-pl-or-qt-mg
68	TM72041401	Hônosawa	ga-mu-chl-bi-pl-or-qt-tm
69	TM72041404	"	si-ga-mu-bi-pl-or-qt
70	HK 72041012	Uchide	ga-mu-bi-pl-or-qt
71	TM67072613	Sakamichi	ga-mu-bi-pl-qt-gr
72	KN67072401	Yachi	si-ga-mu-bi-pl-qt-pyrr-gr
73	TM67072231	Yanoyama	ga-mu-bi-pl-qt-gr
74	TM67072210	"	ga-mu-bi-qt-mg-gr
75	TM67072203	Miyamae	ga-mu-bi-pl-qt-gr-mg
76	HM66090302	Sakune	ga-mu-bi-qt-pl-or-cp
77	HM66090303	Miyamae	ga-bi-mu-qt-pl-or-gr
78	HM66090304	"	ga-mu-bi-pl-qt-gr
79	TM65090410	Onikoshi	ga-bi-mu-qt-qy
80	TM65083165	Kuwakida	si-co-ga-mu-bi-pl-or-gr
81	TM65080508	Eryûda	cum-ho-ga-bi-pl-qt
82	TM65081015	Watarase	si-ga-mu-bi-pl-qt-or-gr

Collectors : AF, Akio FURUKAWA, HK, Hiroshi KANO, HM, Hiroshi MITSUKAWA, KI, Kiyoshi INUMA, TM, Takahiko MARUYAMA, TN, Terukazu NUREKI, YK, Yoshimasu KURODA, YO, Yoshihisa OHIRA, U.N., Hayao UMEMURA and Takeo NAKAMURA, U. T., Katsutoshi URUNO and Takashi TASAKA, MG, Masao GORAI.

Constituent minerals : st staurolite, si sillimanite, ad andalusite, co cordierite, sp spinel, ga garnet, act actinolite, cum cummingtonite, ho hornblende, mu muscovite, bi biotite, chl chlorite, pl plagioclase, or orthoclase, qt quartz, ilm ilmenite, mg magnetite, hm hematite, py pyrite, pyrr pyrrhotite, cp chalcopyrite, rt rutile, gr graphite, tm tourmaline.

Table 2. Chemical compositions of ilmenite.

No.	Locality	TiO ₂	FeO	MnO
1	Sawatari (Area I)	51.4	44.6	4.0
2	Shinden (")	56.4	40.8	2.8
3	" (")	58.6	37.8	3.6
6	Kumakura (")	52.2	37.8	10.0
7	Tairaguchi (")	57.3	37.0	5.7
10	Gozaisho (")	52.3	45.7	1.8
12	" (")	50.2	45.6	4.2
14	Kamiyama (")	51.1	39.9	9.0
16	" (")	49.9	43.3	6.8
19	Nabana (")	51.0	36.1	12.9
20	" (")	51.1	41.6	7.3
40	Kaiya (Area II)	53.8	42.0	4.2
44	Nita (")	57.4	38.8	3.8
70	Nyûdô (")	55.2	44.0	0.8

Note : Numbers are the same as those in Table 1.

(Fig. 2) in which the composition field of almandine and biotite were determined by the present data of this paper. It is noted that the majority of plots are included in the triangle of muscovite-biotite-almandine. It may be said that the rocks in Area II are generally rich in K component than those in Area I. The averages of K₂O are 1.91% and 2.66% in Area I and Area II respectively.

The whole rock compositions are also plotted on Mn-Mg-Fe triangle (Fig. 3), in which the rocks of Area I and Area II can be largely divided into separated groups. The rocks of Area II except No. 53 are richer in Mg and poorer in Mn than those in Area I. As will be stated later, this is quite conformable to the characteristics of biotites in both Areas.

With respect to the common presence of graphite in Area II, it is remarked that the oxidation ratio, $\text{Fe}^{+3}/\text{Fe}^{+2} + \text{Fe}^{+3}$, is a little lower in Area II than in Area I, though the difference is rather less distinct. This ratio is 0.20—0.56 (mean 0.35) in Area I and 0.20—0.44 (mean 0.29) in Area II.

IV. Microprobe Analysis

Microprobe X-ray analyses were performed on a JEOL X-ray microanalyser (JXA-5) having an X-ray take-off angle of 40° in the Institute of Mining College, Akita University, Akita. Operating conditions were 15 kv accelerating voltage, 0.1 to 0.05 microamperes sample current and about 3 microns spot size. Because of the zonal variation in composition always seen in garnets, the ten-second step-scanning analysis was run at intervals of 10 to 100 microns on a traverse. Then

Table 3. Chemical analyses of the host rocks of the studied garnets and biotites.

	Area I									Area I'	
	5	7	9	10	13	20	21	22	24	31	33
SiO ₂	71.48	69.78	60.36	60.82	62.34	71.22	73.14	69.84	62.96	75.56	74.82
TiO ₂	0.33	0.38	0.30	1.03	1.29	0.30	0.10	0.51	0.74	0.32	0.38
Al ₂ O ₃	12.93	13.01	19.90	17.32	16.41	14.56	14.03	13.71	18.08	11.09	10.86
Fe ₂ O ₃	2.24	1.69	3.75	3.27	1.89	1.76	1.34	1.91	1.28	1.18	1.44
FeO	3.81	3.09	2.62	5.10	5.49	2.43	1.66	3.17	4.61	2.87	2.80
MnO	0.42	0.40	0.15	0.26	0.29	0.07	0.22	0.06	0.13	0.10	0.10
MgO	1.99	1.97	0.53	2.43	2.32	1.39	1.16	1.92	2.51	1.97	2.55
CaO	1.59	1.93	1.86	3.76	2.54	1.57	1.66	2.19	0.96	1.63	2.83
Na ₂ O	3.02	4.55	4.90	3.21	4.70	4.12	4.20	3.57	2.01	1.91	1.91
K ₂ O	1.72	1.13	4.07	1.27	0.85	2.13	1.93	2.07	2.76	2.17	1.33
H ₂ O(+)	0.83	1.15	0.48	1.43	1.65	0.81	0.76	0.94	3.40	1.23	0.76
H ₂ O(-)	0.11	0.36	0.71	0.17	0.22	0.10	0.11	0.08	0.26	0.23	0.25
P ₂ O ₅	0.04	0.42	0.13	0.05	0.07	0.03	0.06	0.02	0.10	0.03	0.05
Total	100.51	99.86	99.76	100.12	100.06	100.49	100.37	99.99	99.80	100.29	100.08
Fe ³⁺											
Fe ²⁺ + Fe ³⁺	0.34	0.34	0.56	0.36	0.24	0.39	0.41	0.35	0.20	0.26	0.32

	Area II							
	41	42	43	47	53	54	57	59
SiO ₂	69.68	72.24	68.60	62.01	52.32	65.70	60.35	69.30
TiO ₂	0.51	0.48	0.56	0.53	2.77	0.99	0.60	0.20
Al ₂ O ₃	14.23	13.14	13.92	17.54	17.36	15.02	17.36	15.02
Fe ₂ O ₃	1.73	0.97	2.58	3.01	5.79	1.95	1.69	1.11
FeO	2.62	3.05	2.91	4.04	7.54	5.03	4.87	4.09
MnO	0.09	0.10	0.05	0.11	0.28	0.04	0.19	0.05
MgO	1.52	1.76	2.03	3.31	2.77	2.39	3.14	2.53
CaO	2.62	1.68	2.89	1.54	5.28	2.62	4.35	1.18
K ₂ O	4.11	3.61	3.02	1.90	1.91	3.32	2.82	2.21
Na ₂ O	1.68	2.62	1.90	2.06	2.40	2.31	2.72	3.12
H ₂ O(+)	1.08	0.64	1.33	2.59	0.62	0.84	1.38	1.29
H ₂ O(-)	0.19	0.01	0.41	0.79	0.12	0.14	0.06	0.14
P ₂ O ₅	0.08	0.10	0.03	0.01	0.05	0.05	0.08	0.02
Total	100.14	100.40	100.23	99.44	99.21	100.40	99.61	99.55
Fe ³⁺								
Fe ²⁺ + Fe ³⁺	0.38	0.22	0.44	0.40	0.23	0.26	0.24	0.20

	Area II								
	60	66	67	68	69	70	72	76	78
SiO ₂	54.20	71.10	74.66	73.48	62.48	69.38	69.67	75.10	68.80
TiO ₂	0.96	0.33	0.24	0.06	0.59	0.11	0.61	0.27	0.69
Al ₂ O ₃	19.62	13.09	12.54	12.34	18.19	14.12	15.66	12.04	14.85
Fe ₂ O ₃	2.54	1.13	0.83	1.37	1.74	1.18	1.91	1.06	2.37
FeO	5.13	3.84	2.51	2.87	4.20	3.61	2.67	2.51	3.27
MnO	0.07	tr	0.07	0.10	0.02	0.04	0.21	0.05	0.26
MgO	2.77	2.51	1.53	1.79	2.26	2.32	2.25	1.71	2.01
CaO	2.68	3.11	1.51	1.05	1.38	1.98	0.53	1.10	1.61
K ₂ O	4.31	2.27	2.72	2.72	2.82	3.32	1.39	2.62	3.02
Na ₂ O	3.50	2.17	2.31	3.24	4.39	3.53	2.36	2.82	2.04
H ₂ O(+)	3.54	0.65	0.80	0.43	2.13	0.50	0.02	0.87	1.42
H ₂ O(-)	0.60	0.10	0.13	0.02	0.10	0.04	0.40	0.11	0.09
P ₂ O ₅	0.12	0.07	0.06	0.03	0.05	0.04	0.05	0.04	0.04
Total	100.04	100.37	99.91	99.50	100.35	100.17	99.73	100.30	100.47
Fe ³⁺									
Fe ²⁺ + Fe ³⁺	0.31	0.21	0.22	0.31	0.28	0.22	0.39	0.29	0.40

Fig. 2. Plots of the compositions of host rocks on AKF diagram.

the like procedure was carried on another individual in the same section. For the analyses of biotites the similar method was used.

As standards for the quantitative calculation mainly a suit of natural or synthetic oxides and silicates such as SiO_2 , TiO_2 , Al_2O_3 , Fe_2O_3 , MnO , MgO , CaSiO_3 , $\text{NaAlSi}_3\text{O}_8$, and KAlSi_3O_8 and, for reference, the Anamushi garnet (KANO and HIRANO, 1970) were used and the correction procedure was made after the method introduced by BENCE and ALBEE (1968) with the recent revision by ALBEE and RAY (1970), though no correction was needed when the Anamushi garnet was used as standard for the analysis of garnet. The analytical error in total sum which is largely due to re-

latively high deviation of Si and Al is about $\pm 2\%$.

V. Zonation and Composition of Garnet

Chemical compositions of analysed garnets, 24 from Area I, 4 from Area I' and 43 from Area II, are listed in Tables 4, 5, and 6.

Under the microprobe scanning, all the garnets exhibit pronounced zonation, though its degree and pattern are different from Area I to Area II. Those from Area I generally show great difference in composition between the center and the margin, and their zonation patterns are characterized by the decrease of MnO and the complementary increase of FeO outward from the center. As a consequence the MnO profile takes an inverted "V"-like or "bell"-like shape (Fig. 4). On the other hand MgO increases towards the outside, though its variation is only small. The behavior of CaO is somewhat complicated, but it shows almost the same pattern as MnO . Such pattern of zonation is named the normal type or normal zoning.

Since BANNO (1965) first described a zoned pyroxene-garnet of the Sanbagawa schist from Besshi of Japan, similar zoned garnets of the normal type have been reported from a number of metamorphic terrains in the world, *e.g.* the Kwoick area of British Columbia (HOLLISTER, 1966), Otago of New Zealand (BROWN, 1967), the Tibulon Peninsula of the Franciscan, California (DUDLEY, 1969), Perthshire area of the Dalradian, Scotland (ATHERTON, 1968), the northern area of the Idaho batholith (HIETANEN, 1969), *etc.*

In Japan, according to our probe investigations (KANO, 1971), garnets from Sazare and Shirataki

Fig. 3. Plots of the compositions of host rocks on Mn-Mg-Fe triangle.

($+\text{Fe}_2\text{O}_3$) : Fe_2O_3 is calculated as FeO and added to FeO . ($-\text{Fe}_2\text{O}_3$) : Fe_2O_3 is not added to FeO .

Table 4. Microprobe analyses of garnets in Area I.

		1	2	3	4	5	6	7	8	9	10	11	12
	SiO ₂	37.54	34.81	37.39	37.69	38.41	37.75	38.86	36.21	38.13	36.39	34.67	39.24
	TiO ₂	0.11	—	0.14	0.26	0.15	0.16	0.22	—	0.11	—	—	0.24
	Al ₂ O ₃	21.07	21.38	21.10	20.46	21.02	21.03	21.95	21.69	21.84	21.50	21.30	21.80
	FeO	20.84	28.37	24.99	27.29	19.39	13.49	9.64	19.77	21.38	30.95	22.43	24.68
	MnO	14.51	13.95	11.96	10.74	15.37	23.62	20.04	19.21	17.29	8.37	20.36	12.25
	MgO	0.90	1.70	1.20	1.37	0.77	3.31	0.13	0.84	0.65	2.09	0.80	1.48
	CaO	3.53	0.66	1.36	3.46	4.51	2.19	9.52	1.22	1.11	1.52	1.77	2.06
	K ₂ O	—	—	—	—	—	—	—	—	—	—	—	0.04
	Total	98.53	100.89	98.15	101.28	99.63	101.54	100.35	98.93	100.53	100.82	100.53	101.78
center	MnO	15.9	15.7	12.6	13.6	19.3	27.1	24.0	19.5	19.0	11.8	21.9	15.1
	FeO	19.7	26.8	24.1	24.3	15.1	10.9	7.0	19.0	20.1	29.6	21.8	21.8
	MgO	0.9	1.7	1.3	1.1	0.4	3.6	0.1	1.0	0.7	1.8	0.7	1.2
margin	MnO	13.4	12.4	11.2	9.6	8.6	19.3	16.0	18.8	17.1	5.3	19.3	6.7
	FeO	21.3	29.6	25.3	30.6	29.5	15.7	17.4	20.6	22.2	33.2	23.3	29.3
	MgO	0.9	1.8	1.1	1.6	1.6	3.9	0.2	0.9	0.7	2.3	0.8	1.8

		13	14	15	16	17	18	19	20	21	22	23	24
	SiO ₂	36.52	38.17	39.03	36.14	37.23	38.47	37.15	38.42	39.17	38.96	39.95	35.46
	TiO ₂	—	0.21	—	0.21	0.14	0.02	—	0.28	0.14	0.08	0.12	—
	Al ₂ O ₃	20.69	21.09	20.09	21.11	21.03	21.46	22.15	20.89	21.61	22.06	21.08	22.81
	FeO	22.17	18.61	18.62	23.14	25.15	24.32	19.59	19.73	19.66	21.64	11.16	31.57
	MnO	15.69	16.60	13.33	14.33	12.34	11.38	12.56	15.62	14.98	12.91	20.41	7.84
	MgO	1.10	1.41	1.01	1.47	2.21	2.17	1.50	0.99	1.43	1.40	0.24	1.80
	CaO	2.50	3.23	6.91	3.63	2.75	1.43	6.50	4.82	2.98	3.21	8.71	1.22
	K ₂ O	—	0.04	—	0.04	0.05	0.02	—	0.04	0.00	—	0.04	—
	Total	98.67	99.38	98.99	100.08	100.89	99.26	99.45	100.79	99.95	100.24	101.71	100.70
center	MnO	18.2	15.5	17.4	15.0	13.6	13.4	13.4	19.1	19.4	15.9	23.2	8.8
	FeO	21.6	16.0	14.6	21.4	23.4	22.3	16.9	14.8	13.3	18.1	9.0	32.1
	MgO	1.1	1.2	0.6	1.2	1.8	1.8	1.0	0.6	0.7	0.9	0.2	1.9
margin	MnO	14.5	17.6	11.0	13.4	12.3	9.3	12.3	11.8	12.3	11.0	16.1	6.8
	FeO	23.2	22.0	20.1	24.9	26.6	26.2	22.6	26.0	24.0	26.3	14.1	34.3
	MgO	1.2	1.8	1.2	1.5	2.3	2.8	2.0	1.6	1.9	1.9	0.5	1.8

Table 5. Microprobe analyses of garnets in Area I'.

		31	32	33	34
	SiO ₂	34.23	38.59	35.44	38.27
	TiO ₂	—	0.07	—	0.04
	Al ₂ O ₃	21.56	20.64	21.04	21.38
	FeO	27.86	24.68	23.81	28.34
	MnO	12.62	8.91	12.68	6.48
	MgO	2.97	2.75	4.47	3.12
	CaO	1.00	0.97	2.76	1.38
	K ₂ O	—	—	—	0.02
	Total	100.24	96.61	100.20	99.04
center	MnO	11.5	7.2	12.8	5.2
	FeO	28.2	26.2	23.8	28.8
	MgO	3.3	3.1	4.5	3.3
margin	MnO	13.5	12.6	12.5	8.4
	FeO	27.6	22.8	24.2	27.2
	MgO	2.8	2.2	4.4	2.4

areas of the Sanbagawa belt, Nishiki area of the Sangun belt, and Unazuki area of the Hida belt are proved to be of the normal type of zonation. In the Ryoike belt, garnets from schistose hornfels also show the normal zonation. Here it must be noted that all the garnets mentioned above including those from Area I of Abukuma occur from the low to medium grade regional metamorphic rocks subjected with single cycle progressive metamorphism. The characteristic "bell"-shaped profile of MnO observed in individual grain can be explained in terms of a simple segregation model or by the effect of the Rayleigh fractionation advanced by HOLLISTER (1966) and ATHERTON (1968).

To the contrary all the garnets from Area II show rather small compositional difference between the center and the margin and the zonation

Table 6. Microprobe analyses of garnets in Area I.

	40	41	42	43	44	45	46	47	48	49	50	51	52	53
SiO ₂	39.26	39.18	36.22	35.24	35.30	36.60	38.22	37.48	39.32	39.69	38.97	38.80	35.60	37.94
TiO ₂	0.11	0.12	—	—	—	—	—	—	0.04	0.03	—	—	—	0.12
Al ₂ O ₃	20.91	23.21	21.36	21.93	21.86	21.45	22.35	25.09	33.51	21.83	21.06	22.38	22.07	21.69
FeO	27.34	25.26	23.61	20.73	25.37	24.35	22.73	30.50	23.06	31.23	29.40	28.70	30.27	31.08
MnO	6.98	10.36	15.90	14.12	14.09	14.65	9.04	3.12	10.19	3.94	7.39	6.09	6.97	1.54
MgO	2.63	1.63	2.74	2.09	3.00	2.15	2.83	3.93	3.39	3.36	2.47	2.24	3.17	2.50
CaO	2.11	0.91	1.55	6.38	0.84	0.77	5.29	0.94	1.05	1.17	1.36	1.04	0.99	4.45
K ₂ O	—	—	—	—	—	—	—	—	—	—	—	—	—	0.04
Total	99.33	100.68	101.38	100.49	100.46	99.96	100.46	101.06	100.56	101.26	100.65	99.25	99.07	99.35
center	MnO	6.5	8.6	15.6	12.3	13.0	12.5	8.2	3.0	9.6	3.6	6.4	6.2	1.4
	FeO	27.8	25.6	23.1	21.1	26.0	26.0	25.0	30.0	23.4	31.9	29.5	29.2	31.8
	MgO	2.6	1.8	3.0	1.9	3.3	2.5	3.3	4.2	4.0	3.7	2.9	2.4	2.8
margin	MnO	8.5	11.3	17.1	15.1	15.0	16.5	11.9	3.4	10.6	5.5	8.6	6.0	1.8
	FeO	26.0	24.0	24.8	20.3	25.0	22.8	19.4	32.2	22.2	30.3	29.4	28.2	31.3
	MgO	2.3	1.3	2.5	2.2	2.6	1.7	1.9	2.8	3.2	2.6	2.2	2.1	2.3

	54	55	56	57	58	59	60	61	62	63	64	65	66	67
SiO ₂	36.34	37.43	36.07	37.05	37.01	36.36	34.83	36.50	36.37	34.90	36.17	37.53	34.62	35.94
TiO ₂	—	—	—	—	—	—	—	—	—	—	—	—	—	—
Al ₂ O ₃	22.64	21.85	22.19	21.60	20.71	21.61	21.32	21.74	22.86	21.32	21.73	22.26	21.92	22.33
FeO	31.42	14.00	21.66	29.54	28.81	31.73	32.51	29.84	28.46	29.07	26.11	37.18	24.42	28.22
MnO	5.25	19.47	12.46	5.27	6.41	6.41	5.20	7.55	7.93	8.84	8.06	0.41	10.61	9.82
MgO	2.84	5.57	3.83	3.77	2.75	3.35	3.99	4.80	2.92	4.63	4.88	1.79	4.48	4.06
CaO	1.70	1.65	2.95	2.33	0.71	1.03	1.24	1.40	2.38	1.06	2.79	2.09	4.11	1.30
K ₂ O	—	—	—	—	—	—	—	—	—	—	—	—	—	—
Total	100.19	99.97	99.15	99.55	100.98	100.52	99.09	101.83	100.92	99.82	99.74	101.26	100.16	101.68
center	MnO	4.6	17.8	11.5	4.8	11.3	6.0	4.9	7.2	6.8	7.6	7.3	0.4	9.1
	FeO	32.3	14.3	22.0	30.0	29.5	32.3	32.6	31.1	30.0	30.3	25.5	37.3	29.2
	MgO	3.0	7.1	4.1	4.1	2.7	3.7	4.1	5.4	3.5	5.2	5.1	1.8	4.3
margin	MnO	7.1	21.6	16.4	7.4	11.0	7.8	5.7	9.1	10.2	12.4	9.8	0.4	11.4
	FeO	30.4	13.6	19.5	29.3	28.3	30.9	32.8	29.7	27.6	27.2	26.4	36.5	27.4
	MgO	2.2	5.0	3.0	2.9	2.6	3.0	3.9	3.3	2.2	3.5	4.1	1.8	3.4

	68	69	70	71	72	73	74	75	76	77	78	79	80	81	82
SiO ₂	39.11	40.03	38.44	37.25	36.06	38.10	38.25	39.55	35.60	36.00	36.12	39.63	34.83	36.69	36.84
TiO ₂	0.02	—	0.03	—	—	—	0.11	0.12	—	—	—	0.07	—	—	—
Al ₂ O ₃	21.75	21.45	21.56	22.88	20.97	22.68	23.49	21.02	21.28	21.08	22.04	21.13	21.72	21.91	21.15
FeO	29.32	27.06	24.92	28.55	28.88	27.84	25.25	25.57	27.82	24.44	29.51	16.48	28.13	29.11	33.62
MnO	7.31	9.56	10.38	6.58	7.17	6.95	9.30	7.22	11.25	14.45	9.35	17.70	10.89	4.17	3.91
MgO	3.08	2.74	3.62	2.50	4.28	2.69	3.59	3.71	2.82	3.97	3.57	4.18	4.45	3.81	2.69
CaO	0.86	0.97	1.08	2.10	0.76	2.52	1.13	2.98	0.94	0.57	0.98	2.02	0.79	4.62	0.73
K ₂ O	—	—	—	—	—	—	—	0.04	—	—	—	—	—	—	—
Total	101.46	101.81	100.02	99.86	98.12	100.77	101.13	100.20	99.70	100.69	101.61	98.52	100.81	100.31	98.94
center	MnO	6.7	9.2	10.4	5.6	5.8	5.9	7.7	6.9	10.0	13.4	8.7	16.5	5.2	3.7
	FeO	29.1	27.6	24.6	29.5	30.6	28.1	25.8	23.0	28.1	24.8	29.9	17.0	28.6	34.2
	MgO	3.2	2.8	3.9	2.6	5.0	3.0	4.1	3.0	3.2	4.4	3.9	5.0	3.9	2.9
margin	MnO	9.0	9.9	11.9	7.8	9.3	9.6	11.9	10.3	13.6	16.4	10.9	20.6	3.4	6.1
	FeO	27.9	26.6	23.4	25.9	27.5	26.9	23.4	25.0	26.5	23.1	28.8	15.8	29.5	31.7
	MgO	2.7	2.6	3.1	1.5	3.2	2.1	2.6	2.7	2.4	3.1	3.0	3.5	3.4	2.3

Fig. 4. Profiles of zoned garnets traversed across the diameter. Numbers are the same as those in Table 1 and Fig. 1.

Fig. 5. Mn-Mg-Fe diagram of zoned garnets in Area I, I' and II.

Circles show the compositions of center. Bars or arrows projected therefrom indicate the compositional changes toward the margins.

pattern is in the opposite pattern to that of Area I. The pattern is characterized by the rapid and distinct increase of MnO and the antipathetic decrease of FeO and MgO towards the periphery. Consequently either profiles of MnO, FeO and MgO become the reverse what are seen in the normal zonation mentioned above (Fig. 4). The CaO profile appears usually synpathetic with that of MgO with an exception of No. 46 in which CaO exceeds MgO in amount.

Garnets from Area I' also show the same pattern to those from Area II. As mentioned above, Area I' is represented by the polymetamorphic

hornfels derived from the rocks of Area I. So it is highly probable that garnets from Area I' may be polymetamorphic origin. In this respect the MnO profile of No. 19 which was collected from near the boundary between Area I and Area II is very interesting. It shows fundamentally the normal type zonation but a slight increase in MnO at the periphery may suggest a polymetamorphic effect. The same phenomena are observed in No. 21 etc. To be of special notice, on the other hand, a number of garnets occurring in Area II, e.g. Nos. 45, 55, 56, 70, 71, 74, and 75, exhibit somewhat complex structures characterized by the

Fig. 6. Plots of bulk compositions of garnets on Mn-Mg-Fe diagram.

presence normally zoned cores which may be considered to represent the relic garnets of the earlier metamorphism. The specimen, No. 59, is an example of cracked and, in turn, recrystallized garnet which may have formed originally a single crystal. The specimen, No. 77, shows the coalescing-together of two neighbouring grains.

To our knowledge the zoned garnets of the reverse type have been hitherto reported from the Fanad aureole of Donegal (EDMUNDS and ATHERTON, 1971), the Glen Clova aureole of Dalradian (CHINNER, 1962) and the Minnesota River valley (GRANT and WEIBLEN, 1971), among which the former two are polymetamorphic terrains and the last one is a migmatite terrain. In Japan, garnets

from the Iiba-Raizan area of the Sangun metamorphic belt of northern Kyushu are of the reverse type of zonation (KANO, 1971). According to KARAKIDA (1969) these garnets are found in polymetamorphic hornfels by the Itoshima granodiorite of the Cretaceous age. The hornfels are what were derived from the pelitic schists of the greenschist facies.

The chemical inclination of such zonation is clearly recognized in Mg-Mn-Fe triangle diagram (Fig. 5). Traversing from the center to the outside, garnets in Area I become rich in Fe and poor in Mn, whereas the majority in Area II including Area I' are decreased in Mg and increased in Mn with an exception of No. 81 which is found in a cummingtonite-hornblende bearing biotite schist (see Table 1). Nevertheless it must be remarked that its CaO profile of zonation is of reverse type as like as that of No. 46, though the MnO profile is of the normal type of zonation.

The bulk compositions of the zoned garnets are plotted on Mn-Mg-Fe triangle diagram (Fig. 6). Apparently the garnets in Area I and those in Area II including Area I' are grouped into two separate regions, of which the former is characterized by the low Mg and relatively high Mn/Fe, whereas the latter is of high Mg and relatively low Mn/Fe. Among these in Area II, Nos. 55, 79, and 65 are somewhat extraordinary in composition. In this respect, it is of notice that the former two garnets occur in quarzose mica schists and the latter garnet is found from an abnormally ferruginous and aluminous schists.

On Ca-Mg-(Mn+Fe) diagram the garnets are also well divided into two groups as shown in Fig.

Fig. 7. Plots of bulk compositions of garnets on Ca-Mg-(Fe+Mn) diagram.

Table 7. Microprobe analyses of biotites from Area I.

	1	2	3	4	5	6	7	8	9	10	11
SiO ₂	36.50	35.92	36.54	35.14	38.03	36.85	36.27	36.43	35.89	34.75	33.51
TiO ₂	1.90	1.60	2.05	1.85	1.48	2.07	1.48	1.80	2.51	1.60	2.03
Al ₂ O ₃	17.69	19.20	20.77	17.10	19.58	23.78	18.50	18.17	18.93	18.12	15.67
FeO	20.29	22.64	21.86	19.76	18.84	22.17	19.07	20.64	25.59	21.71	24.07
MnO	0.28	0.17	0.28	0.35	0.21	0.60	0.34	0.63	0.90	0.16	0.60
MgO	8.08	6.64	6.61	8.54	8.50	3.15	9.45	7.85	4.27	8.78	4.82
CaO	0.08	0.20	0.08	0.07	0.08	0.22	0.06	0.06	0.07	0.14	—
Na ₂ O	0.09	0.10	—	—	—	—	—	—	—	—	—
K ₂ O	8.49	7.72	8.25	9.23	8.37	7.31	8.72	9.30	8.52	8.13	9.53
Total	93.41	94.17	96.44	92.03	95.08	96.17	93.89	94.88	96.67	93.39	94.28

	12	13	14	15	16	17	18	19	20	21	22	24
SiO ₂	37.14	36.61	37.08	36.78	33.88	35.05	36.93	35.11	35.76	36.27	36.46	33.24
TiO ₂	1.65	1.57	3.33	2.56	3.31	3.40	2.10	2.20	2.97	2.29	2.78	2.91
Al ₂ O ₃	17.71	16.51	18.68	17.23	17.63	20.20	16.28	16.01	19.20	19.33	19.94	22.14
FeO	19.74	22.54	17.70	21.32	21.05	19.02	19.94	20.78	20.52	18.23	19.25	19.91
MnO	0.27	0.33	0.59	0.41	0.47	0.53	0.56	0.70	0.66	0.45	0.48	0.25
MgO	9.96	8.02	9.35	7.38	8.31	8.87	10.11	9.49	7.55	8.68	8.13	8.80
CaO	0.01	0.21	—	0.08	—	0.07	0.03	0.07	0.03	0.01	0.08	0.01
Na ₂ O	—	—	—	0.08	—	—	—	—	—	—	—	—
K ₂ O	9.00	6.73	9.59	9.03	9.54	9.60	8.75	9.15	9.37	8.78	8.81	8.51
Total	95.49	92.52	96.33	94.87	94.19	96.75	94.71	97.51	96.04	94.04	95.94	95.77

7. In this case, however, Ca content appears to be equally distributed in either group and the peculiarity of Nos. 55, 79, and 65 is indistinguishable.

Miyashiro (1953) remarked the decreasing of MnO of garnet in pelitic-psammitic metamorphic rocks in the progressive metamorphism. Indeed average MnO content of the garnets in Area I is 14.82 wt% and in Area II it is 8.70 wt%. Since the metamorphic grade seems to be higher in Area II than in Area I, these data seem to be accordance with his hypothesis. However this may not be the decisive conclusion, because MnO content of the host rocks is higher in Area I (average 0.27 wt%) than in Area II (average 0.10 wt%). As Atherton said, one reason of higher content of MnO in the garnets in Area I may be attributed to the higher content of MnO in the host rocks.

Atherton (1968) considered that the value of K determined by the formula, $K = (\text{MnO in the center of garnet}) / (\text{MnO in the host rock})$, on pelitic-psammitic rocks regularly decreases from 81 at the garnet isograd to 25 in the kyanite zone. In Abukuma, however, the K-values are variable and rather reverse in tendency. They are 45.4—149.3 (average 75.7) in Area I and 25.3—246 (average 121.3) in Area II. Therefore any critical conclusion cannot be induced from these values. Miyashiro (1953 and 1958) considered that the

high content of MnO in garnet was the result of low pressure metamorphism. Müller and Schneider (1971), however, stated that there was no relationship whatsoever between pressure and the MnO content of garnet, but the relatively high content of MnO in garnets in Abukuma might be due to the high fugacity of oxygen. In Stanvanger they concluded that the higher MnO content of garnets of graphite bearing pelitic schists in "Zone I" were due to the higher oxygen fugacity and the lower MnO content of those of graphite-free pelitic schists in "Zone II" were related to the lower oxygen fugacity.

Nevertheless, according to our study in Abukuma, the garnets from the rocks of Area II rich in graphite show lower contents of MnO than those in the rocks of Area I bringing no graphite. As stated below it is quite difficult to explain what factor controls the MnO content in garnet.

VI. Composition of Biotite

The chemical analyses of biotites coexisting with the garnets are listed in Tables 7 (Area I), 8 (Area II) and 9 (Area I'). They are plotted on the Mn-Mg-Fe triangle diagram (Fig. 8). From the figure it can be said that the biotites in Area II are richer in Mg than those in Area I, though some overlap each other. Judging

Table 8. Microprobe analyses of biotites from Area I'.

	31	32	33	34
SiO ₂	36.62	37.42	36.67	35.78
TiO ₂	2.56	2.63	3.00	2.68
Al ₂ O ₃	19.44	14.40	15.86	17.63
FeO	18.34	20.17	18.43	19.95
MnO	0.43	0.48	0.39	0.37
MgO	8.73	10.48	11.36	8.37
CaO	0.04	0.11	—	0.06
Na ₂ O	—	—	—	—
K ₂ O	8.99	10.43	9.61	9.18
Total	95.39	96.20	95.32	94.07

from the difference in the chemical compositions of the whole rocks of each Area this is natural. TiO₂ content in the biotites is also worthy of notice. It is higher in Area II (average 2.88 wt.%) than in Area I (average 2.14 wt.%). This may be also correlatable to the difference between opaque minerals from both Areas. As already mentioned, they are ilmenite in Area I and magnetite and/or graphite in Area II.

In addition, we could not find any compositional zonation in biotite. Therefore the composition of biotite may be considered to reflect the condition of the last stage of metamorphism.

Table 9. Microprobe analyses of biotites from Area II.

	40	41	42	43	44	45	46	47	48	49	50	51	52	53
SiO ₂	35.90	35.76	35.65	34.92	37.62	33.86	37.01	35.85	36.30	35.88	35.29	36.32	38.07	36.63
TiO ₂	2.74	1.89	2.68	2.75	2.94	3.38	3.33	3.10	2.56	3.18	3.38	2.37	2.60	3.69
Al ₂ O ₃	17.24	21.85	19.47	16.11	18.54	18.57	15.29	21.24	21.04	19.96	21.08	19.26	20.77	16.60
FeO	19.12	19.32	17.22	19.25	19.67	19.83	20.81	15.36	13.90	19.81	19.35	18.14	18.53	22.42
MnO	0.27	0.28	0.59	0.49	0.42	0.68	0.78	0.13	0.34	0.20	0.16	0.19	0.28	0.09
MgO	9.41	8.71	9.24	10.90	7.27	9.10	9.35	10.90	11.61	6.08	7.94	9.29	8.64	7.58
CaO	0.10	0.03	0.03	—	—	—	0.03	0.07	0.04	—	0.03	0.05	—	—
Na ₂ O	—	—	—	—	—	—	—	—	—	—	—	—	—	—
K ₂ O	8.64	8.74	9.10	8.91	9.23	9.28	9.01	8.54	9.24	8.81	9.91	8.82	9.81	9.34
Total	93.42	96.59	93.99	93.33	95.69	94.70	95.61	95.19	95.04	93.91	97.14	94.44	98.70	96.36

	54	55	56	57	58	59	60	61	62	63	64	65	66	67
SiO ₂	35.22	36.87	35.74	34.94	35.91	35.65	35.40	36.42	36.25	35.93	35.33	33.34	36.80	36.47
TiO ₂	3.52	2.20	2.83	2.94	3.12	3.54	3.90	2.92	2.75	3.65	2.38	1.94	2.39	2.74
Al ₂ O ₃	17.16	20.81	18.67	18.62	20.00	19.28	19.58	18.65	18.05	19.38	19.18	19.67	18.29	22.28
FeO	21.81	9.43	14.44	18.94	19.07	19.64	20.05	17.81	19.80	18.21	16.77	24.34	17.66	18.55
MnO	0.29	1.86	0.63	0.27	0.26	0.22	0.17	0.24	0.23	0.41	0.49	0.00	0.41	0.36
MgO	8.40	14.20	11.73	9.87	9.57	7.71	8.05	8.47	9.84	9.37	9.95	5.50	10.25	9.19
CaO	0.03	—	0.03	0.03	—	—	—	0.04	0.04	—	—	0.07	—	0.03
Na ₂ O	—	—	—	—	—	—	—	0.12	—	—	0.10	—	—	—
K ₂ O	9.49	8.46	9.27	8.92	9.84	8.62	9.55	9.07	9.14	9.96	8.90	8.02	9.91	9.77
Total	95.93	93.85	93.34	94.52	97.77	94.66	96.70	93.74	96.10	96.91	93.08	92.88	95.71	95.39

	68	69	70	71	72	73	74	75	76	77	78	79	80	81	82
SiO ₂	35.95	36.56	36.69	35.46	36.38	34.52	35.68	36.69	35.21	36.46	37.66	38.75	38.11	36.76	35.53
TiO ₂	1.96	2.59	2.18	3.35	3.07	2.89	2.77	3.61	2.71	3.86	2.65	2.20	2.28	3.27	3.14
Al ₂ O ₃	19.80	20.77	19.89	19.20	19.82	18.86	20.46	18.36	17.80	18.44	18.10	18.75	20.54	15.96	19.89
FeO	18.75	18.56	16.64	18.36	18.35	18.73	17.14	15.43	19.56	17.67	18.32	10.32	16.66	19.83	20.02
MnO	0.27	0.36	0.44	0.13	0.31	0.16	0.43	0.44	0.48	0.55	0.35	0.97	0.39	0.12	0.18
MgO	8.23	7.94	9.82	7.79	9.68	9.57	10.17	10.53	9.17	10.26	8.84	16.46	9.68	10.36	8.45
CaO	—	—	—	0.05	0.10	0.02	0.03	0.03	—	—	—	—	0.02	0.09	0.02
Na ₂ O	—	—	—	—	—	—	—	—	—	—	—	—	0.39	—	—
K ₂ O	9.27	9.22	9.12	8.92	9.28	9.20	9.24	9.29	9.38	9.84	8.98	8.85	8.93	7.96	8.47
Total	94.23	96.00	94.79	93.26	96.99	93.95	95.93	94.37	94.31	97.08	94.90	96.30	96.99	94.35	95.70

Fig. 8. Plots of the compositions of biotites on Mn-Mg-Fe triangle.

VII. The Host Rocks, Garnets, and Biotites in Area I'

The pelitic-psammitic metamorphic rocks around the Yoshimagawa and the Iritono plutonic masses belong originally to Area I, and apparently it is not easy to distinguish them from the normal rocks in Area I. Under the microscope, however, the grain size is somewhat larger and the texture becomes hornfelsic. K-feldspar, graphite, *etc.* are recognized. The chemical analyses of host rocks are shown in Table 3. In AKF diagram (Fig. 2), Mn-Mg-Fe diagram (Fig. 3), *etc.*, they are plotted in the same field with those of Area II. It means that the polymetamorphism by the Yoshimagawa and the Iritono plutonic masses may have been not isochemical but allochemical and metasomatic in some degree.

Chemical analyses of coexisting garnets and biotites are presented in Tables 5 and 8, respectively. In the Mn-Mg-Fe diagram (Fig. 5) the garnets are plotted in the field of Area II and exhibit the same pattern of zonation as shown in Fig. 9. These chemical features of the host rocks in Area I' are very important in order to consider the genesis of the reverse zonation of garnets in Area II. In view of the above-mentioned chemistry of the rocks, garnets and biotites in Area I', it is quite natural to consider that the rocks in Area II may have been subjected with polymetamorphism associated with some metasomatic change of the bulk composition.

VIII. Discussion

In the Mn-Mg-Fe triangle diagram (Figs. 10 and 11) the relation between the tie-line of garnet and biotite and the bulk composition of the host rock is examined. In this case, Fe_2O_3 of the rocks is combined with FeO and is calculated as total FeO.

Fig. 9. Profiles of MnO and FeO contents of garnets in Area I'.

Fig. 10. Tie-lines of coexisting biotites and garnets and plots of bulk compositions of the host rocks (cross) from Area I on Mn-Mg-Fe triangle.

Fig. 11. Tie-lines of coexisting biotites and garnets and plots of bulk compositions of the host rocks (cross) from Area II on Mn-Mg-Fe triangle.

It is noticed from these figures that the bulk compositions of the host rocks are always plotted near by the corresponding tie-lines of garnet and biotite. Although the plots of No. 5 and No. 9 in Fig. 10 lie exceptionally far away from the corresponding tie-lines, this is due to the presence of abundant magnetite in these rocks. Considering the common including of magnetite or ilmenite in the pelitic-psammitic rocks it is not strange that their plots lie always inside the garnet-biotite-magnetite (or ilmenite) triangle satisfying the quantitative relation expected from the mineralogical phase rule.

In addition there are a few exceptional tie-lines which may be unable to explain their gradient only as the function of P and T. For example, in the case of the mutually intersecting tie-lines of No. 9 and No. 10, another factor controlling the gradient should be considered, for they were collected from nearly the same locality and therefore the conditions of formation may be substantially the same at least concerning T and P. Presumably it may be related to any difference of the bulk composition between both rocks. In this respect attention may be paid to a fact that the oxidation ratio, $\text{Fe}^{+3}/(\text{Fe}^{+2} + \text{Fe}^{+3})$ of No. 10 is remarkably lower than that of No. 9. The ratios are 0.36 in No. 10 and 0.56 in No. 9.

As MIYASHIRO (1953) demonstrated schematically, the gradient of tie-line of garnet and biotite in the triangle may reflect the P-T conditions under which the rock was metamorphosed. It is true that the tie-lines of the lower grade metamorphic rocks of Area I are steeply inclined in general, whereas the higher grade metamorphic rocks of Area II show relatively gentle slopes. Such situation is also the same in the Bohemian

Fig. 12. Tie-lines of coexisting biotites (cross) and garnets (circle) of granulites in Bohemian massif (after MATEJOVSKA, 1970).

Fig. 13. Tie-lines of coexisting biotites (cross) and garnets (circle) of gneisses in Nova Scotia (after PHINNEY, 1963).

granulites (MATEJOVSKA, 1970) compared with the gneisses of Nova Scotia (PHINNEY, 1963). The tie-lines of the former are evidently of lower gradient than those of the latter (Figs. 12 and 13).

Considering these facts, MIYASHIRO's idea appears to be qualitatively right. However, it must be noted that, in the Gozaisho-Takanuki district, character of metamorphism marked on the garnets must be quite different between in Area I and Area II. Because the garnets of Area II show the essentially different zonation from those in Area I as described above. Accordingly these garnets from both Areas can be never considered to have formed under a single cycle metamorphism.

Next then the partition of elements between the

Fig. 14. Fe/Mg relation between garnet and biotite of the metamorphic rocks in Abukuma.

Fig. 15. Mn/Fe relation between garnet and biotite of the metamorphic rocks in Abukuma.

coexisting garnet and biotite is discussed. In this case the direct consideration on Mn-Mg-Fe triangle as MIYASHIRO (1953) discussed is difficult, so we consider the partition coefficients from the following three equations:

They are represented graphically in Figs. 14—16,

where the compositions of garnets were calculated separately either at the margin or at the center.

Equation (1): the apparent partition coefficients,

$$K_D = (\text{Fe}/\text{Mg})_{\text{gar}} / (\text{Fe}/\text{Mg})_{\text{biot}},$$

for each pair of garnet (center)/biotite in Area II including Area I' are 2.6—6.3 (3.7 in average), while for the pairs of garnet (margin)/biotite the values are a little larger than the former, that is, 3.0—7.4 (4.5 in average). It is noted in either diagram that the plots of partition are aligned into a narrow zone. This may suggest that the

Fig. 16. Mn/Mg relation of coexisting biotite and garnet of the metamorphic rocks in Abukuma.

Fig. 17. Fe/Mg relation between biotite and garnet of gneisses in Nova Scotia.

c: area of plots for garnet(center)/biotite of the rocks in Area I, Abukuma (ref. to Fig. 14-right). M: area of plots for garnet (margin)/biotite of the rocks in Area I, Abukuma (ref. to Fig. 14-left).

chemical equilibrium between garnet and biotite would have been attained during the period of garnet growth.

On the other hand those of the rocks in Area I are 3.5—13.8 for the pairs of garnet (center)/biotite and 3.4—8.5 for those of garnet (margin)/biotite which averages are 7.5 and 6.2 respectively. Differing from the rocks of Area II, the plots are

not so well aligned on a line of 45° because of less dispersion of Fe/Mg ratio. However, it is important that the values become slightly smaller at the margin than in the center. Hence both plots of Area II and Area I become closer to each other in the pairs of garnets(margin)/biotite. This is due to the mutually opposite types of garnet zonation in both Areas.

In the Bohemian granulites above cited, the plots of partition pairs are scattered very irregularly, so that the coexisting garnets and biotites are considered to have not approached to the state of chemical equilibrium.

In the Novascotian gneisses, the plots are concentrated in a small area which lies very close to that of the rocks of Area I in Abukuma (Fig. 17). It means that the gneisses would attained to the equilibrium state under the similar P and T conditions as Area I. In other words the metamorphism of Area I may belong to the same type of Nova Scotia, that is, the *Dalradian* type.

Equation (2); the apparent partition coefficients for each pair of garnet(center)/biotite are 9—70 (18.1 in average) in Area II and 20—146 (45.8 in average) in Area I. Those for pairs of garnet (margin)/biotite in Area II including Area I' are 11—97 (23.4 in average) and 12—59 (25.8 in average) respectively. The plots are illustrated in Fig. 15. Fig. 18 presents those of Nova Scotia for reference. The resemblance between both is quite evidently.

Equation (3); the apparent partition coefficients in Area II including Area I' are 28—145 (70 in

Fig. 18. Mn/Fe relation between biotites and garnets of gneisses in Nova Scotia.

Broken lines are the same as those in Fig. 15-left.

Fig. 19. Mn/Mg relation between biotites and garnets in gneisses in Nova Scotia.

Broken lines are the same as those in Fig. 16-left.

average) for the garnet (center)/biotite and 56—317 (111 in average) for the garnet (margin)/biotite. Those in Area I are 82—1451 (352 in average) and 81—447 (153 in average) respectively. The plots are shown in Fig. 16 and those of Nova Scotia are plotted on Fig. 19.

To summarize the above described apparent partition coefficients for the three equations are collectively listed on Table 10 together with those of Nova Scotia. In every reaction, the apparent partition coefficient decreases from the center of garnet to its margin in Area I, while in Area II it is in the opposite way. Hence it is suggested that the reactions of these three equations by the

Table 10. The mean apparent partition coefficients for the pair of coexisting garnet and biotite.

Equation	Area	garnet		Nova Scotia
		center	margin	
(1) Fe/Mg	I II *	7.5 3.7	6.2 4.5	ca. 7
(2) Mn/Fe	I II *	45.8 18.1	25.8 23.4	ca. 26
(3) Mn/Mg	I II *	352 70	153 111	ca. 200

* Area II includes Area I'.

changes of P and/or T are essentially the same in their direction. Although the difference in the coefficients between Area II and Area I become small in the pairs of garnet(margin)/biotite, the value is always lower in Area II than in Area I indicating that the difference may be the function of P and/or T.

The volume change (ΔV) for the equation (1) under the standard state is $+2.67 \text{ cm}^3/\text{mole}$ and thus the increase of pressure may act so as to increase the partition coefficients. About the volume changes for the equations (2) and (3), nothing is known because of the absence of data on molar volume of Mn-biotite. If the molar volume of Mn-biotite does not differ substantially from that of annite or it might be rather greater than that of annite, the ΔV for the equation (2) should be positive. It will be also positive for the equation (3), though the value may be smaller than that for the equation (2). Therefore it seems reasonable to say that the partition coefficients of these three equations may become larger with the increasing of pressure.

On the other hand we have little knowledge on the temperature dependence upon partition equilibrium because of the absence of free energy data on annite even under the standard state. However it seems possible to consider that the partition coefficients may become smaller with the increasing of temperature, on the assumption that the pressure at the time of formation of the inner part of garnet in Area II would have not be so lower than that in Area I. Such assumption is reasonable because the inner part of garnet in Area II can be considered to be a relic as like as kyanite which may be assumed as a relic of older metamorphism.

From these considerations it is concluded that: (1) the partition coefficient is raised by the increasing of P and/or the decreasing of T, or in

other words, the decreasing of P and/or the increasing of T cause its lowering; (2) the change of apparent partition coefficient in Area II is mainly due to recrystallization under the condition of lowering T, while the change in Area I means either the successive increasing of T and/or the successive decreasing P.

Finally with respect to the polymetamorphism in Abukuma, the history of metamorphism is briefly summarized as follows: 1) the first (older) metamorphism might have been accomplished under the condition of the *Dalradian* type, where the temperature might have been higher in Area II than in Area I; 2) after that the second (younger) thermal metamorphism have happened under somewhat lower temperature condition than the maximum of the former metamorphism. This metamorphism was rather of the contact type and both Area II and Area I are the polymetamorphic aureoles superposed on the older metamorphic terrain. In addition it may be remarked that a certain degree of metasomatic change in the host rocks would have happened during the last metamorphism, though the reason is yet indistinct.

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中部阿武隈の泥質-砂質岩源変成岩中の共存するざくろ石-黒雲母の化学的研究

加 納 博・黒 田 吉 益

(要 旨)

阿武隈高原の御齊所-竹貫変成岩についての一連の研究として、砂質-泥質岩中の共存するざくろ石-黒雲母の化学的性質を報告した。EPMAで分析したざくろ石-黒雲母の対は65組、その中、母岩を分析したもの27個である。

ざくろ石はすべて累帯構造をしているが、そのパターンは、MnOが中心に多い正累帯型と、反対に周縁に多い逆累帯型とがある。その分布はFig. 1の境界線で、はっきりとArea I (正累帯型)とI', II (ともに逆累帯型)とに分けられる。IIおよびI'のざくろ石は全組成としてIのものよりMnが少なく、Mgが多い。したがってFe/Mgも小さい。これに似た傾向は母岩にも見られるので、ざくろ石のMnOの変化は母岩の組成による影響が大きいのかも知れない。黒雲母の化学組成につ

いても同様な傾向がある。

Mn-Mg-Fe 三角図中のざくろ石-黒雲母の tie-line の勾配はIで急で、IIで緩くなる (Fig. 10, 11)。しかし、例外もあり、解釈は一樣ではない。ざくろ石-黒雲母間におけるFe-Mg, Mn-Fe, Mn-Mgの分配係数を計算すると、どの場合もIIよりIが大きい。K_Dの対数グラフ上ではIとIIの中心部、周縁部で、それぞれほぼ45°の直線上に投影される。ただ周縁部ではIとIIの間の差が少なくなる。このことは、Iの正累帯型が温度の増進変成作用を示すとすれば、IIの逆累帯型はより低温の複変成作用をうけたことを示すものであろう。Iの分配係数がNova ScotiaのDalradian型の変成帯のものによく似ていることも注目に値する。

地	名		
Abuki	阿吹	Miyamae	宮前
Baba	馬場	Miyamoto	宮本
Daibara	大原	Nabana	名花
Eryûda	江竜田	Nakanosawa	中ノ沢
Furudono	古殿	Nita	仁田
Furudono-Tokusa	古殿町戸草	Nyudô	入道
Gozaisho (Gosaisho)	御齊所	Oinudaira	男犬平
Hanawa	塙	Onikoshi	鬼越
Hanazono	花園	Osogawa	遅川
Hônosawa	宝ノ沢	Onasawa	小名沢
Horiuchi	堀内	Ronden	論田
Huruuchi	古内	Sakamichi	坂路
Ide	井出	Sakune	作根
Idosawa	井戸沢	Sawatari	沢渡
Iiba-Raizan	飯場一雷山	Samegawa	鮫川
Iritôno	入遠野	Shinden	新田
Ishikawa	石川	Shuku	宿
Ishizumi	石住	Tabito	田人
Jûmonji	十文字	Takanuki	竹貫
Kaidomari	貝泊	Tairakuchi	平口
Kaidomari-Tokusa	貝泊村戸草	Takabo	高房
Kaiya	貝屋	Takano	高野
Kamiyama	神山	Uchide	内出
Koshidai	越代	Yachi	谷地
Kumakura	熊倉	Yanoyama	矢野山
Kuwakida	鍬木田	Yazukuri	矢造
Maegi	前木	Yoshimagawa	好間川
Mappo	松風	Yokogawa	横川
		Watarase	渡瀬