SUBCALCIC HORNBLENDES IN THE EPIDOTE AMPHIBOLITES OF THE YAMAGAMI METAMORPHIC COMPLEX IN THE NORTHERN ABUKUMA PLATEAU

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The chemical compositions of subcalcic hornblendes, epidotes, white mics and bulk rocks were reported from the Yamagami epidote amphibolites. The compositional variation of subcalcic hornblende solid solution was discussed in the system of tschermakite-pargasite-eckermannite-glaucophane under the conditions of glaucophanitic metamorphism. The solvus between subcalcic hornblende and alkali amphibole is wide and subcalcic hornblende is soluble to about 1 in the M_4 site alkali content. There are two variation trends of subcalcic hornblendes in glaucophanitic metamorphism. Trend A, that is, the alkali content of the M_4 site increases with rising temperature, is the same as the contributions by Black (1973) and Toriumi (1975). Trend B, in which the alkali content of the M_4 site decreases and that of the vacant site slightly increases with rising temperature, is compatible with the general trend of subcalcic to calcic hornblende variation. Trend A is produced by the reactions among albite, chlorite, epidote and quartz, but Trend B must be created by the reactions among iron ores, carbonates, albite and epidote or by the breakdown of subcalcic hornblende_c to clinopyroxene-garnet-subcalcic hornblende_p or to quartz-subcalcic hornblende_p. The change of the trend from A to B is due to not only the metamorphic grade but also the bulk composition, specially the scarcity of the modal content of chlorite, quartz and albite.

INTRODUCTION

The Yamagami and Matsugadaira metamorphic complexes isolatedly expose in small in the sheared tectonic zone of the northern Abukuma Plateau. Minato (1960) and Kuroda and Ogura (1963) considered that the metamorphic age of the Matsugadaira metamorphic complex is pre-upper Devonian. K-Ar dating on muscovite from the metamorphic rock of the Yamagami district shows 300×10^6 years (Kawano and Ueda, 1965). Kuroda and Ogura (1963) reported the mineral assemblages and the chemical composition of rocks, subcalcic

hornblendes and some other minerals in this district and indicated that these metamorphic rocks were formed under the conditions of glaucophane schist type metamorphism.

Subcalcic actinolites and hornblendes characteristically appear in glaucophanitic metamorphic belts (Iwasaki, 1963; Banno, 1964; Miyashiro, 1965; Onuki *et al.*, 1976), but their chemical properties have not been investigated in detail. At lower temperatures, Black (1973) and Toriumi (1975) established the temperature dependent solvus in the systems of actinolite-glaucophane-tschermakite and actinolite-glaucophane-magneisoriebeckite, and they

concluded that both tschermakite and glaucophane molecules in calciferous amphibole increase with rising temperature at constant P_{H_2O} . On the other hand, Iwasaki (1963), Banno (1964) and Miyashiro (1973) indicated that subcalcic hornblendes transform to calcic hornblendes with increasing metamorphic grade in the epidote amphibolite facies. These succeeding and contrary events, which are the increasing of tschermakite and glaucophane molecules and the decreasing of glaucophane molecule with rising temperature in glaucophanitic metamorphic terranes, are interesting.

We will examine the chemical varia-

tion with increasing metamorphic grade in the system of tschermakite-pargasite-eckermannite-glaucophane.

BRIEF PETROGRAPHY

The epidote amphibolites consist mainly of subcalcic hornblende, epidote, sodic plagioclase (An-7-12), and variable amounts of quartz, white mica, chlorite, rutile and sphene. Twelve modal analyses together with chemical analyses are shown in Table 1. The mineral assemblages indicate that the Yamagami epidote amphibolites were formed under the conditions corresponding to the epidote amphibolite facies. Acces-

Table 1. Bulk chemical compositions and modal contents of the Yamagami epidote amphibolites.

	1	2	3	4	5	6	7	8	9	10	11	12
SiO ₂	45.84	47.63	47.41	46.26	45.84	46.84	51.00	46.38	46.87	45.67	47.57	46.46
TiO ₂	1.85	1.63	1.37	1.47	1.56	1.56	1.53	1.51	1.55	1.54	1.49	1.58
Al ₂ O ₃	16.81	15.18	15.24	16.91	15.48	15.24	16.63	16.13	14.96	16.39	15.76	16.75
Fe ₂ O ₃	3.47	4.50	4.79	5.52	5.94	6.26	3.48	6.10	3.07	4.73	8.15	7.40
FeO	8.30	7.17	6.37	4.49	6.61	5.17	5.44	5.95	5.68	7.40	3.88	4.25
MnO	0.21	0.17	0.15	0.17	0.16	0.20	0.14	0.16	0.17	0.16	0.18	0.19
MgO	9.01	7.27	6.85	5.48	7.04	6.39	5.08	6.78	8.10	6.71	5.47	3.66
CaO	6.03	9.16	10.62	12.08	9.54	12.61	9.49	9.96	7.63	11.29	11.44	13.88
Na ₂ O	3.38	3.16	3.37	1.87	2.72	1.74	3.59	2.60	4.77	2.11	3.16	3.21
K ₂ O	0.84	0.94	0.67	1.65	2.00	0.96	1.12	1.02	0.81	0.44	0.36	0.20
H ₂ O+	3.86	2.68	2.64	3.81	2.64	2.57	2.29	2.67	5.79	3.27	2.26	2.27
H ₂ O-	0.29	0.25	0.15	0.20	0.12	0.10	0.03	0.17	0.24	0.16	0.15	0.08
P ₂ O ₅	0.16	0.16	0.15	0.20	0.19	0.17	0.33	0.28	0.15	0.13	0.18	0.26
Total	100.05	99.90	99.78	100.11	99.84	99.81	100.15	99.71	99.79	100.00	100.05	100.19
Modal %												
Qz	6.8	5.4	-	7.3	4.1	7.9	2.1	2.2	0.1	8.5	0.4	0.0
Pl	10.8	16.2	21.3	13.7	7.3	7.6	26.7	22.8	33.4	9.8	26.0	9.0
Hb	33.4	49.3	43.7	27.5	60.4	49.4	34.7	50.1	27.8	47.2	43.0	62.7
Chl	1.5	5.9	-	2.1	2.3	4.1	0.3	2.1	17.0	0.3	0.6	4.0
Rut	1.8	2.6s	2.2	2.2s	2.7s	2.7s	1.9	2.3s	2.3	1.9s	2.0	2.7
W.Mica	-	4.6	-	18.6	8.9	13.8	+	0.6	6.0	+	+	-
Na-Amp	-	-	-	-	+	-	-	+	1.2	-	-	-
Calc	-	-	+	-	-	-	-	-	8.4	+	-	-
Act	+	+	+	+	+	+	+	+	+	+	+	+
Bi	+	-	-	-	-	-	-	+	-	-	-	-
Others	1.1	0.4	0.3	0.1	0.3	1.2	0.3	0.2	0.7	0.3	0.1	0.2

Chemical compositions were analysed by H. Onuki and modal compositions by M. Tagiri. +: present, -: absent, S: sphene coexists with rutile. Others: garnet, apatite, zircon and opaque minerals.

Table 2. Chemical compositions of subcalciic hornblendes from the Yamagami epidote amphibolites with their atomic ratios (anhydrous basis 0=23).

	1	2	3	4	5	6	7	8	9	10	11	12
SiO ₂	45.65	46.80	46.93	45.82	44.61	45.98	45.18	46.44	44.58	44.95	46.40	45.73
TiO ₂	0.87	0.65	0.53	0.59	0.78	0.74	0.91	0.95	0.92	1.10	0.89	0.71
Al ₂ O ₃	13.68	11.32	10.24	11.56	12.01	11.40	10.43	9.59	11.66	10.03	9.23	10.93
Fe ₂ O ₃	4.31	5.53	5.21	5.28	7.17	5.56	6.53	4.66	5.38	5.05	7.03	5.94
FeO	10.54	10.95	10.76	10.04	9.15	10.30	9.93	10.76	9.29	11.64	10.32	10.97
MnO	0.22	0.21	0.24	0.29	0.23	0.22	0.26	0.25	0.23	0.24	0.28	0.27
MgO	12.29	10.85	11.96	12.15	10.89	11.76	11.43	12.06	13.32	12.09	11.16	11.29
CaO	7.87	8.59	9.37	9.12	9.14	8.70	8.93	9.50	8.31	8.51	8.35	8.13
Na ₂ O	2.62	2.53	2.26	2.37	2.73	2.33	3.04	2.37	2.93	2.44	3.15	3.16
K ₂ O	0.25	0.55	0.24	0.77	0.67	0.74	0.56	0.51	0.50	0.73	0.49	0.52
H ₂ O ⁺	2.03	2.02	2.07	2.12	2.12	2.09	2.63	2.90	2.17	3.25	2.51	2.10
H ₂ O ⁻	0.02	0.09	0.06	0.09	0.24	0.17	0.10	0.06	0.32	0.04	0.18	0.12
Total	100.35	100.09	99.87	100.20	99.74	99.99	99.93	100.05	99.61	100.07	99.99	99.87
Atomic ratio												
Si	6.559	6.799	6.830	6.661	6.551	6.699	6.657	6.825	6.527	6.675	6.834	6.708
Al ^{IV}	1.441	1.201	1.170	1.339	1.449	1.301	1.343	1.175	1.473	1.325	1.166	1.292
Al ^{VI}	0.875	0.738	0.587	0.641	0.629	0.656	0.467	0.486	0.539	0.430	0.435	0.595
Ti	0.094	0.071	0.058	0.065	0.086	0.081	0.101	0.105	0.101	0.123	0.098	0.078
Fe ³⁺	0.466	0.604	0.570	0.578	0.792	0.609	0.724	0.516	0.593	0.564	0.778	0.655
Fe ²⁺	1.266	1.330	1.309	1.221	1.124	1.255	1.223	1.322	1.137	1.445	1.271	1.344
Mn	0.027	0.026	0.030	0.036	0.028	0.027	0.033	0.031	0.028	0.030	0.035	0.033
Mg	2.630	2.348	2.593	2.631	2.382	2.553	2.509	2.640	2.905	2.675	2.448	2.464
Ca	1.211	1.337	1.461	1.420	1.437	1.357	1.409	1.495	1.303	1.354	1.317	1.276
Na	0.730	0.712	0.638	0.667	0.776	0.658	0.867	0.674	0.832	0.703	0.899	0.898
K	0.046	0.101	0.045	0.143	0.125	0.138	0.106	0.095	0.093	0.139	0.092	0.097

Analysts: 1-6, 9 and 12 by H. Onuki and 7, 8, 10 and 11 by M. Tagiri.

sory minerals are calcite, apatite, garnet, alkali amphibole, actinolite, biotite, leucocoxene, zircon and opaque minerals. Actinolite commonly replaces the rim and cleavage of subcalciic hornblendes. Alkali amphibole sometimes grows over the rim of subcalciic hornblende and in chlorite. Garnet alters to chlorite and epidote. Rutile is frequently surrounded by sphene. Opaque minerals are very rare in the rocks. Albite, epidote, actinolite and quartz veins appear mostly, and calcite, biotite and

sphene veins are rare in the rocks. Minerals of the modal analysis do not include vein minerals.

CHEMICAL COMPOSITIONS OF SUB-CALCIC HORNBLLENDE, EPIDOTE AND WHITE MICA

The chemical compositions of twelve subcalciic hornblendes are shown in Table 2. These analyses indicated in Fig. 1 by means of the representation proposed by Tagiri and Onuki (1974) show a restricted

Fig. 1. Subcalcic hornblendes from the Yamagami epidote amphibolites.

Tr=tremolite, Ts=tschermakite,
Ed=edenite, Pa=pargasite,
Ka=katophorite, Gl=glaucophane.

compositional range. The mean alkali content of the M_4 site and its standard deviation are 0.475 and 0.078, respectively. The alkali content of the vacant site is about from 0.2 to 0.5 and the trivalent cation content about from 2.3 to 3.0. The mean Fe^{3+}/Fe^{2+} ratio and its standard deviation are 0.49 and 0.10, respectively.

The mean chemical compositions of four white micas and nine epidotes coexisting with subcalcic hornblendes are shown in Table 3 with their standard deviations. White micas in the epidote amphibolites are

Table 3. Chemical compositions of white mica and epidote from the Yamagami epidote amphibolites with their standard deviations.

	White mica (n=4)		Epidote (n=9)	
		s.d.		s.d.
SiO_2	47.31	0.48	37.61	0.12
TiO_2	0.34	0.03	0.19	0.05
Al_2O_3	29.19	1.49	24.41	0.37
Fe_2O_3	3.36	1.40	12.20	0.39
FeO	1.27	0.16	0.06	0.05
MnO	0.02	0.01	0.18	0.02
MgO	2.97	0.12	0.29	0.11
CaO	0.21	0.05	22.66	0.20
Na_2O	0.94	0.26	0.19	0.06
K_2O	10.09	0.49	0.06	0.03
H_2O^+	4.34	0.10	2.00	0.05

Analyst H. Onuki

composed of the solid solution of about 40 molecular percent phengite, about 20 molecular percent paragonite and about 40 molecular percent muscovite. The average $Fe^{3+}/(Fe^{3+}+Al)$ ratio of analysed epidotes is about 0.24. Each of the wet chemical analyses on white micas and epidotes give an uniform composition in contrast with the variety of the host rock compositions.

SUBCALCIC HORNBLENDE SOLID SOLUTION

Though subcalcic hornblendes are a characteristic mineral of glaucophanitic metamorphism, they also sometimes appear in other metamorphic terranes. Now, we used forty-two data by wet chemical analysis apart from those of this paper as follows, Lacroix (1942), Van der Plas (1959), Iwasaki (1963), Kuroda and Ogura (1963), Banno (1964), Coleman *et al.* (1965), Binns (1967), Kanehira (1967), Leake (1968), Hashimoto (1973), Onuki and Tanaka (1974), Onuki *et al.* (1976) and Onuki (unpublished). The data by EPMA are not utilized because of the uncertainty of Fe^{3+}/Fe^{2+} ratio in this paper.

Fig. 2. The relation between calcic subcalcic and alkali amphiboles. Rie=riebeckite, Ric=richterite, Ec-Arf=eckermannite-arfvedsonite. Other abbreviations are the same as in Fig. 1.

Fig. 3. The M4 site alkali versus the A site alkali diagram. Square: Subcalcic actinolite from glaucophanitic metamorphic terranes. Open circle: Subcalcic hornblende from the lowest grade part of the epidote amphibolite facies in glaucophanitic metamorphic terranes. Solid circle: That from the middle to high grade part of the epidote amphibolite facies in glaucophanitic metamorphic terranes. Half solid circle: That from the Yamagami epidote amphibolite. Small solid circle: That from amphibolite. X: That from diorite and syenite. Cross: That from granulite and nepheline gneiss. Open circle with X: "Magneisio-arfvedsonite" from Zone D of the Bessi-Ino district (Banno, 1964). Subscriptions C, D and E represent Zones C, D and E of the Bessi-Ino district (Banno, 1964). Abbreviations are the same as in Fig. 2.

The solid solution of subcalcic hornblendes may be characterized by the substitutions of $*Ca \rightleftharpoons NaNa$ and $*NaSi \rightleftharpoons *CaAl$ on calciferous hornblendes, so that we describe the relation between the alkali contents of the vacant site A and M_4 site. The star * indicates the vacant site of amphiboles. The graphical relation between the amphiboles is indicated in Fig. 2 by means of Tagiri and Onuki's method (Tagiri and Onuki, 1974). Fig. 3 shows the tschermakite-pargasite-glaucophane section selected from Fig. 2. The end-members of tschermakite, pargasite, glaucophane-riebeckite, katophorite and eckermannite-arfvedsonite are shown in this diagram, and those of tremolite, edenite, and richterite exist in the background of this plane. The forty-two data noted above and the twelve data of this paper are plotted in Fig. 3 together with the compositions of three subcalcic actinolites. Subcalcic hornblendes in the Yamagami epidote amphibolites are situated in the same field as Zone D of the Bessi-Ino district (Banno, 1964). Those from the lowest grade part of the epidote amphibolite facies are relatively poor in the alkali content of the vacant site and rich in that of the M_4 site. Those from the higher grade part of the epidote amphibolite facies have a wide variation. The alkali content of the M_4 site in metamorphic calcic hornblendes are less than about 0.5, in general.

Two compositional variation trends of hornblendes are generally supposed during glaucophanitic metamorphism by many authors. Black (1973) and Toriumi (1975) indicated that both tschermakite and glaucophane molecules in calciferous amphibole increase with rising temperature under constant P_{H_2O} at lower temperatures. Banno (1964) and Miyashiro (1973) reported that subcalcic hornblendes transform to

calcic hornblende with increasing temperature under the conditions of the epidote amphibolite facies. We name the former Trend A and the latter Trend B. In Trend A, the alkali content of the M_4 site increases, and in Trend B, that of the M_4 site decreases and that of the vacant site slightly increases with rising temperature. In Fig. 3, subcalcic hornblendes from the lowest grade part of the epidote amphibolite facies which corresponds to Zone C of the Bessi-Ino district are higher in $(Na + K \text{ in } M_4)/(Na + K \text{ in } A)$ ratio than those from the higher grade part of the epidote amphibolite facies with some exceptions. Calcic hornblendes also appear commonly in the epidote amphibolite facies. Trend A does not appear in Fig. 3 because of the scarcity of the wet chemical analyses on actinolites and subcalcic actinolites, but Trend A is clearly shown by Black (1973) and Toriumi (1975). Their contributions suggest that Trend A probably continues until glaucophane becomes unstable at higher temperatures and is succeeded by Trend B.

On the other hand, subcalcic hornblendes in garnet-clinopyroxene bearing epidote amphibolites (so-called hornblende eclogites) have the wide range of both alkali contents of the M_4 site and the vacant site. Some subcalcic hornblendes in "hornblende eclogites" equal those of the lowest grade part of the epidote amphibolite facies in maximum glaucophane molecular content. If subcalcic hornblendes in "hornblende eclogites" stably associate with clinopyroxenes and garnets, as they are soluble in the same alkali content of the M_4 site as those from the lowest grade part of the epidote amphibolite facies under "hornblende eclogite" conditions of the epidote amphibolite facies, it is assumed that Trend

B is due to the effect of bulk composition on the solid solution of subcalcic hornblende. For example, "magnesian arfvedsonites" occur from the albite-quartzose schists of Zone D in the Bessi-Ino district (Banno, 1964). The chemical property of two "magnesian arfvedsonite" resemble closely the intermediate between katophorite and subcalcic hornblende solid solution than the ideal arfvedsonite $(Na Na_2 Fe^{2+} Fe^{3+} Si_3 O_{22} (OH)_2)$. The alkali content of the M_4 site and vacant site in them are 0.139 and 0.877, and 0.799 and 0.410, respectively. These two "magnesian arfvedsonite" are also plotted in Fig. 3.

Subcalcic hornblendes from amphibolites, granulites, nepheline gneisses, syenites and diorites show a different property in composition. Their alkali contents of the M_4 site are relatively less than those from glaucophanitic metamorphic terranes and increase according as that of the vacant site increases, and subcalcic amphiboles approach katophorite-richterite. Allen *et al.* (1975) produced subcalcic hornblendes at 10–18 Kb and 880–1000°C with controlling oxygen fugacity. The alkali content of the M_4 site and vacant site in their subcalcic hornblendes is less about 0.66 and about 0.52, respectively. In their experiments, most subcalcic hornblendes crystallize at the starting material of andesitic composition, but not at tholeiitic, alkali basaltic and nephelinite compositions.

There is nothing that the alkali content of the M_4 site in subcalcic hornblende is over about 1. A large miscibility gap exists between glaucophane and tschermakite-pargasite (Klein, 1969; Black, 1973; Toriumi, 1974).

RECRYSTALLIZATION OF SUBCALCIC HORNBLENDES

When epidotes exist in the rocks, subcalcic hornblende bearing mineral assemblages are shown in the tetrahedral system of $1/2\text{Na}_2\text{O}-\text{CaO}-(\text{MgO}+\text{FeO})-\text{SiO}_2$, and each mineral is projected from epidote on the plane of $1/2\text{Na}_2\text{O}-(\text{MgO}+\text{FeO})-\text{SiO}_2$ (Fig. 4). Amphibole solid solution is also plotted in Fig. 4, which is contrasted with Fig. 3. Amphibole solid solution is divided into two parts by the tie line of albite and chlorite. One is named hornblende_c and the other hornblende_p. Hornblende_c is in the tetrahedron of albite-quartz-epidote-chlorite and hornblende_p is outside the tetrahedron. Subcalcic hornblende_c corresponds to that of the lowest grade part of the epidote amphibolite facies and subcalcic hornblende_p to that of the epidote amphibolite facies. Hornblende_c can recrystallize by means of the reaction among albite, epidote, quartz and chlorite. In contrast to hornblende_c, hornblende_p must be produced by breakdown reactions of some minerals in the tetrahedron of albite-epidote-quartz-chlorite or by reactions between some minerals outside the tetrahedron. Although the breakdown reaction of glaucophane-riebeckite makes subcalcic hornblende richer in alkali content, that seems to finish at the lowest grade part of the epidote amphibolite facies (Iwasaki, 1963; Banno, 1964).

Consequently, the compositional variation of subcalcic hornblende in the basic metamorphic rocks of glaucophanitic metamorphism is summarized as follows. In the closed system, the Na/Ca ratio of the remainder subtracted subcalcic hornblende molecule from the bulk composition decreases according as that of subcalcic horn-

Fig. 4. Projection from epidote onto $1/2 \text{Na}_2\text{O}-(\text{MgO}+\text{FeO})-\text{SiO}_2$ face of mineral compositions. Ab=albite, Ep=epidote, Qz=quartz, Chl=chlorite, Cpx=clinopyroxene, Gar=garnet. Other abbreviations are the same as in Fig. 1.

blende increases with rising temperature. After subcalcic hornblende reached the solvus between subcalcic hornblende and alkali amphibole, both the Na/Ca ratios of subcalcic hornblende and the remainder are fixed at the solvus. When hornblende progressively recrystallize still more, the five cases seem to take place according to the following reactions.

The first four cases accompany with some spending of albite, epidote, quartz and

chlorite. The fifth case is the breakdown of subcalcic hornblende_c. Albite or albite molecule in plagioclase and epidote usually exist in basic metamorphic rocks except some eclogite. When chlorite and quartz vanished or when they were spent all for the formation of garnet, reaction (1) is succeeded by reactions (2) and (3). Reactions (2) and (3) produce the decrease of the alkali content of the M₄ site and the increase of that of the vacant site. Accordingly, Trend B is due to the bulk composition, specially the scarcity of chlorite and quartz, in addition to metamorphic temperature. In "hornblende eclogite", albite are so less that reaction (1) is not replaced by reactions (2) and (3), and the alkali content of the M₄ site remain maximum. Reaction (5) probably proceeds under "hornblende eclogite" conditions of the epidote amphibolite facies. Fig. 4 shows that this subcalcic hornblende_D becomes much richer in alkali content than subcalcic hornblende_c of reaction (5). The chemical property of this subcalcic hornblende_D is compatible with those from "hornblende eclogites".

As the chemical compositions of the Yamagami epidote amphibolites are about the same as those of the rocks described by Banno (1964) from the Bessi-Ino district, subcalcic hornblende compositions indicate that the Yamagami epidote amphibolites were recrystallized under the conditions of the middle to high grade part of the epidote amphibolite facies.

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北部阿武隈山地，山上変成岩類の緑れん石角閃岩中のサブカルシック角閃石

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12 個の緑れん石角閃岩およびそれらのサブカルシック角閃石の化学組成を示した。サブカルシック角閃石の固溶体をチエルマツク閃石，パーガス閃石，エツケルマン閃石，らん閃石系について示し，その生成反応を $\text{CaO}-1/2 \text{Na}_2\text{O}-(\text{MgO}+\text{FeO})-\text{SiO}_2$ 系で考察を行なった。らん閃石変成帯のサブカルシック角閃石固溶体の変化傾向には相対する二つがある。緑れん石角閃岩相の最低温部では M_4 サイトのアルカリが温度上昇とともに増加するが，高温部では M_4 サイトのアルカリが減少し，A サイトのアルカリが増大する。後者の変化傾向は複雑である。前者のサブカルシック角閃石はアルバイト，緑れん石，緑泥石，石英間の反応で生じ得るが，後者は生じ得ない。後者の変化傾向は変成温度と母岩の緑泥石，石英，アルバイトの含量に依存している。