

Fluorine-rich calcic amphiboles in ultrahigh-temperature mafic granulite from Tonagh Island in the Napier Complex, East Antarctica: Preliminary report

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Abstract: Fluorine-rich calcic amphiboles occur in ultrahigh-temperature mafic granulite from Tonagh Island in the Napier Complex, East Antarctica. The amphiboles are subdivided into two types: high-grade brownish to pale brownish amphibole and retrograde greenish amphibole. The brownish amphibole is pargasitic, occurring as a subhedral grain in two-pyroxene mafic granulite. It contains up to 2.2 wt% fluorine, which corresponds to an F/(F+Cl+OH) ratio of 0.54 (sample B98020801). In sample B98020802B, brownish amphibole is rimmed by an orthopyroxene-plagioclase-quartz corona, probably formed by a devolatilization reaction. It has a lower fluorine content of 0.21–0.52 wt% (F/(F+Cl+OH)=0.04–0.13). Pale brownish amphibole is a minor phase in metapyroxenite. It has the highest fluorine content (2.6 wt%, F/(F+Cl+OH)=0.60, sample B98012802F). Greenish amphibole is present as fine-grained aggregates with quartz, rimming ortho- and clinopyroxenes. Fluorine is almost absent in this amphibole.

Holloway and Ford (1975) experimentally determined that pargasite with a F/(F+Cl+OH) ratio of 0.48 is stable up to 1100°C at 5 kbar, and that F/(F+Cl+OH) increases with increasing temperature. The fluorine-rich calcic amphiboles in mafic granulites are therefore stable under UHT metamorphic conditions. The formation of an orthopyroxene-plagioclase-quartz corona in sample B98020802B can therefore be explained by breakdown of the amphibole due to its low-fluorine content. The origin of the fluorine is not known, but it is probably derived from basic magma at the stage of protolith formation.

key words fluorine-rich amphibole, mafic granulite, Tonagh Island, Archean Napier Complex, East Antarctica

1. Introduction

The Napier Complex of Enderby Land, East Antarctica, is an example of a late-Archean high-grade terrane which has undergone granulite-facies metamorphism at a temperature higher than 1100°C (*e.g.* Ellis, 1980, Harley, 1985, Sheraton *et al*, 1987, Hokada *et al*, 1999). The Amundsen Bay area, located in the western part of the Napier Complex, is known as the highest grade region of the complex where sapphirine-quartz and osumilite-bearing mineral assemblages are common (Sheraton *et al*, 1987, Harley and Hensen, 1990). The presence of an orthopyroxene-sillimanite-garnet-quartz assemblage in pelitic gneiss (Sheraton *et al*, 1987) and inverted pigeonite in meta-ironstone and mafic to intermediate granulites (Sandiford and Powell, 1986, Harley, 1987) provide additional evidence of ultrahigh-temperature (UHT) metamorphism.

In spite of the UHT metamorphic conditions of the Napier Complex, biotite is present as a common hydrous mineral in many lithologies including aluminous, quartzofeldspathic, mafic, and ultramafic granulites (*e.g.* Harley, 1985, Sheraton *et al*, 1987). Although most of the biotite is retrograde in origin, some biotite is regarded as a product of UHT metamorphism. Such high-grade biotites are characterized by their high fluorine contents up to 8 wt% (Motoyoshi, 1998). Experimental studies of fluorine-bearing biotite indicate that the stability field of biotite expands to higher temperature with increasing fluorine content in biotite (*e.g.* Peterson *et al*, 1991, Hensen and Osanaï, 1994).

Calcic amphibole is also known as a common hydrous mineral in UHT mafic and ultramafic granulites of the Napier Complex (Ellis and Green, 1985, Sheraton *et al*, 1987). Although most amphiboles are probably of retrograde origin, some formed during or prior to peak UHT metamorphism (Tsunogae *et al*, 1999, Owada *et al*, 1999). However, the fluorine contents of calcic amphiboles of the Napier Complex have not been reported before. Although fluorine-rich amphiboles have been identified elsewhere in hydrothermal veins (Damman, 1989) and contact metamorphic aureoles (Droll, 1983), those in high-grade rocks have not been discussed in detail. In this study we identified fluorine-rich calcic amphibole in mafic granulite from Tonagh Island. This is the first report of high-fluorine amphibole from the Napier Complex.

2. Geology of Tonagh Island

Detailed regional geological and structural characteristics of Tonagh Island are given in Osanaï *et al* (1999). Generally the island is composed of layered gneisses of various rock chemistry. Orthopyroxene-bearing quartzofeldspathic gneiss is the most abundant lithology in the area. It is commonly interlayered with mafic granulite, garnet-bearing felsic gneiss, and magnetite-quartz gneiss. Ultramafic granulite is present as thin layers or lenses in quartzofeldspathic and mafic granulites. Peak metamorphic conditions of Tonagh Island were determined by Hokada *et al* (1999) as $T > 1100^{\circ}\text{C}$ using ternary-feldspar equilibrium.

There are several isotopic ages from metamorphic rocks of Tonagh Island. The protolith age of granulites was determined to be 3800 Ma for mafic and ultramafic granulites by the Sm-Nd isochron method (Owada *et al*, 1994) and 3230–3260 Ma for orthopyroxene-bearing quartzofeldspathic gneiss by the SHRIMP zircon U-Pb method.

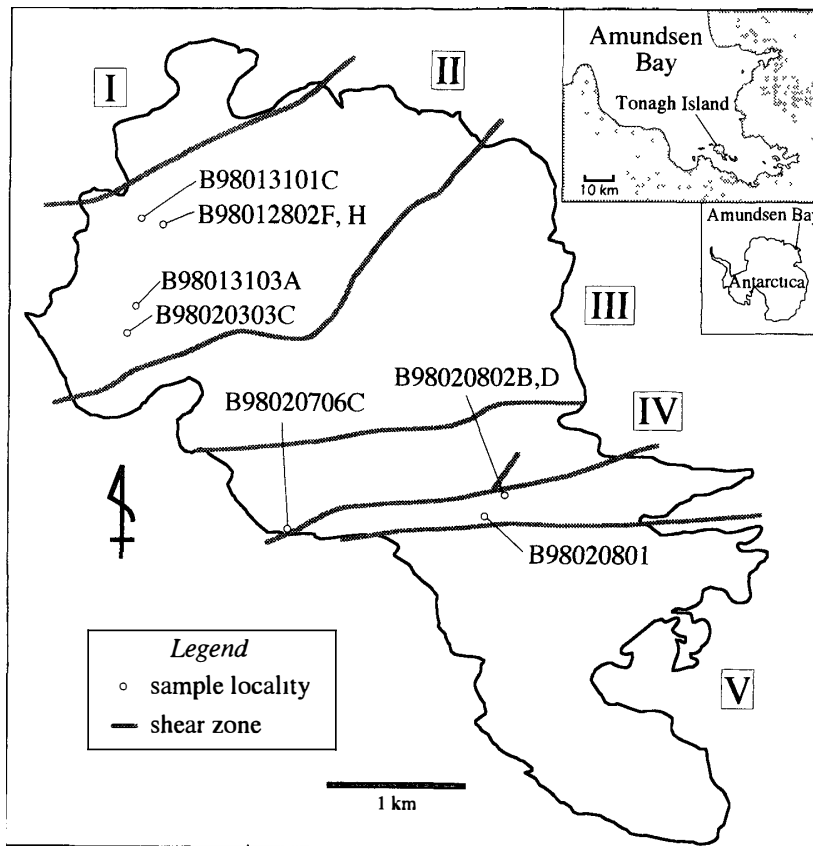


Fig 1 Location map of Tonagh Island with localities of representative samples of mafic granulite discussed in this study. I to V denote crustal units separated by shear zones after Osanai *et al.* (1999) and Toyoshima *et al.* (1999)

(Shiraishi *et al.*, 1997). A metamorphic age of *ca.* 2500 Ma was derived from whole-rock Sm-Nd isochrons of quartzofeldspathic gneisses and ultramafic granulites by Owada *et al.* (1994) and from metamorphic overgrowth of zircon around magmatic crystals by Shiraishi *et al.* (1997).

The high-grade rocks from Tonagh Island are subdivided into five units (Units I to V) by high-angle and ENE-WSW to E-W trending shear zones (Osanai *et al.*, 1999; Toyoshima *et al.*, 1999). Approximate positions of the shear zones are shown in Fig. 1. Unit I corresponds to the northwestern portion of the island. There occur layered gneisses which vary in thickness from several to several tens of centimeters. Thick layers of mafic granulite (up to 100 m in thickness) are present in Units II, III, and IV. Unit V is different from the other units because of the more common occurrences of orthopyroxene- and garnet-bearing quartzofeldspathic gneisses.

3. Petrography and mineral chemistry of calcic amphiboles

3.1. Analytical procedure

Chemical analyses of minerals were performed by an electron microprobe analyzer at the Research Center of Coastal Lagoon Environments, Shimane University, using an automated JEOL JXA-8800 instrument. The data were obtained under conditions of 15

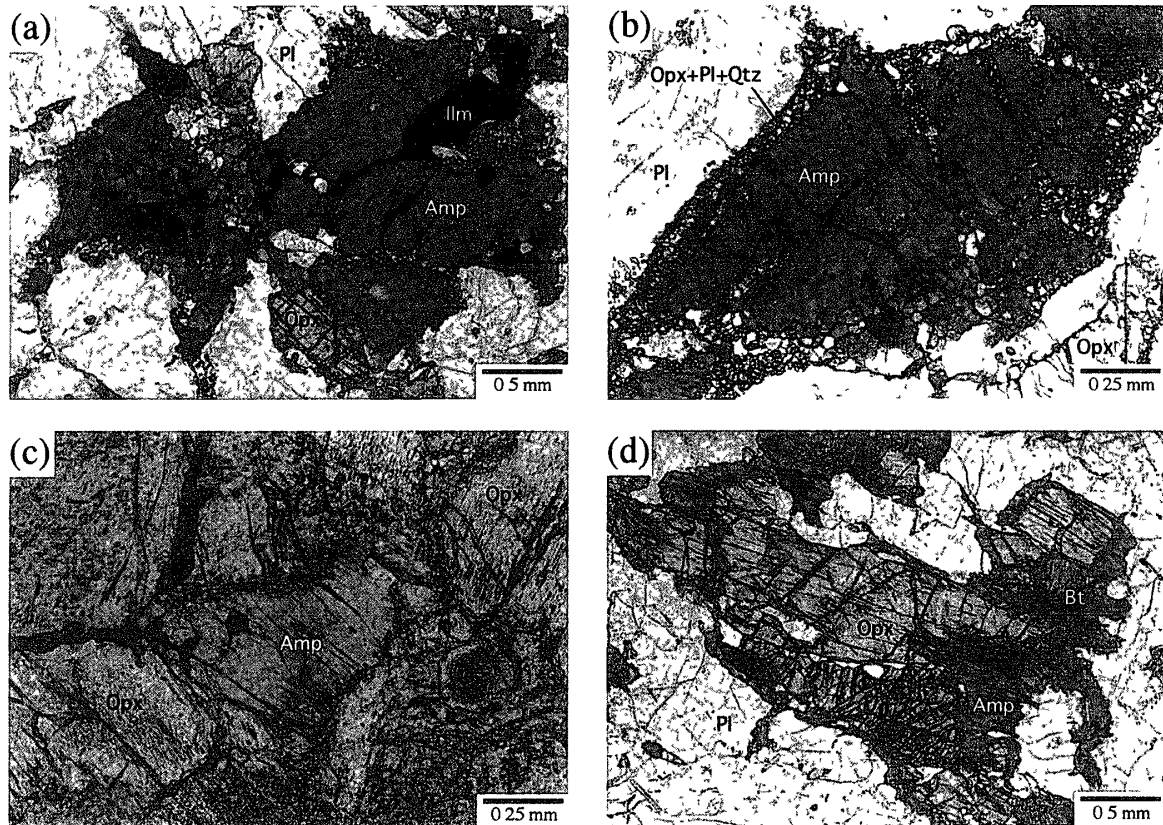


Fig 2 Photomicrographs of calcic amphibole-bearing mafic granulites and metapyroxenite. All photographs are under polarized light. (a) Brownish amphibole in sample B98020801 (type 2). It coexists with orthopyroxene, clinopyroxene, plagioclase, and quartz. (b) Brownish amphibole in sample B98020802B (type 2). It is rimmed by orthopyroxene-quartz-plagioclase corona which was probably formed by a devolatilization reaction (1). Ilmenite lamella is present in the amphibole. (c) Pale brownish amphibole in type 6 metapyroxenite (sample B98012802F). The rock has a granoblastic texture and is composed of orthopyroxene and minor ilmenite, calcic amphibole, and clinopyroxene. Amphibole is present along grain boundaries of pyroxenes. (d) Greenish amphibole-quartz symplectite around orthopyroxene in type 3 mafic granulite (sample B98012802H). Biotite is also present as a hydrous phase around the pyroxene. The texture is evidence of retrograde hydration by infiltration of water-bearing fluid.

kV accelerating voltage and 20 nA sample current, using data processing by an Oxide-ZAF model correction program supplied by JEOL.

For fluorine analysis, pure fluorite (CaF_2) was adopted as a standard sample, using the $\text{F-K}\alpha$ line reflected by a TAP crystal. Although the $\text{Fe-L}\alpha$ line might possibly interfere with the $\text{F-K}\alpha$ line, its effect was completely eliminated. As discussed below, the fluorine content of green amphiboles (*ex* sample B98012802H) is therefore nearly zero even if they have high Fe content ($\text{Fe}/(\text{Mg} + \text{Fe}) > 0.5$). The count of the $\text{F-K}\alpha$ line is very low (less than 120 cps) counting the time of the peak and backgrounds are set as 300 and 150 s, respectively. Accuracy of the fluorine analysis was examined by ten repeated analyses of fluorite crystal. The result was 48.46 ± 0.39 wt%, which is close to the known fluorine content of pure fluorite (48.67 wt%). The error corresponds to the standard deviation of the result. Ten repeated analyses of a single spot of amphibole in sample B98020801

Table 1 Representative electron microprobe analyses of calcic amphibole in mafic granulite

Sample No Rock type Color	amphibole (number of oxygens = 23)									
	B98020801	B98020801	B98020802B	B98020802B	B98013101C	B98012802H	B98020706C	B98020303C	B98012802F	B98013103A
	2	2	2	2	3	3	3	4	6	6
	brownish				greenish				pale brownish	
	core	rim	core	rim	core	core	core	core	core	core
SiO ₂	40.78	41.68	40.23	40.29	46.69	42.88	42.82	43.00	43.87	43.20
Al ₂ O ₃	11.22	10.98	12.96	13.52	9.69	12.73	11.84	12.14	12.52	13.39
TiO ₂	1.56	1.86	2.37	1.85	0.56	1.22	0.97	1.03	0.77	1.42
Cr ₂ O ₃	0.00	0.00	0.24	0.26	0.00	0.05	0.03	0.00	0.36	0.00
FeO*	15.38	15.37	14.64	15.35	15.09	17.20	17.72	15.76	4.74	4.99
MnO	0.08	0.05	0.09	0.13	0.23	0.14	0.12	0.12	0.02	0.03
MgO	11.39	11.05	10.21	9.91	12.34	9.82	9.39	10.64	17.71	17.26
CaO	11.70	11.67	11.35	11.39	11.74	11.55	11.44	11.84	13.34	12.68
Na ₂ O	1.82	1.61	1.28	1.25	1.09	1.17	1.54	1.14	1.99	2.04
K ₂ O	2.64	2.71	3.10	3.16	0.46	1.34	1.20	1.76	1.78	2.02
F	2.05	1.93	0.47	0.40	0.00	0.01	0.03	0.12	2.58	1.73
Cl	0.26	0.26	1.44	1.45	0.01	0.00	0.10	0.23	0.02	0.01
Total	98.89	99.17	98.38	98.95	97.88	98.11	97.21	97.78	99.70	98.77
Total***	97.97	98.30	97.86	98.46	97.88	98.10	97.17	97.68	98.61	98.04
Si	6.269	6.361	6.164	6.149	6.864	6.419	6.502	6.463	6.339	6.251
Al	2.032	1.974	2.339	2.432	1.678	2.246	2.119	2.150	2.132	2.283
Ti	0.180	0.213	0.273	0.212	0.061	0.137	0.111	0.116	0.083	0.154
Cr	0.000	0.000	0.029	0.031	0.000	0.006	0.004	0.000	0.041	0.000
Fe ²⁺	1.977	1.961	1.875	1.959	1.855	2.153	2.249	1.980	0.573	0.604
Mn	0.011	0.007	0.011	0.016	0.028	0.018	0.016	0.015	0.003	0.004
Mg	2.608	2.512	2.329	2.253	2.703	2.189	2.124	2.382	3.812	3.720
Ca	1.927	1.907	1.863	1.862	1.848	1.853	1.860	1.906	2.064	1.965
Na	0.542	0.477	0.381	0.369	0.309	0.339	0.452	0.331	0.557	0.573
K	0.517	0.528	0.605	0.615	0.086	0.257	0.232	0.337	0.327	0.373
Total	16.064	15.941	15.871	15.899	15.433	15.616	15.669	15.680	15.933	15.927
F	0.997	0.930	0.228	0.195	0.000	0.005	0.012	0.058	1.180	0.789
Cl	0.034	0.033	0.187	0.187	0.002	0.000	0.027	0.030	0.004	0.001
Mg/(Fe+Mg)	0.569	0.562	0.554	0.535	0.593	0.504	0.486	0.546	0.869	0.860
F/(F+Cl+OH)**	0.499	0.465	0.114	0.097	0.000	0.003	0.006	0.029	0.590	0.395
Cl/(F+Cl+OH)**	0.017	0.017	0.094	0.094	0.001	0.000	0.014	0.015	0.002	0.001

*Total Fe as FeO, **Calculated for stoichiometry of amphibole, ***Total correlated for F and Cl

showed its fluorine content to be 1.97 ± 0.09 wt%

For chlorine analysis, a granular aggregate of halite crystals (NaCl) was adopted as a standard sample, using the Cl-K α line reflected by a PET crystal. There is no significant interference with the Cl-K α line. Ten repeated analyses of a single spot of amphibole in sample B98020802B show its chlorine content to be 1.42 ± 0.03 wt%. Analytical totals were corrected for F and Cl in the way that the wt% of oxygen corresponding to the wt% of F and Cl was subtracted from the analytical total.

Calcic amphiboles in about 20 samples of amphibole-bearing mafic granulite from Tonagh Island were analyzed. Localities of representative samples in this study are shown in Fig. 1. Compositions of calcic amphibole in the samples are listed in Table 1.

3.2 Mineral assemblages of mafic granulite

Mafic granulites of Tonagh Island were subdivided into five rock types (types 1 to 5) on the basis of mineral assemblages. Type 6 is a sample of ultramafic granulite (metapyroxenite). Although there are some other types of ultramafic granulite on Tonagh Island, they are not discussed in this paper because they lack amphibole. Mineral abbreviations are after Kretz (1983). Minerals in italics are secondary phases.

Type 1 Pl + Opx + Cpx + Qtz (\pm Ilm, Bt)

Type 2 Pl + Opx + Cpx + Qtz + brownish Amp (\pm Ilm, Bt)

Type 3 Pl + Opx + Cpx + Qtz + *greenish* Amp (\pm Ilm, Bt)

Type 4 Pl + Opx + Cpx + Qtz + *greenish* Amp + Grt (\pm Ilm, Bt)

Type 5 Pl + Opx + Cpx + Qtz + Grt (\pm Ilm, Bt)

Type 6 Opx + Cpx + Amp (\pm Ilm, Pl, Bt)

Type 1 is an anhydrous mineral assemblage and is common in all units on Tonagh Island. Type 2 contains brownish amphibole with high fluorine as discussed below. They are present in Units I and IV. Type 3 is characterized by retrograde amphibole-quartz symplectite around ortho- and clinopyroxenes. Types 4 and 5 contain retrograde garnet around ortho- and clinopyroxenes, but Type 5 contains no retrograde amphibole. Types 3 and 4 are present in Units II, III, and IV, whereas type 5 is present in Units I and V. Type 6 is a metapyroxenite and is composed of granoblastic orthopyroxene, clinopyroxene and minor amphibole.

Calcic amphiboles discussed in this study are subdivided into two types: high-grade brownish to pale brownish amphibole and retrograde greenish amphibole. Brownish to pale brownish amphibole is present in granulites of types 2 and 6, whereas greenish amphibole is present in those of types 3 and 4. High-grade amphibole is present as a subhedral mineral and its grain size is the same as those of coexisting ortho- and clinopyroxenes. Retrograde amphibole is fine-grained and is present around pyroxenes as a secondary phase.

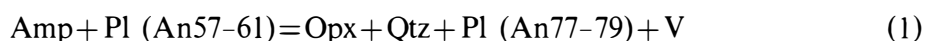
Although some samples were collected close to shear zones, there are few hydrated samples except sample B98020706C. Infiltration of retrograde H₂O-rich fluid which formed greenish amphibole is therefore regarded as a local event and not associated with a shear zone.

3.3. Brownish amphibole

Brownish amphibole is coarse-grained (up to 0.4 mm in sample B98020801) and

subhedral. Sample B98020801 comes from a body of mafic granulite more than 5 m in thickness within orthopyroxene-bearing quartzofeldspathic gneiss. It contains orthopyroxene, clinopyroxene, plagioclase, ilmenite, biotite, and apatite as well as brownish amphibole (Fig 2a). The amphibole is compositionally pargasite (Leake *et al.*, 1997) and characterized by high Na+K content at the A-site, up to 1.00 (based on 23 oxygens). The Ti content of the amphibole is high, ~0.20 pfu. It contains up to 2.2 wt% fluorine, which corresponds to F/(F+Cl+OH) ratio of 0.54.

Sample B98020802B is from a layer of mafic granulite 50–60 cm in thickness in orthopyroxene-bearing quartzofeldspathic gneiss. Although it shows a similar mineral assemblage to that of sample B98020801, brownish amphibole in this sample is rimmed by fine-grained aggregates of orthopyroxene, quartz, and plagioclase (Fig 2b). The composition of the product plagioclase (An_{77–79}) is slightly more An-rich compared with that of matrix plagioclase (An_{57–61}). The product fine-grained orthopyroxene has higher Al₂O₃ content (1.7 to 1.8 wt%) than the matrix (1.0 to 1.4 wt%). The texture suggests formation by a devolatilization reaction (1) of brownish amphibole



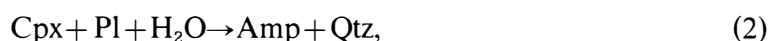
Composition of the brownish amphibole in the sample is similar to that of sample B98020801 except for its titanium, fluorine, and chlorine contents. It has the highest Ti content up to 0.28 at the core of the mineral, whereas it decreases toward the rim (Table 1). It has a lower fluorine content (0.21–0.52 wt%, F/(F+Cl+OH)=0.04–0.13) than that of sample B98020801, but has higher chlorine content of 1.2 to 1.5 wt% (Cl/(F+Cl+OH)=0.16–0.20).

Type 6 ultramafic granulite of sample B98012802F shows granoblastic texture of orthopyroxene and minor clinopyroxene and ilmenite. It contains a small amount of pale brownish amphibole along grain boundaries of pyroxenes (Fig 2c). The amphibole has a high Mg/(Fe+Mg) ratio (up to 0.88) probably due to a high-Mg bulk-rock chemistry. It shows the highest fluorine content up to 2.6 wt% which corresponds to F/(F+Cl+OH) ratio of 0.60. Chlorine content in the amphibole is very low.

Sample B98013103A is also a sample of type 6. It contains orthopyroxene, clinopyroxene, ilmenite, and pale brownish amphibole. As shown in Fig 3, the amphibole is slightly rich in Ti and poor in Si compared with that in sample B98012802F. Fluorine content of the amphibole varies from 1.8 to 2.0 wt% which corresponds to a F/(F+Cl+OH) ratio of 0.38–0.46. Chlorine content in the amphibole is also very low.

3.4. Greenish amphibole

This type of amphibole occurs as a secondary phase in type 4 and 5 mafic granulite. Sample B98012802H is composed of plagioclase, orthopyroxene, clinopyroxene, quartz, ilmenite, biotite, and greenish amphibole. The amphibole is present as fine-grained aggregates with quartz, rimming ortho- and clinopyroxenes (Fig 2d). In Fe-rich samples (*e.g.* sample B98020303C), retrograde garnet also occurs with the amphibole and quartz. The textures suggest retrograde infiltration of fluid and crystallization of greenish amphiboles by reactions (2) to (5)



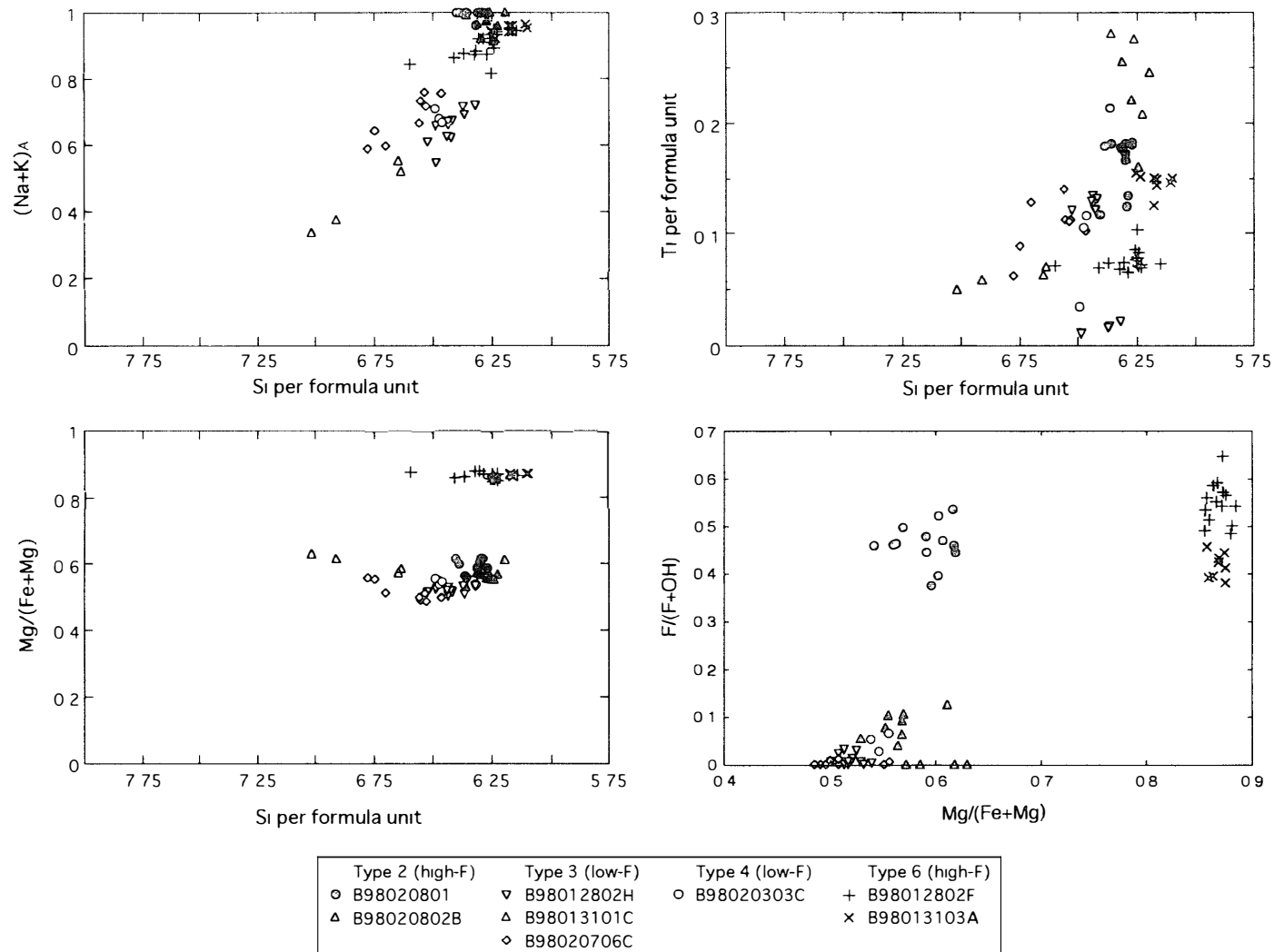
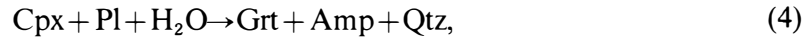
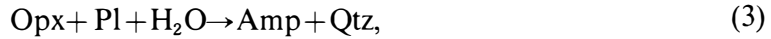


Fig 3 Compositional diagrams showing variety in composition of calcic amphiboles in mafic granulites of Tonagh Island



As shown in Fig 3, greenish amphiboles are classified as pargasite or hornblende. They are characterized by low-Na+K in the A-site and low Ti compared with brownish amphiboles. Generally the fluorine contents of greenish amphiboles formed by reactions (2) and (3) are low (less than 0.01 wt%, samples B98013101C, B98012802H, and B98020706C). Greenish amphibole formed by reactions (4) and (5) has a slightly higher fluorine content of 0.10–0.38 wt% (sample B98020303C). This is, however, still lower than that of brownish amphiboles. The F/(F+Cl+OH) ratio of the two types of greenish amphibole are <0.01 and 0.03–0.09, respectively. Their chlorine contents are very low (<0.1 wt%) except for sample B98020303C (0.23–0.31 wt%).

4. Discussion

As shown in Table 1, brownish amphiboles have higher fluorine content than greenish amphiboles. The low fluorine content of the latter and their occurrence as secondary phases indicate that some H₂O-bearing fluid was introduced during retrograde metamorphism. Chlorine content of the amphibole is very low except for sample B98020802B in which a devolatilization reaction (1) occurred.

Brownish amphiboles have a wide range in fluorine contents: 0.21 wt% (sample B98020802B) to 2.62 wt% (sample B98012802F). The cause of this variation is not known, but it is probably related to bulk rock chemistry. We therefore analyzed apatite coexisting with calcic amphibole, because it is the only mineral with a higher fluorine content than amphibole. As shown in Table 2, the fluorine content of apatite increases with increasing fluorine content in calcic amphibole in the same sample. Owada *et al.* (1999) pointed

Table 2 Representative electron microprobe analyses of apatite coexisting with calcic amphibole

	Apatite (number of oxygens=12.5)			
Sample No	B98020801	B98020802B	B98020303C	B98020706C
Rock type	2	2	4	3
CaO	53.37	53.83	54.13	54.10
P ₂ O ₅	41.72	41.80	42.72	42.11
F	6.90	6.01	5.97	3.78
Cl	0.05	0.74	0.12	1.01
Total	102.03	102.37	102.94	101.00
Total*	99.12	99.68	100.40	99.18
Ca	4.913	4.933	4.885	4.926
P	3.035	3.027	3.046	3.030
Total	7.948	7.960	7.931	7.956
F	1.875	1.625	1.589	1.017
Cl	0.007	0.107	0.017	0.145

*Total correlated for F and Cl

out, on the basis of euhedral crystal shape of apatite and its occurrence as inclusions in magmatic olivine in ultramafic granulite, that at least some apatites are igneous in origin. Higher fluorine content in apatite and calcic amphibole in sample B98020801 than those in sample B98020706C indicates that the fluorine content is controlled by bulk rock chemistry. The fluorine may therefore be derived from Archean basic magma at the stage of protolith formation. This implies a variation of fluorine content in the magma. However, further studies on fluorine contents in amphiboles and bulk rock are needed to fully understand the origin of fluorine.

Holloway and Ford (1975) experimentally investigated the stability of a pargasite of intermediate F/(F+Cl+OH) content ($\text{NaCa}_2\text{Mg}_4\text{Al}^{\text{VI}}\text{Si}_6\text{Al}_2^{\text{IV}}\text{O}_{22}[(\text{OH})_{0.57}\text{F}_{0.43}]_2$). Although their experiment should not be applied directly to a natural system, the results indicate that the fluorine-bearing pargasite is stable up to 1100°C at 5 kbar, and that the F/(F+Cl+OH) of amphibole increases with increasing temperature. The fluorine-rich calcic amphibole discussed in this study can therefore be regarded as a stable mineral during UHT metamorphic conditions. A devolatilization reaction of brownish amphibole in sample B98020802B (Fig 2b) occurred under UHT metamorphic conditions because of its lower fluorine content ($\text{F}/(\text{F}+\text{Cl}+\text{OH}) < 0.13$) compared with the other brownish amphiboles ($\text{F}/(\text{F}+\text{Cl}+\text{OH}) = 0.36\text{--}0.60$). Investigation of reaction textures and fluorine contents of amphibole is therefore useful to appraise the peak metamorphic temperature of UHT metamorphic complexes.

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