Högbomite from the Prince Olav Coast, East Antarctica: An example of oxidation-exsolution of a complex magnetite solid solution?

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Abstract

Högbomite occurs in upper amphibolite-facies quartz-andesine-biotite gneisses from a partially migmatized metamorphic complex at Sinnan Rocks and Cape Ryūgū, Prince Olav Coast, East Antarctica. Högbomite forms small grains (≤ 0.4 mm) along the margins of magnetite or, less commonly, enclosed in it, and it appears either to replace magnetite or to overgrow it. Högbomite is in direct contact with quartz, garnet, sillimanite, cordierite, anthophyllite, hercynite, rutile, and corundum; staurolite, kyanite, K-feldspar, cummingtonite, gedrite, and hornblende are also present. Hematite containing ilmenite exsolution lamellae is more abundant than independent (primary) ilmenite; most ilmenite is secondary. Minor secondary chlorite and muscovite are ubiquitous. Electron microprobe analyses of högbomite in nine samples yield 3.9–8.2 wt% TiO_2, 57–63 wt% Al_2O_3, 0–0.2 wt% Cr_2O_3, 18–25 wt% Fe (as FeO), 0.04–1.4 wt% MnO, 3.5–6.4 wt% MgO, and 2.7–11 wt% ZnO; in eight samples, Zn varies inversely with (Mg + Fe). The Antarctic högbomite could have formed by a process analogous to Buddington and Lindsley's oxidation-exsolution of titaniferous magnetite to ilmenite, whereby a magnetite containing Al, Zn, Mg, and Ti broke down under oxidizing conditions during a later metamorphic event to form högbomite. Associated rutile + magnetite and cordierite + corundum assemblages indicate $T \leq 500$ °C and $P_{H_2O} < P_{Total}$ for högbomite formation.

Introduction

Högbomite, a complex Fe-Mg-Al-Ti oxide related to the spinel group and once thought to be rare, has been reported from an increasing number of metamorphic parageneses, most recently summarized by Petersen et al. (1989). Nonetheless, the stability relations of högbomite are elusive. Högbomite defies easy analysis because of possible H_2O, variable Fe^{3+}-Fe^{2+} ratios, and compositional differences among stacking modifications (Petersen et al., 1989). With few exceptions (e.g., Moleva and Myasnikov, 1952; McKie, 1963) högbomites are fine-grained and are commonly intergrown with other phases. Textures suggest that högbomite formed under lower temperature and pressure conditions than those indicated by the primary silicate assemblages (e.g., Angus and Middleton, 1985; Grew et al., 1987).

We report new localities for högbomite, the first in Antarctica: Sinnan Rocks and Cape Ryūgū, Prince Olav Coast (44°30'–44°E), briefly described by Grew et al. (1989c). Högbomite occurs in upper amphibolite-facies quartzo-feldspathic gneisses and is intimately associated with magnetite. This association appears to constitute a distinct class of högbomite parageneses. The textural rela-

tions suggest that högbomite formed from Ti-Zn-Mg-Al-bearing magnetite at relatively low temperatures during a later metamorphic event by oxidation-exsolution of ilmenite from titaniferous magnetite (Buddington and Lindsley, 1964).

Geology and general petrology of Sinnan Rocks and Cape Ryūgū

Rocks exposed at Sinnan Rocks (68°56'S, 44°34'E) and at Cape Ryūgū, about 15 km to the west of Sinnan Rocks, on the Prince Olav Coast, are well-layered gneisses, migmatite, and amphibolite of the Lützow-Holm complex, part of the Precambrian Shield of East Antarctica (Nakai et al., 1980; Hiroi et al., 1983a, 1983b, 1983c; Shiraishi et al., 1987). These two exposures represent the lowest-grade portion of a metamorphic sequence 700 Ma in age ranging without break from the upper amphibolite facies to the granulite facies. Characteristic prograde assemblages at Sinnan Rocks and Cape Ryūgū include sillimanite + garnet + cordierite + biotite + plagioclase + quartz in pelitic rocks and cummingtonite + hornblende + biotite + plagioclase + quartz in intermediate to mafic rocks. The prograde metamorphic path is recorded by the widespread survival of relict kyanite and staurolite. The

TABLE 1. Mineral assemblages of the högbomite-bearing rocks

	Qtz	Pl*	Bt	Ms**	Chl**	Grt	Crd	St	Sil	Ky	Hbl	Ilm†	Ilm‡	Hem	Mag
Sinnan Rocks															
74021102	x	49	x	x	x	x	x	x	x	x	—	—	x	x	x
81020802B	x	x	x	x	x	x	x	—	x	—	—	—	—	x	x
81020910C	x	44	x	—	x	x	—	x	—	—	x	x	x	x	x
81020912	x	x	x	x	x	x	—	x	x	x	—	0	0	0	0
81021005A	x	44	x	x	x	x	—	—	x	x	—	—	x	x	x
81021005B	x	x	x	x	x	x	x	—	x	—	—	—	x	x	x
81021107	x	x	x	—	x	x	—	—	x	x	—	0	0	0	0
81021108	x	x	x	—	—	—	—	—	x	x	—	0	0	0	0
81021301	x	x	x	x	x	—	x	—	x	x	—	—	—	x	x
81021302B	—	x	x	x	—	—	—	—	x	—	—	0	0	0	0
81021401B	x	42	x	x	x	x	x	x	x	x	—	—	x	x	x
81S57	x	x	x	x	x	x	—	x	—	—	—	x	x	x	x
81S74B	x	x	x	x	x	x	—	—	x	x	—	—	x	x	x
81S80	x	x	x	x	x	x	—	—	x	x	—	—	x	x	x
81S84	x	x	x	x	x	—	x	—	x	x	—	—	x	x	x
81S86A	x	x	x	—	x	x	—	—	—	—	—	—	x	x	x
81S86B	x	x	x	x	x	x	—	—	—	—	x	—	x	x	x
81S88	x	x	x	—	—	x	—	—	x	x	—	x	x	x	x
Cape Ryūgū															
704A	x	49	x	x	x	x	x	x	x	x	—	x	x	x	x

Note: x = Present, 0 = unidentified opaques present in covered thin sections. Mineral abbreviations mostly from Kretz (1983): Qtz = quartz, Pl = plagioclase, Bt = biotite, Ms = muscovite, Chl = chlorite, Grt = garnet, Crd = cordierite, St = staurolite, Sil = sillimanite, Ky = kyanite, Ilm = ilmenite, Hem = hematite, Mag = magnetite, Rt = rutile, Hög = högbomite, Hc = hercynite, Mnz = monazite, Ap = apatite, Zrn = zircon, Ged = gedrite, Crn = corundum, Ath = anthophyllite, Cum = cummingtonite, Kfs = K-feldspar, Cal = calcite, Aln = allanite, Mrg = margarite.
* Anorthite content from microprobe analyses.
** Exclusively secondary.
† Apparently primary.
‡ As oxidation-exsolution lamellae in Mag, or other texture suggesting a secondary origin.
§ Mostly as blebs in Pl, some interstitial to Pl.

Lützow-Holm complex of the Prince Olav Coast was affected by a second metamorphic event (probably at 500 Ma) associated with emplacement of granite and pegmatite under lower-pressure conditions such that andalusite formed locally.

MINERALOGY AND PETROLOGY OF HÖGBOMITE-BEARING ROCKS

Högbomite was studied in 18 samples from Sinnan Rocks, 17 of which are quartz-plagioclase-biotite gneisses (the other lacks quartz) and one sample of quartz-plagioclase-biotite gneiss from Cape Ryūgū. Of these, 15 have been studied in polished thin sections (Table 1). Most of these rocks are pelitic and contain abundant garnet (typically as poikilitic porphyroblasts) with cordierite or sillimanite or both, whereas anthophyllite is the dominant ferromagnesian silicate in sample 81020910C (Hiroi et al., 1983b, 1983c).

Magnetite grains are commonly relatively coarse, euhedral or subhedral, and poikilitic (Fig. 1). Hematite forms relatively coarse grains with ilmenite exsolution lamellae. Ilmenite also forms lamellae in magnetite and margins around hematite grains where these are in contact with magnetite (secondary ilmenite in Table 1). Independent ilmenite grains and some ilmenite in magnetite, both textural types locally containing hematite lamellae, appear to be primary. Fine opaque grains included in garnet were identified as hematite and ilmenite in the polished sections.

Corundum occurs in 81021005B with högbomite, rutile, and cordierite (enclosing quartz) in an aggregate surrounded by magnetite containing secondary ilmenite, suggesting a local corundum-rutile-högbomite-cordierite-magnetite-ilmenite assemblage.

Högbomite forms small grains (up to 0.4 mm) along the margins of magnetite (Fig. 1) or less commonly, enclosed in magnetite, but only in very rare cases does it form grains isolated from magnetite, e.g., by a silicate mineral or ilmenite. Högbomite is in direct contact with all the major silicate minerals including quartz (Fig. 2A). Oxide-mineral assemblages inferred from direct contacts include högbomite-magnetite-ilmenite-hematite, högbomite-rutile-magnetite, and högbomite-magnetite-hercynite. No contacts were found with K-feldspar, staurolite (Fig. 2B), kyanite, or hornblende, probably because these minerals occur in only minor amounts. Most commonly, högbomite is in contact with plagioclase, muscovite, or chlorite (Fig. 2C), and only rarely is högbomite (with opaque oxide) enclosed in garnet (this paragenesis was noted in a covered section, so that identification of the opaque mineral was not possible). Högbomite grains in places appear to embay or even replace magnetite (Fig. 2B), whereas elsewhere it forms overgrowths on magnetite (Fig. 2C). Only a few grains of hercynite appear in any given section, and in general högbomite encloses this hercynite and appears to be replacing it (Fig. 2C). In addition, hercynite forms fine lamellar intergrowths with högbomite, similar to those reported by Petersen et al.

TABLE 1.—*Continued*

Rt	Hög	Hc	Mnz	Ap	Zrn	Other
			Sinnan Rocks			
x	x	x	x	x	x	Ged
x	x	—	x	—	x	Crn
x	x	x	—	x	—	Ath, Cum
—	x	—	x	x	—	—
x	x	x	x	x	x	—
x	x	—	x	x	—	Crn
—	x	x	—	x	x	—
—	x	(?)	x	x	x	—
—	x	—	x	x	x	—
x	x	x	x	x	x	Kfs§
(?)	x	x	—	x	—	—
x	x	—	x	x	x	Cal**
x	x	—	x	x	x	—
x	x	—	—	x	—	—
x	x	x	—	x	x	—
—	x	—	x	x	x	Aln
x	x	x	x	x	x	Aln, Mrg(?)
x	x	x	—	x	x	—
			Cape Ryūgū			
x	x	x	x	x	x	Kfs
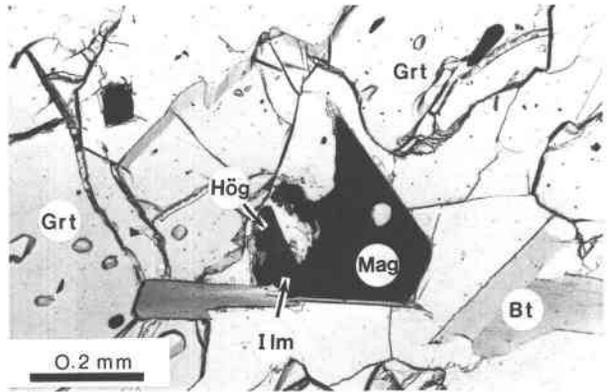
Fig. 1. Photomicrograph of Prince Olav Coast högbomite. Sample 81S88, plain light. Högbomite and ilmenite adjacent to euhedral magnetite near garnet in plagioclase and quartz. Abbreviations are in Table 1.

(1989), and occurs rarely as inclusions in garnet. In one section, högbomite and ilmenite form a selvage between hematite and magnetite. Like högbomite, rutile occurs as small grains next to, or, rarely, in magnetite (Fig. 2D). Inclusions of rutile are also found in garnet.

Except for högbomite + quartz, the assemblages with högbomite are typical of those reported previously (e.g., reviews by Grew et al., 1987; Petersen et al., 1989). Högbomite and quartz commonly occur in the same rock at other localities, but only Spry and Scott (1986) and Spry and Petersen (1989) explicitly described contacts between these two minerals.

MINERAL CHEMISTRY

Methods

Chemical analyses were carried out on the JEOL JXA 733 electron microprobe (wavelength-dispersive system) at the National Institute of Polar Research (NIPR) where data were processed by the Bence-Albee method and on a Hitachi S550 scanning electron microscope (energy-dispersive system) at Chiba University, where data were processed by a method developed by Mori and Kanehira (1984).

Twelve oxides (see Table 2, plus Na_2O) were analyzed on the NIPR microprobe, whereas the Chiba microprobe automatically determined which oxides were present in amounts greater than 0.1 wt% and analyzed only these. For each sample and microprobe, 1–4 grains of högbomite were analyzed at 1–14 spots per grain (4–19 analyses per section), while 1–4 grains of other minerals were analyzed at 1–8 spots per section, except staurolite (≤33 spots per section). As a rule, the same högbomite grains were analyzed by both microprobes. In most cases, the analyzed minerals lie inside an area 0.5 to 2 cm across.

Comparison of analytical data from the Chiba and NIPR microprobes reveals systematic differences for Al_2O_3 and SiO_2, but good agreement for TiO_2, total Fe (as FeO), MnO (staurolite), MgO (except 704A), and ZnO (Table 2). The poorer agreement and lower precision in the data for sample 704A could be the result of compositional heterogeneity (see below).

Högbomite

Analyses of högbomite in nine samples yield 3.9–8.2 wt% TiO_2, 57–63 wt% Al_2O_3, 0–0.2 wt% Cr_2O_3, 18–25 wt% Fe (as FeO), 0.04–1.4 wt% MnO, 3.5–6.4 wt% MgO, 2.7–11 wt% ZnO, and negligible NiO (≤0.1 wt%) (selected analyses listed in Table 3). Ni, Sn, and Co were not detected in a computerized element scan on the NIPR microprobe (the detection limit is a few tenths of one percent) of högbomite in 74021102, 81021005A, 81021401B, and 81S57. The reported values for CaO and K_2O in the NIPR analyses are interpreted to be negligible, whereas Na_2O values were discarded, as they are undoubtedly due to interference from Zn $L\alpha$. A manual element scan and detailed analysis for which the detection limit is estimated to be 0.001 wt% (10 ppm) was performed on a högbomite in sample 81021005A. These analyses revealed minor CoO and traces of SnO_2, NiO, and possibly CaO (Table 3). The (Zn + Mn) contents of the Prince Olav Coast högbomites (Table 3) are sufficiently high to lie in the range for högbomites from massive sulfide deposits (Spry and Petersen, 1989).

In a given sample from Sinnan Rocks, högbomite Ti and Al contents are more or less constant within a grain, whereas Zn varies inversely with (Fe + Mg) (Fig. 3). In the Cape Ryūgū sample Al varies inversely with Ti and with total Fe (Fig. 4), or possibly Al varies with Fe^{3+} only.

There is no agreement as to how to calculate a högbomite formula including the Fe^{3+}-Fe^{2+} ratio because so many factors are unknown, most importantly structure type, hydroxyl content, and the proportion of cation vacancies. Only one högbomite specimen has been studied in detail crystallographically: Gatehouse and Grey (1982) refined the structure of an $8H$ polytype and reported full
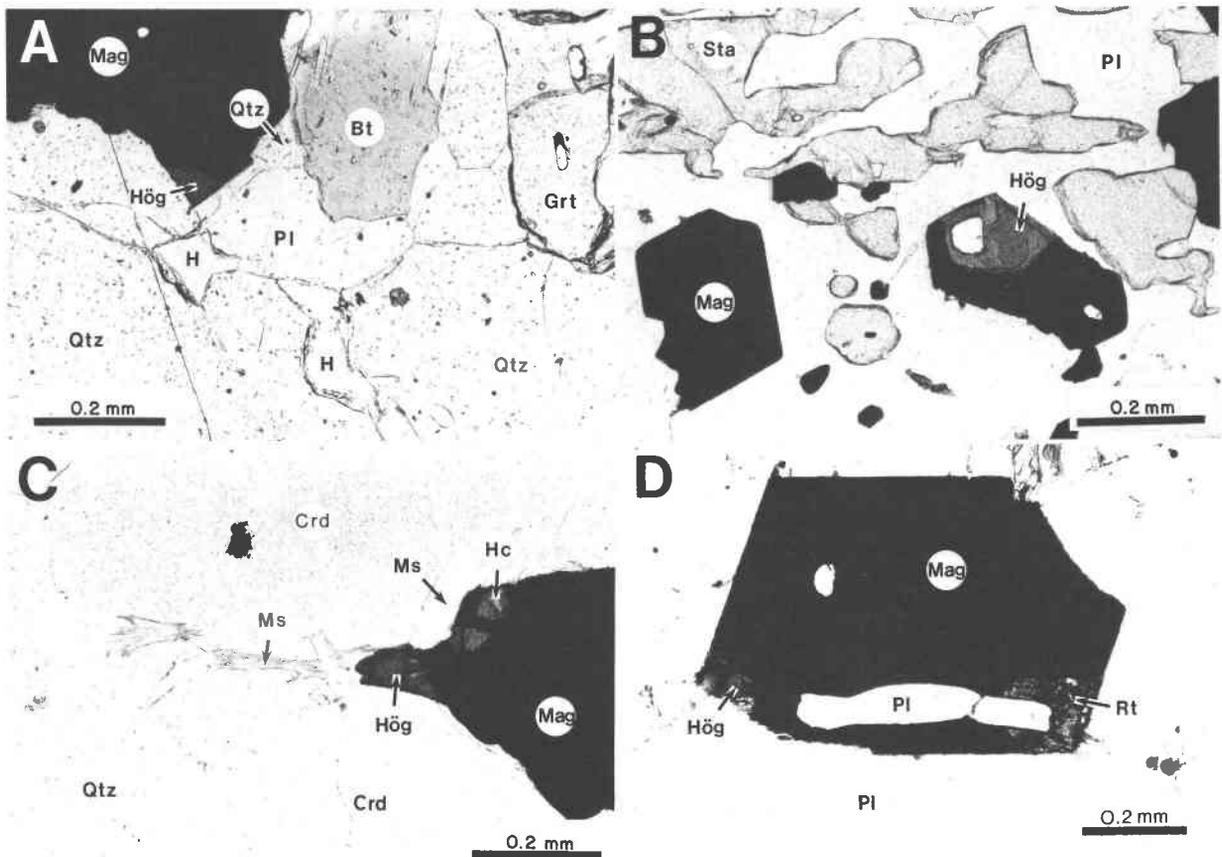
Fig. 2.   Photomicrographs of Prince Olav Coast högbomite. (A) Sample 81021401B, plain light. Högbomite and magnetite are in direct contact with quartz, which separates them from biotite and plagioclase. H = holes in the section. Quartz identification confirmed by electron microprobe. (B) Sample 74021102, plain light. Högbomite, euhedral magnetite, staurolite in plagioclase. Högbomite does not touch staurolite, but there is no evidence of a reaction between them. (C) Sample 81020802B, plain light. Högbomite (darker, Hög), which encloses hercynite (lighter, Hc) and muscovite around magnetite in cordierite. (D) Sample 81021005A, plain light. Högbomite, rutile, and magnetite in plagioclase. Abbreviations are in Table 1.

occupancy of 22 cations and an anion composition of (30 O + 2 OH). If the högbomites in Table 3 are assumed to have 22 cations, 31 oxygens (30 O + 2 OH), then calculated Fe^{3+} contents are 1.208–1.682 and Fe^{3+}-Fe ratios are 0.28–0.48. Such values are substantially higher than Fe^{3+}-Fe ratios characteristic of spinel-hercynite and ferromagnesian silicates, but similar to the 0.34 Fe^{3+}-Fe ratio Gatehouse and Grey (1982) calculated for their högbomite. Because Fe varies inversely with Mg and Zn, we infer that variations in Fe are largely due to variations in Fe^{2+}. In addition, the associated Ti-poor magnetite + titaniferous hematite ± ferrian-ilmenite assemblage would have buffered the Fe^{3+}-Fe^{2+} ratio of högbomite to relatively high values, thereby lending credence to the substantial Fe^{3+} contents estimated from stoichiometry.

Another indication of the relatively oxidizing conditions for högbomite formation are the Fe_2O_3 contents of Al_2SiO_5, ranging from 0.68 to 1.65 wt% in sillimanite and 1.14–1.19 wt% in kyanite, values characteristic of moderately to highly oxidized metapelites (Grew, 1980). Corundum Fe_2O_3 contents are similar at 0.75–1.13 wt% (sample 81021005B). If Fe^{3+} contents are relatively constant from sample to sample, variations of Mg-Fe, Mn-Fe, Zn-Fe would follow, to a first approximation, variations in the corresponding ratios with Fe^{2+}.

Ilmenite and hematite

Relict ilmenite enclosed in garnet is ferrian and Mn-poor (Table 4). In contrast, secondary ilmenite, which is contiguous to magnetite or forming lamellae in it, contains 1.9–18 wt% MnO (Table 4), while element scans of ilmenite lamellae in hematite of samples 81021005B and 81S57 indicate substantial MnO contents in these ilmenites as well. Hematite hosts in samples 74021102, 81020802, and 81S57 contain less than 1 wt% MnO, suggesting that MnO was concentrated in ilmenite as it exsolved from hematite. Measured ZnO contents in ilmenite and hematite range from 0 to 0.16 wt%, values not significantly above background.

TABLE 2. Comparison of analyses on the Chiba University (CU) and National Institute of Polar Research (NIPR) microprobes (in wt%)

Sample	74021102		81021401B		704A	
Mineral	Staurolite		Högbomite			
Probe	CU(6)	NIPR(2)	CU(7)	NIPR(5)	CU(7)	NIPR(4)
SiO_2	27.10(0.17)	25.90(0.04)	—	—	—	—
TiO_2	0.57(0.03)	0.61(0)	5.23(0.15)	5.17(0.12)	4.82(0.28)	5.14(0.24)
Al_2O_3	54.87(0.21)	53.92(0.32)	60.97(0.25)	59.75(0.53)	61.11(0.87)	59.25(0.60)
Cr_2O_3	0.26(0.11)	0.24(0.01)	—	0.04(0.04)	—	0.04(0.05)
FeO	10.24(0.10)	10.21(0.71)	21.18(0.24)	20.68(0.61)	22.38(0.67)	22.83(0.56)
MnO	1.52(0.05)	1.58(0.27)	0.29(0.03)	0.19(0.03)	0.20(0.03)	0.13(0.07)
MgO	3.49(0.08)	3.42(0.08)	4.01(0.11)	4.03(0.15)	5.73(0.26)	5.04(0.36)
ZnO	1.35(0.08)	1.28(0.08)	6.89(0.45)	7.02(0.54)	4.21(0.32)	4.01(0.14)
NiO	—	0.05(0.10)	—	0.06(0.03)	—	0.02(0.02)
CaO	—	0.01(0.01)	—	0.02(0.01)	—	0.01(0.01)
K_2O	—	0.01(0.02)	—	0.01(0.01)	—	0.02(0.03)

Note: Numbers in parentheses are standard deviations; where there are only two measurements, the spread of values is given. Staurolite: from one analytical area; högbomite: same grain. N = number of analyses.

Rutile

Analyses of rutile in sample 81021005B gave 98.3–98.9 wt% TiO_2 and 0.5–0.8 wt% FeO. Element scans in samples 81S57 and 81021005A showed no evidence for Nb, a potentially major impurity in metamorphic rutile (Rumble, 1976; Grew et al., 1987).

Magnetite and hercynite

Magnetite is essentially pure Fe_3O_4 (e.g., Table 5). Maximum values of impurities are TiO_2, 0.15 wt%; Cr_2O_3, 0.26 wt%; Al_2O_3, 0.36 wt% (NIPR probe); V_2O_3, 0.45 wt% (Chiba probe) and MnO, 0.67–0.86 wt% (in sample 81021301, Chiba probe). However, the significance of

TABLE 3. Selected individual analyses of högbomite

| Sample no. | 74021102 | 81020802B | 81020910C | 81021005A* | 81021005B | 81021301 | 81021401B | 81S57 | 704A | |
Probe	NIPR	NIPR	NIPR	NIPR	CU	CU	NIPR	CU	CU	CU
					Wt%					
SiO_2	0.02	0	0	0	—	—	0	—	—	—
TiO_2	5.14	4.45	5.81	5.31	5.00	4.61	5.37	5.65	4.74	7.41
SnO_2	—	—	—	0.008(0.3)	—	—	—	—	—	—
Al_2O_3	60.10	59.51	59.81	59.16	60.26	60.75	60.32	60.85	61.25	58.53
Cr_2O_3	0.09	0.07	0	0	—	0.05	0.01	0.06	—	—
FeO	18.38	19.81	20.17	21.89	23.01	19.14	21.58	22.13	22.72	24.78
MnO	1.28	0.94	0.34	0.58	0.76	1.50	0.23	0.69	0.19	0.26
MgO	4.72	4.08	5.67	4.12	3.91	3.75	4.11	4.88	5.62	5.00
ZnO	8.92	9.05	4.97	5.72	5.38	9.44	6.28	5.17	4.12	2.74
NiO	0.01	0	0.02	0.009(1)	—	—	0.04	—	—	—
CoO	—	—	—	0.072(2)	—	—	—	—	—	—
CaO	0	0	0.01	0.001	—	—	0.02	—	—	—
K_2O	0	0	0.01	0	—	—	0.02	—	—	—
H_2O(calc)	1.43	1.41	1.42	1.40	1.42	1.43	1.42	1.45	1.45	1.43
Total	100.09	99.32	98.23	98.27	99.74	100.67	99.40	100.88	100.09	100.15
				Formulae for 22 cations						
Ti	0.811	0.711	0.922	0.853	0.792	0.727	0.853	0.880	0.738	1.168
Sn	—	—	—	0.001	—	—	—	—	—	—
Al	14.863	14.885	14.891	14.901	14.956	15.010	15.020	14.857	14.935	14.457
Cr	0.015	0.011	0	0	0	0.008	0	0.010	—	—
Fe	3.225	3.516	3.564	3.913	4.052	3.356	3.813	3.834	3.931	4.343
Mn	0.228	0.168	0.061	0.105	0.136	0.266	0.042	0.121	0.033	0.046
Mg	1.476	1.290	1.786	1.311	1.227	1.172	1.293	1.507	1.733	1.562
Zn	1.382	1.419	0.775	0.903	0.836	1.461	0.980	0.791	0.629	0.424
Ni	—	—	—	0.002	—	—	—	—	—	—
Co	—	—	—	0.012	—	—	—	—	—	—
O	30.250	30.159	30.368	30.304	30.270	30.236	30.363	30.314	30.205	30.396
X_{Fe}	0.69	0.73	0.67	0.75	0.77	0.74	0.75	0.72	0.69	0.74

Note: Values <0.05 wt% not considered significant (except 81021005A). H_2O calculated assuming 2 OH per 22 cations (Gatehouse and Grey, 1982). X_{Fe} = Total Fe/(Total Fe + Mg).
* Analysis combines a point analysis with averages and standard deviations (in parentheses) obtained by Y. Kondo on the same grain at four points, with ranges in wt% as follows: SnO_2, 0.004–0.010; NiO, 0.008–0.010; CoO, 0.070–0.074.
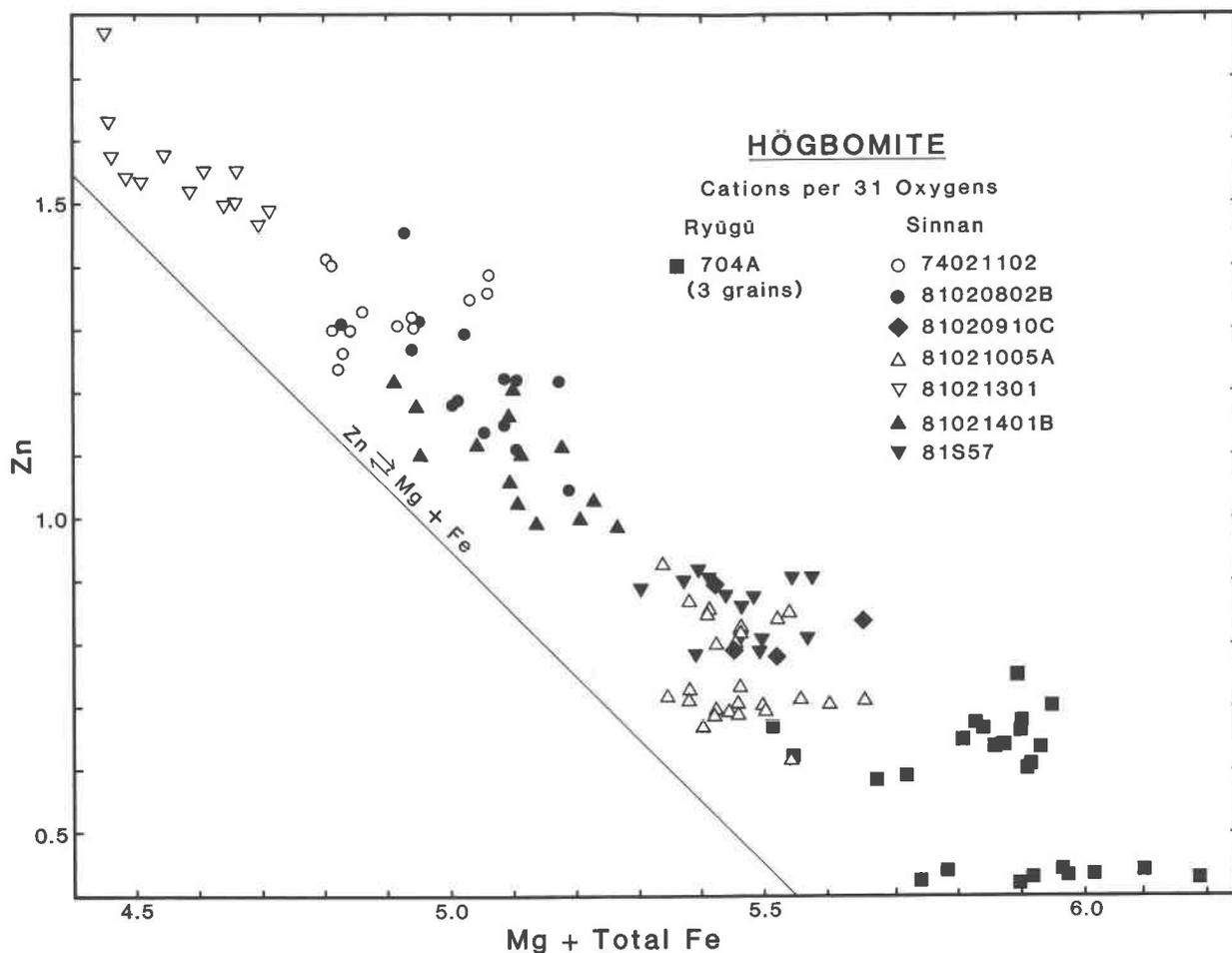
Fig. 3. Plot of Zn vs. Mg + total Fe in individual analyses of högbomite from Sinnan Rocks and Cape Ryūgū. Substitution line Zn ⇌ Mg + Fe is for reference only.

Al_2O_3 and MnO values less than 0.4 wt% is difficult to assess, because of the differences in MnO and Al_2O_3 values between the Chiba University and NIPR microprobes (Table 5). Few average values of TiO_2, Cr_2O_3, MgO, and NiO exceed 0.1 wt% and those of ZnO do not exceed 0.2 wt%; none of these appears to be significant.

Hercynite enclosed in garnet is richer in Fe^{3+} and poorer in Zn than secondary hercynite at the margin of a magnetite grain, a relationship analogous to that for Fe^{3+} in ilmenite from the same specimen (Table 4).

Ferromagnesian silicates

Relatively high MnO contents are characteristic not only of högbomite, but also of the ferromagnesian silicates in some of the samples, notably garnet (Table 6), staurolite (Tables 2, 7), and cordierite (Table 8). Staurolite in sample 74021102 contains 1.6 wt% MnO, equivalent to 0.36 Mn per formula unit of 46 oxygens. This is more than twice the maximum of 0.15 Mn among the 82 staurolites

reviewed by Griffen et al. (1982). The maximum MnO content of cordierite (1.09 wt% sample 81021301B, Table 8) is among the highest reported in metamorphic cordierite (Deer et al., 1986). Biotite alkali contents and analytical totals are low (Table 9). Hiroi et al. (1983a) reported compositional zoning in garnet and compositional heterogeneity in biotite in the Sinnan Rocks samples. Staurolite is also heterogenous, e.g., in sample 704A, ZnO = 0.73–1.07 wt%, MgO, 2.44–3.09 wt%.

Compositional relationships

Because the gneisses along the Prince Olav Coast have been affected by two metamorphic events (Hiroi et al., 1983a), equilibrium chemical relations attained during the earlier, higher-grade event were most likely disrupted during the second event, during which re-equilibration was probably not complete. Compositional zoning and heterogeneity, particularly the marked differences in composition between relict and secondary hercynite and
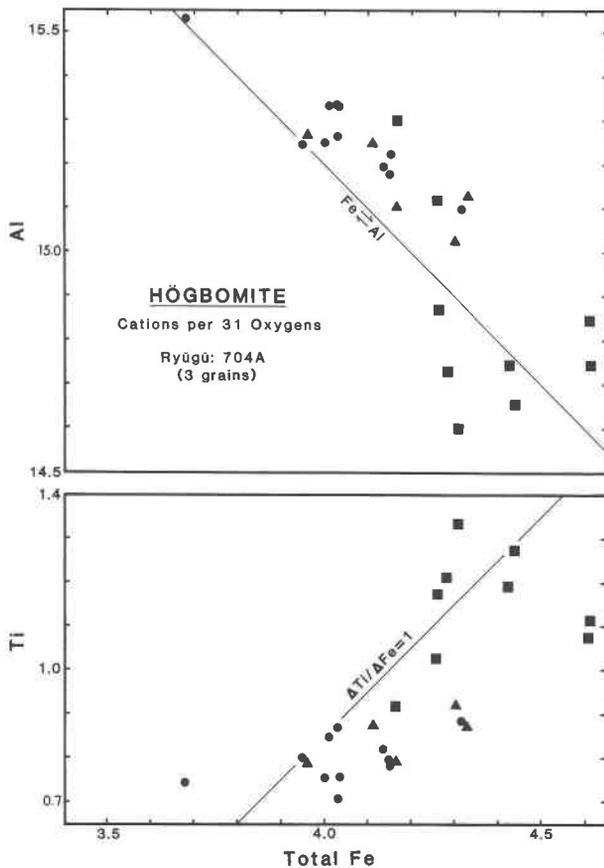
Fig. 4. Plot of individual analyses of högbomite on three grains in sample 704A, Cape Ryūgū. Lines for Al-Fe and Ti-Fe = 1 are for reference only.

Fig. 5. Plot of atom ratio Mg-Fe in associated phases in the Sinnan Rocks and Cape Ryūgū (704A only) samples (adapted from Albee, 1968, Fig. 25-3). Samples are listed in increasing Mg-Fe ratio of högbomite. Length of horizontal bar is the range of measured ratios in a given sample; vertical bar is the midpoint; filled circles are individual analyses. Equal partition coefficients are represented by equal distances between these midpoints, and lines joining these would be parallel if minerals crystallize in chemical equilibrium. The major-silicate assemblages are as follows: Qtz-Bt-Pl-Grt-Sil-Crd except samples 81021301 (lacks Grt), 81021005A (lacks Crd) and 81020910C, which is Qtz-Bt-Pl-Ath-Grt±Hbl±Cum (Table 1).

between relict and secondary ilmenite, are clearcut evidence for such incomplete re-equilibrium.

In order to further assess the extent of this disruption and possible re-equilibration, we have plotted Mg-Fe, Mn-Fe, and Zn-Fe ratios of coexisting ferromagnesian minerals in semilog plots (Figs. 5–7), a format devised by Albee (1968). Chemical equilibrium would be indicated by compositional homogeneity (a small range of values for a given ratio in each mineral) and covariation of Mg-Fe, Mn-Fe, and Zn-Fe ratios for each mineral and among the minerals from sample to sample in a given assemblage (Albee, 1968). Even allowing for differences in the mineral assemblages, our data show marked deviations from these equilibrium trends, for example, inverse correlation of högbomite and cordierite Mg-Fe ratios (Fig. 5) and inverse correlation of Mn-Fe and Mg-Fe ratios in högbomite (Figs. 5 and 6). The concordant trends in staurolite composition suggest that staurolite may have equilibrated during the earlier prograde event and equilibrium relations survived to a limited extent during the later event. Högbomite Zn-Fe and Mn-Fe ratios apparently adjusted to the corresponding ratios in associated stau-

rolite (Figs. 6–7), suggesting an approach to equilibrium during the later event in some small, localized areas.

The situation at Sinnan Rocks and Cape Ryūgū is thus similar to that at Molodezhnaya Station, 60 km to the east, where compositional zoning in garnet and heterogeneity in biotite, as well as nonequilibrium variations in Mn-Fe ratios of coexisting ferromagnesian minerals, are attributed to a second metamorphic event accompanying pegmatite emplacement at 500 to 550 Ma (Grew, 1981).

IMPLICATIONS FOR HÖGBOMITE PETROGENESIS

Högbomite crystallized late in the polymetamorphic evolution of the Sinnan Rocks–Cape Ryūgū area, most likely coevally with rutile, muscovite, and chlorite during the later, lower-grade event that is estimated to be 500 Ma. The close association of högbomite with magnetite suggests that a possible origin for the högbomite is external granule oxidation-exsolution from magnetite, that is, exsolution involving oxidation and limited migration of the exsolved material to the edge of the grain, a process proposed by Buddington and Lindsley (1964) for the formation of ilmenite granules in magnetite. An exsolution-like origin of högbomite from magnetite was also suggested by Devaraju et al. (1981) and Grew et al. (1989b). More commonly, breakdown and oxidation of magnetite results in exsolution of spinel-hercynite and oxidation-exsolution of ilmenite. Rare hercynite occurs in the Prince

TABLE 4. Selected point analyses of hercynite, hematite, and ilmenite

Sample no.	74021102	81020802B	81020910C	81020910C	80120910C	81020910C	81021005B	81021401B	704A
Mineral	Hem	Hem	Hc*	Hc**	Ilm*	Ilm**	Ilm**	Ilm**	Ilm**
Probe	NIPR	NIPR	CU	NIPR	CU	NIPR	CU	NIPR	CU
Wt%									
SiO_2	0	0	—	0	0.15	0.03	—	0	—
TiO_2	7.99	7.96	—	0	47.50	51.17	52.20	47.88	49.84
Al_2O_3	0.11	0.15	60.20	60.10	—	0.02	—	0.01	—
Cr_2O_3	0.03	0.11	0.12	0	—	0	—	0.02	—
Fe_2O_3	82.12	84.04	3.10	1.68	9.54	0.85	—	8.35	4.52
FeO	6.44	6.84	21.99	17.98	41.17	42.93	28.09	39.95	42.70
MnO	0.57	0.32	—	0.32	0.40	2.84	18.18	1.85	2.09
MgO	0.09	0	10.25	8.24	0.74	0.09	—	0.69	—
ZnO	0	0.05	4.09	11.41	—	0.03	—	0.03	—
NiO	0	0	—	0.05	—	0.05	—	0.00	—
Total	97.35	99.47	99.75	99.78	99.50	98.01	98.47	98.78	99.15
Formulae for given cations and oxygens									
Si	0	0	—	0	0.004	0.004	—	0	—
Ti	0.162	0.159	—	0	0.905	0.992	1.005	0.920	0.957
Al	0.003	0.005	1.934	1.965	—	0	—	0	—
Cr	0	0.002	0.003	0	—	0	—	0	—
Fe^{3+}	1.671	1.676	0.064	0.035	0.182	0.016	—	0.161	0.087
Fe^{2+}	0.146	0.152	0.501	0.417	0.873	0.925	0.601	0.853	0.911
Mn	0.013	0.007	—	0.007	0.009	0.062	0.394	0.040	0.045
Mg	0.004	0	0.416	0.341	0.028	0.003	—	0.026	—
Zn	0	0	0.082	0.234	—	0	—	0	—
Ni	0	0	—	0.001	—	0.001	—	0	—
Cations	2	2	3	3	2	2	2	2	2
Oxygens	3	3	4	4	3	3	3	3	3

Note: Oxide values < 0.05 wt% and ZnO ≤ 0.05 wt% are not significantly above background. Fe^{3+}-Fe^{2+} ratios calculated from stoichiometry.
* Enclosed in garnet.
** Adjacent to magnetite.

Olav Coast rocks, as well as oxidation-exsolution lamellae of ilmenite. However, conditions in the Sinnan-Ryūgū rocks were more favorable for the formation of högbomite instead of hercynite. In fact högbomite replaced most

TABLE 5. Compositon of magnetite in 704A (average over two grains)

Probe (N)	CU (5)	NIPR (5)
Wt% (standard deviation)		
SiO_2	—	0(0.0)
TiO_2	0.12(0.09)	0.05(0.04)
Al_2O_3	≤0.04	0.24(0.07)
Cr_2O_3	—	0.04(0.02)
V_2O_3	0.40(0.05)	—
FeO (meas)	92.70(0.38)	90.02(0.45)
Fe_2O_3 (calc)	68.68	66.56
FeO (calc)	30.90	30.13
MnO	0.41(0.09)	0.02(0.04)
MgO	—	0.04(0.03)
CaO	—	0.01(0.01)
Na_2O	—	0.07(0.08)
K_2O	—	0.02(0.03)
ZnO	—	0.02(0.03)
NiO	—	0.02(0.02)
Total	100.51	97.22
Formulae for 3 cations, 4 oxygens		
Ti	0.004	0
Al	0	0.011
Cr	0	0.001
V^{3+}	0.012	—
Fe^{3+}	1.981	1.988
Fe^{2+}	0.990	1.000
Mn	0.013	0

Note: Fe^{3+}-Fe^{2+} ratio by stoichiometry. Measured FeO not included in totals. N = number of analyses.

hercynite that did form and grew with, or instead of, ilmenite and rutile. At the peak metamorphic conditions (680 °C, 6–7 kbar, Hiroi et al., 1983a), the magnetite probably incorporated minor Ti, Al, Mg, and Zn, the last released by the breakdown of staurolite, relics of which contain 0.5–1.4 wt% ZnO in the högbomite-bearing samples. In some cases, however, högbomite appears with ilmenite in a reaction rim between hematite and magnetite. This reaction could be regarded as a variation of the oxidation-exsolution process in which hematite-magnetite contacts served as suitable sites for högbomite nucleation.

Temperatures of högbomite formation were probably in the 400–500 °C range, based on the presence of the assemblage rutile-magnetite (e.g., Southwick, 1968; Mielke and Schreyer, 1972; Rumble, 1976; Braun and Raith, 1985). The assemblage ilmenite-hematite in the intergrowths of hematite and ilmenite appears to be the higher-temperature equivalent of rutile-magnetite as related by the reaction

$$FeTiO_3 \text{ (in Ilm)} + Fe_2O_3 \text{ (in Hem)}$$
$$= Fe_3O_4 + TiO_2. \quad (1)$$
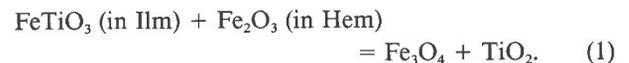
Rumble (1970) calculated a $\Delta\bar{G}$ = 3 kcal for this reaction at T = 400–600 °C, P = 1 kbar, a value no larger than the uncertainties in the calculation. Given this insignificant value for $\Delta\bar{G}$, reaction 1 must be sensitive to compositional and kinetic factors. Mn in ilmenite and hematite could stabilize the rhombohedral oxides to lower temperatures, whereas kinetics would favor the development of similar structures, that is, ilmenite exsolution

TABLE 6. Selected analyses of garnet

Sample no.	81020910C	81021005A	81021401B	704A
Probe	NIPR	CU	NIPR	NIPR
		Wt%		
SiO_2	37.93	38.09	38.74	38.61
TiO_2	0	—	0.04	0.02
Al_2O_3	21.60	21.85	21.60	21.37
FeO	27.10	24.86	29.56	27.55
MnO	1.45	8.85	1.26	1.13
MgO	7.83	5.32	7.60	7.44
CaO	2.95	1.69	1.99	3.79
Total	98.86	100.66	100.79	99.91
	Formulae for cations per 12 oxygens			
Si	2.979	2.991	3.000	3.005
Ti	0	—	0.002	0.001
Al	2.000	2.022	1.971	1.960
Fe	1.780	1.632	1.914	1.793
Mn	0.096	0.589	0.083	0.074
Mg	0.917	0.623	0.877	0.863
Ca	0.248	0.142	0.165	0.316
Total	8.020	7.999	8.012	8.012
X_{Fe}	0.66	0.72	0.69	0.68

Note: Cr_2O_3 below detection. All Fe as FeO.

TABLE 8. Selected analyses of cordierite

Sample no.	81020802B	81021005B	81021301	81021401B	704A
Probe	CU	CU	CU	NIPR	NIPR
			Wt%		
SiO_2	48.97	46.48	48.35	49.32	48.97
TiO_2	—	—	—	—	0.01
Al_2O_3	32.91	33.13	33.65	33.02	32.81
FeO	3.96	4.99	3.67	5.48	5.78
MnO	0.51	0.67	1.09	0.01	0.13
MgO	11.48	10.97	11.69	10.23	10.50
CaO	—	—	—	0.04	0.03
Na_2O	0.14	0.26	0.40	0.16	0.27
K_2O	—	—	—	0.03	—
Total	97.97	96.50	98.85	98.29	98.50
	Formulae for 18 oxygens				
Si	4.977	4.840	4.890	5.012	4.985
Ti	—	—	—	—	0.003
Al	3.942	4.065	4.010	3.954	3.936
Fe	0.337	0.435	0.310	0.466	0.492
Mn	0.044	0.059	0.094	0.001	0.011
Mg	1.739	1.703	1.763	1.549	1.592
Ca	—	—	—	0.004	0.003
Na	0.028	0.052	0.079	0.031	0.053
K	—	—	—	0.004	—
Total	11.067	11.154	11.146	11.021	11.075
X_{Fe}	0.16	0.20	0.15	0.23	0.24

Note: All Fe as FeO.

lamellae in hematite in preference to a rutile + magnetite mixture. Consequently, ilmenite-hematite intergrowths could have formed in the same rock along with rutile-magnetite. Thus, högbomite is interpreted to be stable with rutile + magnetite and manganiferous ilmenite-hematite at T ≤ 500 °C.

At these low temperatures, survival of cordierite + corundum (sample 81021005B) suggests that $P_{H_2O} < P_{Total}$. Under experimental conditions at $P(H_2O) \geq 2$ kbar in the $MgO-Al_2O_3-SiO_2-H_2O$ system, cordierite-corundum is stable relative to chlorite-Al_2SiO_5 only above 530 °C (Seifert, 1973); theoretical calculations also give a mini-mum of 530 °C assuming cordierite is hydrous and chlorite is clinochlore (Berman et al., 1986; Perkins et al., 1986). In the iron-bearing natural system, formation of chlorite + Al_2SiO_5 would be expected at somewhat higher temperatures because chlorite more readily incorporates iron than cordierite (Guidotti et al., 1975). Even under conditions of fluid $X(H_2O) = 0.5$, muscovite + quartz would form at the expense of K-feldspar + Al_2SiO_5 at

TABLE 7. Selected individual analyses of staurolite

Sample no.	74021102	81020910C	81021401B	704A
		Wt%		
SiO_2	27.21	25.91	26.31	27.12
TiO_2	0.59	0.44	0.31	0.48
Al_2O_3	54.91	55.20	54.65	53.78
Cr_2O_3	0.34	—	—	—
FeO	10.20	11.63	12.86	12.45
MnO	1.56	0.10	0.26	0.20
MgO	3.42	3.32	2.42	2.75
ZnO	1.44	0.54	1.06	0.90
Total	99.67	97.14	97.87	97.68
	Formulae for 46 oxygens			
Si	7.415	7.213	7.332	7.544
Ti	0.120	0.093	0.064	0.101
Al	17.638	18.112	17.950	17.629
Cr	0.072	—	—	—
Fe	2.324	2.709	2.998	2.897
Mn	0.360	0.022	0.061	0.047
Mg	1.390	1.378	1.006	1.140
Zn	0.290	0.112	0.219	0.185
Total	29.609	29.639	29.630	29.543
X_{Fe}	0.63	0.66	0.75	0.72

Note: All Fe as FeO. All analyses done at Chiba University. $X_{Fe} = Fe/(Fe + Mg)$.

TABLE 9. Selected analyses of biotite (NIPR probe)

Sample no.	81020910C	81021401B	704A
		Wt%	
SiO_2	37.46	36.36	36.74
TiO_2	2.63	2.48	2.27
Al_2O_3	16.41	18.23	17.28
Cr_2O_3	0.00	0.00	0.00
FeO	14.04	15.82	15.64
MnO	0.02	0.00	0.00
MgO	14.74	12.71	13.66
CaO	0.00	0.00	0.00
Na_2O	0.20	0.31	0.45
K_2O	8.03	8.53	7.84
Total	93.53	94.44	93.88
	Formulae for 22 oxygens		
Si	5.609	5.457	5.524
Al(IV)	2.391	2.543	2.476
Al(VI)	0.505	0.681	0.586
Ti	0.296	0.280	0.257
Cr	0.000	0.000	0.000
Fe	1.758	1.985	1.967
Mn	0.003	0.000	0.000
Mg	3.290	2.844	3.062
Ca	0.000	0.000	0.000
Na	0.058	0.090	0.131
K	1.534	1.633	1.504
Total	15.444	15.513	15.507
X_{Fe}	0.35	0.41	0.39

Note: All Fe as FeO.
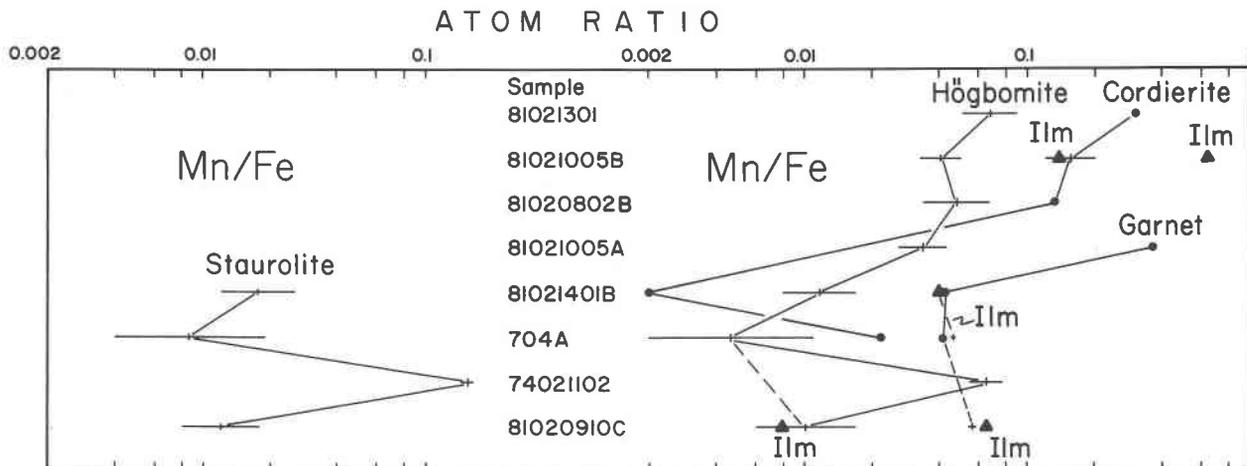
ATOM RATIO

Fig. 6. Plot of atom ratios Mn-Fe (for explanation, see Fig. 5). Dashed lines between samples 704A and 81020910C are added to emphasize covariation. Individual ilmenite (Ilm) analyses (triangles) have not been connected where variations are too great.

550 °C, P_{Total} = 2 kbar (Kerrick, 1972). Consequently, the appearance of secondary muscovite in sample 704A and in gneisses with sillimanite and K-feldspar from Sinnan Rocks (Hiroi et al., 1983b) is not inconsistent with the preservation of cordierite + corundum.

Högbomite form the upper amphibolite to granulite-facies rocks of the eastern Sør Rondane Mountains (Grew et al., 1989a) has several features in common with the Prince Olav material. The rocks of eastern Sør Rondane Mountains were affected by a later, discrete event in the amphibolite facies and retrogression in the greenschist facies. In the two samples analyzed, högbomite occurs in close association with magnetite. Associated sillimanite in one sample contains 0.94–1.12 wt% Fe_2O_3, that is, as much as the Prince Olav sillimanite, but neither of the samples analyzed contains hematite, only ferrian ilmenite (≤5 mol% hematite in solid solution, Grew et al., 1989a). Thus conditions must have been relatively oxidizing, though less so than in the Prince Olav gneisses.
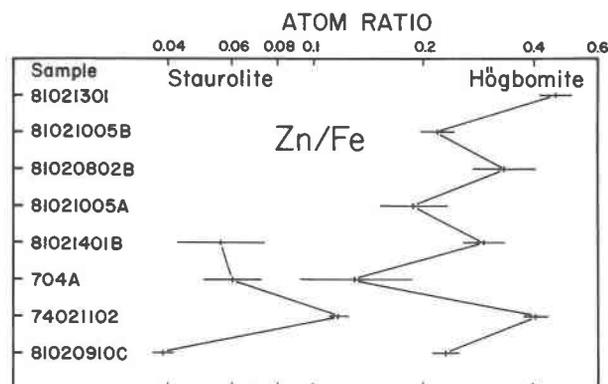
In general, högbomite parageneses can be placed into three broad groups: (1) magnetite association, (2) spinel-hercynite association, and (3) independent grains. The groups are not mutually exclusive and two types may be found in a single thin section. Examples of the magnetite association, which characterizes most of the samples from the Prince Olav Coast and Sør Rondane Mountains, include the Connemara hornfels (Leake, 1965; Angus and Middleton, 1985); Manitouwadge, Canada, cordierite-gedrite gneiss (Petersen et al., 1989); Benson Mines, New York, magnetite paragneiss (Petersen et al., 1989); and Madangere, India, Fe-Ti ore for which Devaraju et al. (1981) also propose an exsolution-like origin. The spinel-hercynite association, possibly the most abundant, is well illustrated by Gavelin (1916, type material), Friedman (1952), Zakrzewski (1977), Gieré (1986), Beukes et al. (1986), and Petersen et al. (1989). The best example of independent, clearly primary högbomite is McKie's (1963) högbomite-chlorite-enstatite-tremolite-dolomite skarn, whereas examples of secondary högbomite grains independent of other oxides are illustrated by Grew et al. (1987), Wilson (1977), and Rammlmair et al. (1988).

Despite the close association of the three groups, two distinct processes may be involved in their formation. On the one hand, most of the independent högbomite may have developed by reactions among other oxides, as well as silicates, such as those discussed by Ackermand et al. (1983), Grew et al. (1987), and Petersen et al. (1989). On the other hand, development of högbomite by alteration of spinel-hercynite or oxidation-exsolution of magnetite may involve only nearby oxides. In such parageneses, kinetic factors may play an important role and högbomite formation could represent metastable rather than stable equilibrium. Low and variable water activities would be conducive to metastable development of högbomite, incomplete exchange of constituents among the minerals, and the appearance of assemblages such as högbomite

ATOM RATIO

Fig. 7. Plot of atom ratio Zn-Fe (for explanation, see Fig. 5).

and quartz. The Antarctic högbomite-quartz association could be analogous to the corundum-quartz association that Motoyoshi and Matsueda (1987) and Motoyoshi et al. (1990) found to have formed from a metastable re-action of quartz and spinel in fluid-deficient conditions during the retrograde stage of a granulite-facies event. In the case of högbomite, polymetamorphism may result in a favorable environment for crystallization. Sufficient water should be available for högbomite, which is pre-sumed to contain OH (Gatehouse and Grey, 1982), to form instead of, or together with, spinel-hercynite. How-ever, water activities must be low enough such that, at fairly low temperatures, the development of water-rich phases like chlorite is limited. The right combination of compositional and mineralogical features needed for hög-bomite formation, in particular, composition of magne-tite, oxygen activity, and water activity, may not be com-monly met in a given polymetamorphic terrain. As a result, högbomite is rarely abundant, and in many cases, can be readily overlooked if not specifically sought.

ACKNOWLEDGMENTS

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