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Al^{iv}/Al^{vi} PARTITIONING IN CALCIFEROUS AMPHIBOLES FROM THE FROOD MINE, SUDBURY, ONTARIO

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Abstract

The quartz diorite host rock of the nickel–copper sulfide ores at the Frood mine, Sudbury, Ontario contains three amphibole phases: green ferrohornblende, blue-green ferrotschermakitic hornblende and cummingtonite. Textural relations and compositional data indicate that the blue-green hornblende developed by prograde metamorphism under high-pressure, middle amphibolite facies conditions. The green hornblende is a relict primary phase. The distribution of Al^{iv} and Al^{vi} in calciferous amphiboles partly reflects paragenesis; criteria have been established for rudimentary recognition of fields of unaltered igneous calciferous amphibole, low-pressure metamorphic hornblende and high pressure metamorphic hornblende. However, the Al^{vi} content is also dependent on the availability of charge-balancing cations, particularly Fe^{3+}, and this severely limits any quantitative approach to the paragenesis–composition relationship.

Sommaire

La diorite quartzifère minéralisée en cuivre et nickel de la mine Frood (Sudbury, Ontario) contient trois amphiboles: ferrohornblende verte, hornblende bleu-vert ferrotschermakitique et cummingtonite. Tant les relations texturales que les données chimiques indiquent un développement de la hornblende bleu-vert par métamorphisme progressif sous haute pression, dans les conditions du faciès amphibolite moyen. La hornblende verte représente une phase primaire relique. La distribution d'Al^{iv} et Al^{vi} dans les amphiboles calcifères reflète, jusqu'à un certain point, les conditions paragénétiques; nous proposons certains critères pour caractériser les amphiboles calcifères ignées fraîches, les hornblendes métamorphiques de basse et de haute pression. Toutefois, la teneur en Al^{iv} dépend aussi des cations disponibles pour équilibrer les charges, surtout Fe^{3+}, ce qui limite sérieusement toute étude quantitative de la relation entre la paragenèse et la composition.

(Traduit par la Rédaction)

Introduction

The Frood–Stobie orebody (Hawley 1962, 1965; Souch *et al.* 1969) is the largest deposit of copper–nickel sulfide in the Sudbury, Ontario mining district and much of the discussion of the origin of the Sudbury ores has centred on it. Recently, Fleet (1977) has suggested that the Frood orebody was emplaced by a hydrothermal replacement process, and argued that the chemical heterogeneity of the ore and abundant sulfide–silicate replacement textures are inconsistent with direct formation by sulfide–liquid immiscibility (Hawley 1962, 1965). In this earlier study (Fleet 1977), it was suggested that the amphibole assemblage in the host rock for the sulfide ore (green hornblende + blue-green hornblende + cummingtonite) was the product of a partial (or incomplete) static metamorphism of preexisting quartz diorite. The present paper establishes this metamorphic event through study of the chemical compositions and textural relations of the coexisting amphiboles. The distribution of Al between tetrahedral and octahedral structural positions of hornblendes (Al^{iv}/Al^{vi}) is particularly useful in this respect. At the same time we take this opportunity to clarify the somewhat confused status of the dependence of Al^{vi} content of hornblende on equilibrium pressure.

Wandke & Hoffman (1924) classified the amphiboles associated with the Sudbury ores as (a) primary green hornblende in unmineralized quartz norite and quartz diorite, (b) blue-green hornblende, associated with and replaced by sulfide and (c) actinolite, an alteration product of pyroxene. Michener (1940) and Hawley (1962) suggested that the blue-green hornblende is hastingsite or a similar variety. The Frood-mine hornblende investigated by Hawthorne & Grundy (1973) is a ferrotschermakite; this is presumed to be the blue-green variety and their analysis is reproduced in Table 1.

Al^{iv}/Al^{vi} Ratio and Paragenesis

The Al^{iv} and Al^{vi} contents of hornblende from selected suites of igneous and metamorphic rocks and of pargasite and kaersutite from miscellaneous volcanic rocks are plotted in Figure 1. Ideally, in the absence of crystal-chemical (particularly charge-balancing) constraints, the Al^{iv}/Al^{vi} ratio should be a function of temperature and pressure, with Al^{iv} increasing with

TABLE 1. REPRESENTATIVE ELECTRON MICROPROBE ANALYSES OF FROOD MINE AMPHIBOLES

	Hornblende					Cummingtonite	
	gn[1]	bl-gn,rim	bl-gn	bl-gn	Hawthorne	massive	within gn hb
	5681	5681	424	18229	& Grundy	5681	5681
SiO_2	44.4	40.9	40.7	41.4	40.12	53.2	53.1
TiO_2	1.6	0.3	0.4	0.4	0.87	0.0	0.0
Al_2O_3	7.7	15.8	15.2	15.4	18.67	0.8	0.2
Cr_2O_3	0.0	0.1	0.1	0.2	-----	0.1	0.0
Fe_2O_3	----	----	----	----	2.64	----	----
FeO	22.8[2]	23.0[2]	22.0[2]	20.1[2]	16.75	29.5[2]	30.8[2]
MnO	0.3	0.3	0.3	0.3	0.27	0.9	1.0
MgO	8.1	4.8	5.5	6.6	5.48	12.6	12.3
CaO	11.2	11.2	11.4	11.5	11.65	0.7	0.5
Na_2O	0.8	1.7	1.7	1.6	0.80	0.1	0.0
K_2O	0.9	0.5	0.5	0.5	0.75	0.0	0.0
	97.8	98.6	97.8	98.0	98.00	97.9	97.9
	recalculated on basis of 23 oxygens						
Si	6.75	6.19	6.20	6.24	6.00	8.00	8.03
Al	1.25	1.81	1.80	1.76	2.00	0.00	0.00
	8.00	8.00	8.00	8.00	8.00	8.00	8.03
Al	0.13	1.02	0.93	0.97	1.30	0.14	0.04
Ti	0.18	0.03	0.05	0.05	0.10	0.00	0.00
Cr	0.00	0.02	0.01	0.03	----	0.01	0.00
Fe^{3+}	0.54[3]	0.32[3]	0.33[3]	0.29[3]	0.30	----	----
Mg	1.83	1.08	1.25	1.48	1.22	2.82	2.77
Fe^{2+}	2.36	2.59	2.47	2.24	2.10	3.71	3.90
Mn	0.04	0.04	0.04	0.04	0.02	0.11	0.13
	5.08	5.10	5.08	5.10	5.04	6.79	6.84
Ca	1.82	1.82	1.86	1.86	1.86	0.11	0.08
Na	0.23	0.50	0.50	0.47	0.23	0.03	0.00
K	0.17	0.10	0.10	0.10	0.14	0.00	0.00
	2.22	2.42	2.46	2.43	2.23	0.14	0.08

[1] gn, green; bl-gn, blue-green; gn hb, green hornblende
[2] total iron
[3] Fe^{3+} calculated by procedure of Papike *et al.* (1974).

increasing temperature of equilibration and Al^{vi} increasing with increasing pressure of equilibration.

The data for igneous calciferous amphiboles are fairly consistent with the suggestion of Harry (1950) that Al^{iv} increases progressively with increase in crystallization temperature. However, although igneous amphiboles are characterized by relatively low Al^{vi} contents, their Al^{iv}/Al^{vi} ratios do not vary in any systematic way with supposed equilibration pressure. Most of them have $Al^{iv}/Al^{vi} \geqslant 3.3$ and this value is suggested tentatively as a limiting boundary for the field of unaltered igneous amphibole.

The chemical composition of metamorphic hornblende is, in general, more dependent on whole-rock composition than on the temperature and/or pressure of metamorphism (Shido & Miyashiro 1959, Engel & Engel 1962, Binns 1965, Bard 1970). However, Ti content does show an increase with increase in metamorphic grade (Binns 1965, Raase 1974) and Al^{vi} appears to increase with increase in pressure of metamorphism in regionally metamorphosed terrains (Raase 1974, Graham 1974). With the qualification that independent estimation of pressure of metamorphism of amphibolites is open to some ambiguity, the data presented in Figure 1 do tend to support the pressure dependence of Al^{vi}

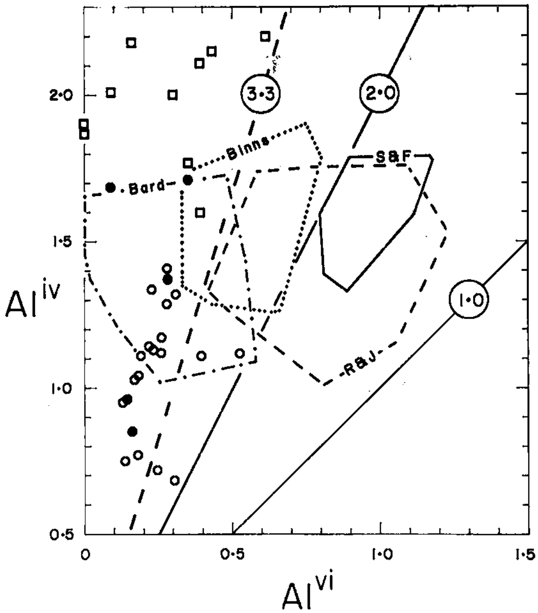

FIG. 1. Al^{iv} and Al^{vi} contents of hornblende from selected igneous and metamorphic rocks: ○ Sierra Nevada batholith (Dodge *et al.* 1968); ● Glen Tilt complex (Deer 1938); □ miscellaneous volcanic pargasite and kaersutite, superior data of Leake (1968); Bard: Aracena metamorphic belt, SW Spain (Bard 1970); Binns: Broken Hill district, New South Wales (Binns 1965); S&F: Hastings region, Ontario (Sampson & Fawcett 1977); R&J: Orange area, Massachusetts and New Hampshire (Robinson & Jaffe 1969); field of unaltered igneous calciferous amphibole defined by $Al^{iv}/Al^{vi} \geqslant 3.3$; solid line with $Al^{iv}/Al^{vi} = 2.0$ approximates the 5 kbar boundary of Raase (1974); field of low-pressure metamorphic hornblende defined by $Al^{iv}/Al^{vi} > 2.0$; field of high-pressure metamorphic hornblende defined by $Al^{iv}/Al^{vi} < 2.0$.

content. The boundary between low- and high-pressure assemblages in Figure 1 is chosen as $Al^{iv}/Al^{vi} = 2.0$. This value is equivalent, more or less, to the 5 kbar line of Raase (1974) and is consistent with the reference data in his study. However, the correlation between Al^{vi} content (or Al^{iv}/Al^{vi}) and pressure of metamorphism is only qualitative. The broad composition fields for the individual terrains may be due, in part, to variation of metamorphic grade (Binns 1965) but it seems that much of the spread in Al^{vi} and Al^{iv} contents simply reflects variation in whole-rock composition. This is a consequence of constraints on Al^{iv}–Al^{vi} partitioning in the hornblende structure reflecting the availability of charge-balancing cations. Charge balancing for Al^{iv} can be effected in a

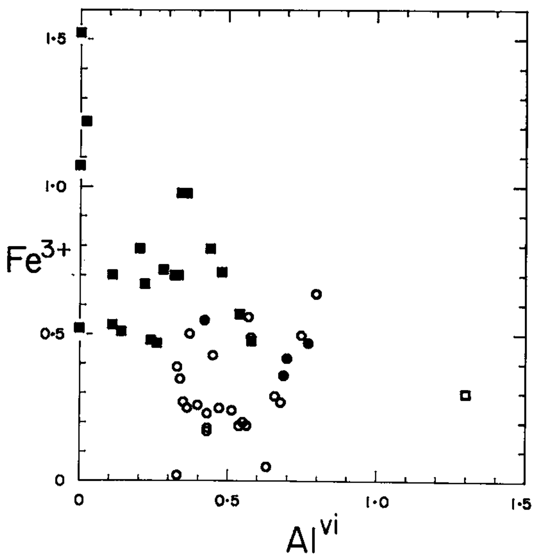

FIG. 2. Inverse correlation of Fe^{3+} and Al^{vi} in hornblende: ■ Aracena metamorphic belt, SW Spain (Bard 1970); ○ Broken Hill district, New South Wales (Binns 1965); □ Frood mine (Hawthorne & Grundy 1973).

variety of ways, principally by combinations of Al, Fe^{3+} and Ti in octahedrally coordinated *M* positions and Na and K in the *A* position. Fe^{3+} is particularly significant in this respect. Inspection of the Fe^{3+}–Al^{vi} distribution data for the Aracena metamorphic belt (Fig. 2) shows that the anomalously low Al^{vi} contents (Fig. 1) are associated with anomalously high Fe^{3+} contents. If the Aracena hornblende had more normal Fe^{3+} contents its composition field in Al^{iv}–Al^{vi} space would be displaced out of the field of igneous amphibole. Similarly, those analyses from the Sierra Nevada batholith (Dodge *et al.* 1968), which plot outside the field of unaltered igneous hornblende (Fig. 1), have low Fe^{3+} contents.

The complexity of charge balancing in hornblende appears to prohibit a meaningful correction of observed Al^{iv}/Al^{vi} ratios or Al^{vi} contents to a common pressure scale. Furthermore, the problem of correction for Fe^{3+} content would be exacerbated by the error in estimating its value from electron microprobe data. However, Al^{iv}–Al^{vi} partitioning does seem to reflect paragenesis in a rudimentary though rather imprecise way, and cautious use of this principle seems justified.

Petrography and Mineral Analysis

The present study was made on the three hand specimens (5681, 424, 18229) of the earlier investigation (Fleet 1977). The quartz diorite from the Frood mine consists essentially of amphibole (40 to 50 modal %), biotite, quartz, plagioclase and chlorite with accessory apatite, magnetite, zircon and chromite. Three varieties of amphibole have been recognized: green hornblende, blue-green hornblende and cummingtonite. These will be described in their apparent order of formation.

Green hornblende, apparently a primary phase, is characterized by the pleochroic scheme: α colorless to very pale yellow-brown; β light green-brown; γ green. It forms subhedral ragged grains partly rimmed by blue-green hornblende. Green hornblende contains very fine lamellae of cummingtonite that do not extend into the blue-green hornblende rims. Single-crystal *b*-axis precession photographs show that the cummingtonite lamellae occur in two orientations, parallel to ($\bar{1}01$) and (100). This is consistent with previous work on lamellar phases in hornblende (Ross *et al.* 1969, Immega & Klein 1976). The precession photographs suggest cummingtonite proportions of about 10% parallel to ($\bar{1}01$) and 2% parallel to (100).

Cummingtonite has been identified only in 5681, in which it amounts to about 20 modal %. It occurs principally as massive aggregates of parallel to subradiating grains. The cummingtonite aggregates are up to several mm in diameter and may be pseudomorphs of primary ferromagnesian minerals, particularly hypersthene. The individual grains are rimmed and, apparently, partly replaced by blue-green hornblende. Cummingtonite also occurs as sporadic colorless, seemingly bleached areas within the green hornblende.

Blue-green hornblende is characterized by α colorless to very pale yellow-brown, β green, γ blue-green; it occurs in a variety of habits. Much of it is in the form of spongy complexly interlocked grains that vary from fine-grained fairly massive aggregates to coarse-grained clots of only a few individuals. These appear largely to have developed by recrystallization of earlier cummingtonite. Individual poikiloblastic grains showing a complete variation from xenoblastic to idioblastic outlines are also present. These commonly appear to have developed by replacement and recrystallization of earlier green hornblende and biotite. In addition, blue-green hornblende occurs as rims on green hornblende and cummingtonite. Blue-green hornblende is subordinate in amount to green hornblende in 5681 but is the dominant amphibole in both 424 and 18229.

Biotite forms brown ragged flakes closely associated with hornblende. Plagioclase forms generally equant grains of andesine composition zoned to more sodic margins. The textural relations with quartz vary from euhedral plagioclase completely enclosed by a continuous quartz grain to rounded and embayed plagioclase surrounded by a quartz grain mosaic. In the latter case the quartz grain embayments intercept the zoning, and there is little doubt that the plagioclase zoning is primary and has persisted through the metamorphic event(s) recorded in these rocks. The chlorite is ripidolite. It occurs as individual blades and as sheaths of blades principally within biotite but having cross-cutting relationships with all of the other important silicate phases. All three hand specimens contain fine-grained mosaic patches of quartz up to several mm in diameter; these mav contain, in addition to plagioclase, one or more of blue-green hornblende, biotite and chlorite.

The rims of blue-green hornblende on green hornblende appear to be both overgrowths on and replacements of relict igneous hornblende. They show greatest development against adjacent biotite grains. Commonly the contact between the rim and green hornblende core has the appearance of being the original biotite–green hornblende grain boundary; it may be marked by fine inclusions of magnetite. Furthermore, blue-green hornblende commonly develops idioblastic faces against biotite, which suggests that it grew at the expense of the latter. There is little evidence that the blue-green amphibole in 5681 and 424 crystallized under non-hydrostatic stress. However, 18229 displays a weak foliation.

Mineral analyses were made with a MAC 400 electron microprobe fitted with the KRISEL automation system. The data were reduced on-line with the MAGIC data reduction program. The analyses (Table 1) were calibrated against the following standard materials: Si, Mg, Ca: diopside; Al, Ti: kaersutite; Cr: chromite; Fe: orthopyroxene; Mn: rhodonite; Na: albite; K: orthoclase. Precision of the analytical method is indicated by replicate determinations of the kaersutite calibration monitor, giving, in cation proportions: Si 5.896 ± .034; Al^{iv} 2.104 ± .034; Al^{vi} 0.365 ± .036; Ti 0.664 ± .012; Fe_{total} 1.524 ± .019; Mg 2.507 ± .048; Ca 1.560 ± .011; Na 0.838 ± .036; K 0.311 ± .006. More than one hundred individual spot analyses were made. Representative data are given in Table 1 (averages of two spot analyses), and representative Al^{iv} and Al^{vi} data are plotted in Figure 3 (averages of up to five spot analyses).

Discussion

According to the classification of Leake (1968), the green variety of the Frood mine hornblende is ferrohornblende and the blue-green variety is ferrotschermakitic hornblende. The blue-green hornblende has higher Al_2O_3 and lower SiO_2 and MgO than the primary green hornblende. The textural data, presented in the previous section, strongly suggest that the blue-green hornblende has developed through prograde metamorphism of quartz diorite. The compositional data are fully consistent with this conclusion, and this is most strikingly indicated by the Al^{iv}–Al^{vi} partitioning data (Fig. 3).

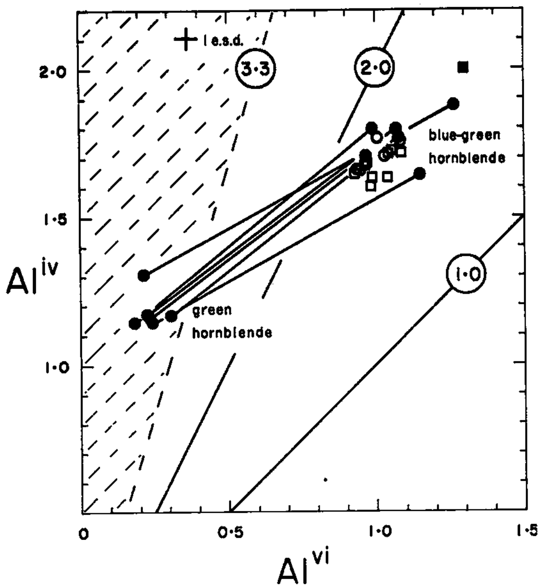

FIG. 3. Al^{iv} and Al^{vi} contents of green and blue-green hornblende from the Frood mine: ● 5681, tie-lines join green hornblende core and blue-green hornblende rim from same grain: ○ 424; □ 18229; ■ Frood mine (Hawthorne & Grundy 1973); + kaersutite microprobe monitor (bars indicate 1 *e.s.d.*); shaded area: field of unaltered igneous calciferous amphibole; other paragenetic fields defined in Figure 1.

The green hornblende analyses plot in the field of igneous hornblende, in the vicinity of composition plots for hornblende from intermediate to felsic plutonic rocks. This clearly represents a relict primary phase of the quartz diorite, perhaps modified slightly by subsequent metamorphism.

The blue-green hornblende analyses plot well within the field of high-pressure metamorphic hornblende. There is no significant difference in Al^{iv}–Al^{vi} partitioning in the blue-green hornblende from the various textural varieties and hand specimens. In general, the blue-green hornblende simply shows an addition of tschermakite component relative to the primary hornblende, and this is emphasized by the tie-lines between the corresponding green hornblende cores and blue-green hornblende rims (Fig. 3). The Frood mine ferrotschermakite investigated by Hawthorne & Grundy (1973) has a significantly higher tschermakite component than the blue-green hornblende of the present study. Sampling and petrographic information were not reported for it but its existence does point to some variation in either whole-rock composition or metamorphic conditions within the Frood–Stobie offset.

Although the dependence of Al^{vi} content on Fe^{3+} has been stressed in an earlier section, the calculated Fe^{3+} contents of both green and blue-green hornblende are in no way anomalous. They are consistent with the Fe^{3+} contents of the analyses used to define the paragenetic fields of Figure 1; they do not limit, therefore, the present paragenetic application of Al^{iv}–Al^{vi} partitioning.

Many previous investigators have noted that the color of hornblende in metabasic rocks is associated with metamorphic grade (*e.g.*, Shido & Miyashiro 1959, Engel & Engel 1962, Binns 1965). With progressive metamorphism the color changes through the sequence blue-green, green, brown-green, reddish-brown. The blue-green hornblende from the Frood mine is clearly equivalent to that frequently observed in metabasic terrains. The textural evidence presented in the previous section suggests that the Frood mine plagioclase has not recrystallized during the metamorphism. Its composition may have been modified by diffusion but most probably reflects both the primary composition and metamorphic grade. Hence, the conditions of metamorphism of the Frood–Stobie offset appear to be high-pressure, middle amphibolite facies.

Sampson & Fawcett (1977) have documented an assemblage of coexisting blue-green hornblende, cummingtonite and chlorite, apparently in stable equilibrium under high-pressure, middle amphibolite facies conditions. However, these three phases do not appear to be in equilibrium in the Frood mine metaquartz diorite. Ripidolite is almost certainly a retrograde phase. The relative stability of cummingtonite is less certain. It could, in fact, be coexisting with blue-green hornblende. However, the textural evidence presented above suggests that the massive variety is partly replaced by blue-green hornblende in 5681 and, apparently, wholly replaced by it in 424 and 18229. Hence, cummingtonite probably developed as an intermediate phase in the metamorphism of preexisting hypersthene.

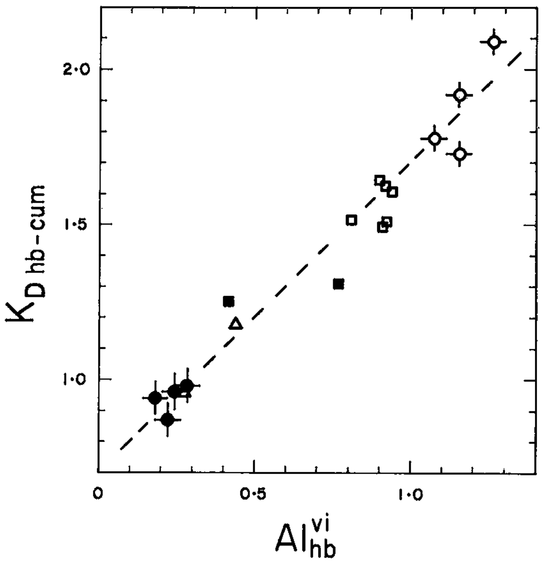

FIG. 4. Dependence of the distribution coefficient for the partition of total Fe and Mg between hornblende and cummingtonite ($K_{D\ hb\text{-}cum}$) on Al^{vi} content of hornblende: ● 5681 green hornblende–cummingtonite (bars indicate 1 *e.s.d.*); ○ blue-green hornblende-cummingtonite; △ Immega & Klein (1976); ■ Robinson & Jaffe (1969); □ Sampson & Fawcett (1977).

The stability of coexisting amphiboles has received considerable attention in the literature; much of this is referenced in Immega & Klein (1976) and Sampson & Fawcett (1977). It should be emphasized that the green hornblende is regarded as a relict phase in the metamorphosed assemblages of the present study and its persistence in these rocks is reaction-rate controlled. The presence of the lamellar phase may be taken as evidence that the green hornblende has equilibrated to some extent with the cummingtonite, although lamellae in amphiboles may form through either exsolution or replacement. The distribution coefficient ($K_{D\ hb\text{-}cum}$) for the partition of Fe^{2+} (approximated by total Fe) and Mg between hornblende and cummingtonite in 5681 is about 1.0 for green hornblende–cummingtonite and about 2.0 for blue-green hornblende–cummingtonite. These data would seem to confirm lack of equilibrium between these phases. However, a literature survey of comparable data (Fig. 4) reveals, somewhat surprisingly, that ($K_{D\ hb\text{-}cum}$) is dependent on Al^{vi} content. This dependency also holds for the more general case of partition between calciferous amphibole and Ca-poor amphibole and is, perhaps, related to a space-fitting constraint, the larger Fe^{2+} cation compensating the smaller Al cation. Thus, Fe^{2+} and Mg could well be 'equilibrated' in the Frood-mine amphiboles. This condition would not necessarily contradict the textural evidence, as it is theoretically possible for the more mobile components of complex silicates to equilibrate by exchange even though the phases themselves may not be in *stable* equilibrium. Furthermore, cummingtonite and blue-green hornblende probably formed within a similar temperature interval. As discussed above, the appearance of cummingtonite is due to favorable kinetic conditions, and its persistence indicates incomplete whole-rock equilibration.

The present study reaffirms the earlier suggestion (Fleet 1977) that the host rock of the Frood orebody has been subjected to prograde, amphibolite facies metamorphism. This was probably associated with the Penokean orogeny (Brocoum & Dalziel 1974) which has been dated indirectly at 1.7–1.75 Gyr. (Hurst & Farhat 1977). However, further speculation on the geological conditions under which metamorphism took place must await more detailed study of the country rocks in the vicinity of the orebody.

ACKNOWLEDGEMENT

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