

## Aluminum in hornblende: An empirical igneous geobarometer

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### ABSTRACT

Electron-microprobe analyses of hornblendes from five calc-alkalic plutonic complexes representing low- and high-pressure regimes define a tightly clustered linear trend in terms of total Al ( $Al^T$ ) and tetrahedral Al ( $Al^{IV}$ ) contents per 23 oxygens:  $Al^{IV} = 0.15 + 0.69Al^T$ ,  $r^2 = 0.97$ . Hornblendes from shallow-level intrusions predominate for  $Al^T \leq 2.0$ ; hornblendes from epidote-bearing intrusions within the accreted terranes of the western Cordillera all have  $Al^T \geq 1.8$ . Rocks from both pressure regimes span similar ranges in bulk-rock  $SiO_2$  content, have a common magmatic mineral assemblage (plagioclase, hornblende, biotite, K-feldspar, quartz, sphene, magnetite or ilmenite,  $\pm$  epidote), and probably crystallized at similar temperatures.

Data collated from the literature on calcic amphiboles from other plutonic complexes and from phase equilibrium experiments using natural rocks or synthetic analogue compositions show a similar  $Al^T$ - $Al^{IV}$  trend and systematic pressure effects. High-pressure calcic amphiboles have high  $Al^T$ , independent of composition. Although  $Al^{IV}$  is temperature dependent, temperature alone cannot account for the observed differences in  $Al^T$  between low- and high-pressure compositions.

The relation between  $Al^T$  in hornblende and pressure (in kilobars), for data from calc-alkalic plutons with the common mineral assemblage, is  $P = -3.92 + 5.03Al^T$ ,  $r^2 = 0.80$ . The  $Al^T$  content of hornblende is suggested as an indicator of pressure to within  $\pm 3$  kbar for crystallization of plutonic rocks of appropriate bulk composition and mineral assemblage.

### INTRODUCTION

Hornblende is the amphibole most commonly observed in calc-alkalic granitoid rocks. Data in the literature on calc-alkalic plutonic rocks and data from experimental studies using natural rocks as starting materials show that the composition of amphibole varies with bulk composition, pressure ( $P$ ), temperature ( $T$ ), and oxygen fugacity ( $f_{O_2}$ ). Wones (1981) summarized aspects of the chemical variability in amphiboles and other mafic silicates as indicators of intensive variables in granitic magmas. Compositions of amphibole from two shallow intrusive complexes (the Mount Princeton batholith in the Sawatch Range of Colorado and the Pioneer batholith in southwestern Montana) and from three plutons with evidence for deeper levels of emplacement within the crust (a pluton at Round Valley near Riggins, Idaho; a pluton at Moth Bay in the Ketchikan area of southeast Alaska; and the Ecstall pluton in northwestern British Columbia) were studied using the electron microprobe. The amphiboles in all complexes are calcic but show essential differences in composition, particularly in Al content. Differences in Al content of calcic amphiboles are directly related to the depth of emplacement of the plutons.

New data on amphiboles from these five occurrences are summarized, along with descriptive information and evidence for pressure estimates for each complex. These data are combined with other data from similar rocks

described in the literature with an attempt to distinguish effects of  $P$ ,  $T$ ,  $f_{O_2}$ , and bulk composition on the composition of amphibole. These data show that pressure is a dominant factor in accounting for the observed differences in Al contents of calcic amphiboles in calc-alkalic plutonic rocks. Table 1 cites some largely empirical previous suggestions on the influence of intensive parameters on amphibole composition.

To simplify nomenclature, the term "hornblende" is used in the general sense (Hawthorne, 1983), subsuming the appropriate names recommended by the International Mineralogical Association Subcommittee on amphibole definitions (Leake, 1978), such as edenite, magnesiohornblende, and pargasite. These more restrictive terms will be used only where distinction is necessary.

### DESCRIPTION OF THE DATA BASE

The data base for this study consists of (1) several hundred new microprobe analyses of hornblendes from five intrusive complexes in western North America; (2) analyses from the literature on well-studied intrusive complexes where estimates of  $T$ ,  $P$ , and  $f_{O_2}$  have been made and where mineral assemblage and bulk compositional data are available; and (3) analyses from the literature describing experiments on relevant natural rock compositions at controlled  $P$ ,  $T$ , and  $f_{O_2}$  conditions. Hammarstrom (1984) tabulated the new data set and discussed

Table 1. Examples of the compositional dependence of amphibole on  $T$ ,  $P$ ,  $f_{O_2}$ , and bulk composition\*

Increasing value of	$T$	$P$	$f_{O_2}$	Concentration	References
Si				+	Cawthorn, 1976; Wones and Gilbert, 1982; Spear, 1981
Al <sup>v</sup>	+				Stephenson, 1977; Helz, 1982
Al <sup>iv</sup>	-	+			Stephenson, 1977; Leake, 1971; Helz, 1982; Raase, 1974; Thompson, 1947
Ti	+		-	+	Binns, 1965; Helz, 1973; Stephenson, 1977; Spear, 1981; Raase, 1974; Anderson, 1980; Czamanske and Wones, 1973
Mn	-				Wones and Gilbert, 1982; Allen et al., 1975
Fe <sup>3+/(Fe<sup>3+</sup> + Fe<sup>2+</sup>)</sup>	-				Stephenson, 1977
Mg/(Mg + Fe <sup>2+</sup> )	+		+	+	Helz, 1982; Czamanske and Wones, 1973
A-site	+				Helz, 1982
Alkalis				+	Stephenson, 1977; Spear, 1981
Al <sup>iv</sup>		+			Spear, 1981

\* E.g., Ti in amphibole tends to increase with increasing  $T$  and increasing concentration of Ti in the system, whereas Ti in amphibole tends to decrease with increasing  $f_{O_2}$ .

details of analytical technique, formula calculation, and site partitioning.<sup>1</sup> Analyses drawn from the literature were recalculated for comparison with our data on the basis of the 23-oxygen anhydrous formula and the total Fe content as FeO. An attempt was made to screen out compositions representing late-stage alteration rather than primary magmatic crystallization by imposing Leake's (1971) proposed limit of  $Si \leq 7.5$  for "igneous" hornblendes and by limiting the data set to analyses with  $Ca \geq 1.6$ . Furthermore, analyses on hornblendes described as "late" on textural grounds were omitted.

### Bulk composition and mineralogy

Information on each complex is presented in Table 2. All the complexes are dominated by granodiorite or tonalite;<sup>2</sup> both the Mount Princeton and Pioneer batholiths include earlier and more mafic plutons (gabbro and quartz diorite) as well as later and more felsic plutons (hornblende and biotite granite, biotite granite, and two-mica granite) emplaced within the same magmatic cycles. The pluton at Round Valley is a tonalite occurring west of the Idaho batholith; the pluton is geochemically distinct from the main batholith rocks (Fleck and Criss, 1985). The pluton at Moth Bay is composed of tonalite and granodiorite (Zen and Hammarstrom, 1984a). The Ecstall pluton is a synmetamorphic and syntectonic composite intrusion (Hutchison, 1982; Crawford and Hollister, 1982) that includes diorite, quartz diorite, granodiorite, quartz monzonite, monzodiorite, and granite according to Hutchison's (1982), Fig. 44) rock classification.

The rocks are typically calc-alkalic and quartz-normative, having minor normative corundum or wollastonite (Table 3). All of the plutons overlap in  $SiO_2$  content and Al saturation index,  $A/CNK$  [molar  $Al_2O_3/(CaO + Na_2O + K_2O)$ ]. The Ecstall pluton tends to be more alu-

minous than the Pioneer or Mount Princeton batholith for a given  $SiO_2$  content (Hutchison, 1982), especially at low  $SiO_2$  contents; however, there is considerable scatter in the Ecstall data. The Ecstall sample included in this study plots near the Pioneer whole-rock chemical trend and is on the low- $Al_2O_3$  side of the range of Ecstall compositions (Woodsworth et al., 1983). The pluton at Round Valley is low in  $K_2O$ , and rocks from the area have been described as trondhjemitic (Barker et al., 1979).

The five intrusive complexes have basically the same mineral assemblage: Plagioclase (andesine to oligoclase; see Table 2), potassic feldspar, biotite, hornblende, sphene, quartz showing textural evidence for overlapping crystallization with the other minerals, apatite, and magnetite or ilmenite. Magmatic epidote occurs in the plutons at Round Valley, Moth Bay, and in the Ecstall pluton (Table 2). The epidote appears to be later than hornblende and shows a reactive relation to it (Zen and Hammarstrom, 1984a). Opaque oxide minerals are scarce in the epidote-bearing plutons, but magnetite, ilmenite, or an oxidized Fe-Ti oxide phase is present in all. Pyroxene is rare, is never a major phase, and is sheathed in later hornblende.

This common mineral assemblage, plus the silicate melt originally present, totals 9 phases (10 if a vapor phase was present; epidote does not count because it is not in equilibrium with hornblende). The corresponding chemical components, 11 in number, are  $SiO_2$ ,  $Al_2O_3$ ,  $TiO_2$ ,  $Fe_2O_3$ ,  $FeO$ ,  $MgO$ ,  $CaO$ ,  $Na_2O$ ,  $K_2O$ ,  $P_2O_5$ , and  $H_2O$ . For specified values of  $T$ , total  $P$ ,  $f_{H_2O}$ , and  $f_{O_2}$  the relatively small differences in bulk composition should be reflected in relative abundance of phases rather than in compositions of individual minerals. In terms of the phase rule, then, 11 components - 10 (or 9) phases + 2 = 3 (or 4) degrees of freedom, and the system is invariant at specified values of  $T$ ,  $P$ , and  $f_{O_2}$  (and  $f_{H_2O}$ ). For an invariant system, changes in bulk composition can only be accommodated by changes in modal proportions of phases.

### Oxygen fugacity

Abundance of sphene and magnetite in most of these rocks, the biotite compositions, and the compositions of

<sup>1</sup> To obtain a copy of the data set, order Document AM-86-315 from the Business Office, Mineralogical Society of America, 1625 I Street, N.W., Suite 414, Washington, D.C. 20006, U.S.A. Please remit \$5.00 in advance for the microfiche.

<sup>2</sup> Names of rocks conform to the Streckeisen (1973) nomenclature unless otherwise stated.

Table 2. Data for plutons and hornblendes studied for this report

	Mount Princeton batholith	Pioneer batholith	Pluton at Round Valley	Pluton at Moth Bay	Ecstall pluton
Location	Sawatch Range, Colorado	Pioneer Mountains, SW Montana	near Riggins, Idaho	Ketchikan, SE Alaska	E of Prince Rupert, B.C.
Dominant rock types*	quartz monzonite, (granite), andesitic to latitic volcanics	(gabbro), quartz diorite, tonalite, granodiorite, granite	tonalite	tonalite, granodiorite	(diorite), (quartz diorite), granodiorite
	Rock chemistry				
SiO <sub>2</sub> (%)	61–69	50–70	55–73	63	57 (54–63)
A/CNK**	0.86–1.01	0.82–1.02	0.89–1.07	0.95	0.86 (0.94–99)
	Minerals (magmatic)				
Amphibole	x	x	x	x	x
Biotite	x	x	x	x	x
Plagioclase (An)	x (50–20)	x (70–20)	x (36–25)	x (43–16)	x (45–34)
K-feldspar	x	x	x	x	x
Quartz	x	x	x	x	x
Sphene	x	x	x	x	x
Magnetite	x	x	x	x	x
Ilmenite			x	x	x
Epidote			x	x	x
P estimate	1 kbar	~2 kbar	8 kbar	8 kbar	8 kbar
Basis for estimate	coeval volcanics, contact metamorphic assemblage, regional setting	stratigraphic reconstruction, contact aureole, cooling history, miarolitic cavities		kyanite-bearing metamorphic assemblage	metamorphic assemblage
	stability field for magmatic epidote and association with gneisses and migmatites				
	Hornblende (cations per 23 oxygens)				
Al <sup>IV</sup>	0.41–1.95	0.88–1.78† 0.74–2.62‡	2.57–2.62	1.78–2.66	2.17–2.28
Al <sup>VI</sup>	0–0.38	0.11–0.52† 0.04–0.93‡	0.59–0.78	0.37–0.80	0.47–0.58
Ti	0.01–0.29	0.04–0.32† 0.03–0.41‡	0.08–0.12	0.04–0.16	0.09–0.12
	Al <sup>VI</sup> = 0.04 + 0.79Al <sup>IV</sup> ; r <sup>2</sup> = 0.97, n = 168		Al <sup>VI</sup> = 0.47 + 0.53Al <sup>IV</sup> ; r <sup>2</sup> = 0.86, n = 104		
No. of analyses	123	45 175	16	67	21
References (geology)	Crawford, 1913; Dings and Robinson, 1957; Toulmin, 1976; Toulmin and Hammarstrom (in prep.)	Zen et al., 1975, 1980; Zen, 1987; Snee, 1982	Hamilton, 1963; Barker et al., 1979; Zen and Hammarstrom (unpubl. data)	Smith and Diggles, 1978; Berg et al., 1978	Hutchison, 1982; Crawford and Hollister, 1982; Woodsworth et al., 1983
References (mineralogy)	Hammarstrom, 1984	Hammarstrom, 1982, 1984	Zen and Hammarstrom, 1984a; Hammarstrom, 1984	Zen and Hammarstrom, 1984a; Hammarstrom, 1984	Zen and Hammarstrom, 1984a; Hammarstrom, 1984

\* Rock types and compositions in parentheses occur, but are not included in this study. \*\* A/CNK = molar Al<sub>2</sub>O<sub>3</sub>/(CaO + Na<sub>2</sub>O + K<sub>2</sub>O). † Rims only. ‡ All.

magmatic epidote suggest that all these rocks crystallized under relatively oxidizing conditions—probably in the  $f_{O_2}$  range bracketed by the nickel-bunsenite (NB) and hematite-magnetite (HM) buffers. Liou (1973) demonstrated that epidote has maximum Fe content at high  $f_{O_2}$  (Ps<sub>33</sub> at HM), becomes more aluminous with decreasing  $T$  and  $f_{O_2}$  (Ps<sub>25</sub> at NB), and is unstable at  $f_{O_2}$  values lower than those defined by the quartz-fayalite-magnetite (QFM) buffer. None of the magmatic epidotes studied thus far are as Fe rich as Ps<sub>33</sub>; they are commonly zoned from Fe-rich cores to more aluminous rims (e.g., Ps<sub>20</sub> core, Ps<sub>18</sub> rim).

### Temperature

No mineral assemblages suitable for direct geothermometry occur in these rocks (the K-feldspars are Or rich, and two feldspar geothermometry typically yields subsolidus temperatures). However, studies by Wyllie (1977) and by Kenah and Hollister (1983) show that for hydrous

magmas with tonalite to granodiorite compositions, the melting intervals lie between 700 and 900°C and are nearly independent of pressure. This is considered a “normal” hydrous magmatic temperature range for the rocks in this study based on their hydrous mineralogy. Data in Table 2 show that plagioclase compositions largely overlap and that hornblende Ti contents [Ti enters amphibole more readily at increased temperatures (Anderson, 1980; Raase, 1974; Helz, 1973)] are similar for all five complexes.

### Pressure

The Mount Princeton batholith and the Pioneer batholith are shallow-level posttectonic intrusions emplaced in a cratonic environment. Estimated final pressures of emplacement and consolidation for the studied rocks of both complexes are no more than 1–2 kbar. For the Mount Princeton batholith, the estimate is based on the age of overlying volcanic rocks, which allows no more than 2

Table 3. Chemical analyses of hornblende-bearing rocks

	EC	AK-1	7-30-5*	7435*	M547-1*	IVP*
SiO <sub>2</sub>	57.20	63.07	65.20	63.80	61.10	65.70
Al <sub>2</sub> O <sub>3</sub>	18.10	19.03	18.60	15.50	15.70	16.20
Fe <sub>2</sub> O <sub>3</sub>	2.80	1.77	0.52	2.50	2.30	2.80
FeO	3.90	2.49	2.80	2.40	3.60	2.10
MgO	2.77	1.16	1.10	1.60	2.40	1.50
CaO	6.69	6.31	5.50	3.30	5.60	4.50
Na <sub>2</sub> O	4.45	4.17	5.00	3.60	3.30	3.00
K <sub>2</sub> O	1.55	1.61	0.96	4.30	3.00	2.60
TiO <sub>2</sub>	0.75	0.47	0.41	0.67	0.72	0.42
MnO	0.13	0.09	0.08	0.11	0.20	0.08
P <sub>2</sub> O <sub>5</sub>	0.31	—	0.17	0.28	0.14	0.21
Other	0.54	—	0.57	1.03	0.77	0.82
Σ	99.19	100.17	100.91	99.99	98.81	99.93
Normative minerals						
Q	7.5	16.8	17.7	17.4	15.6	26.6
C	—	—	—	—	—	0.82
Or	9.2	9.5	5.6	25.6	17.9	15.4
Ab	38.0	35.2	41.9	31.8	28.3	25.4
An	25.0	28.4	25.2	13.5	19.4	20.8
Wo	2.7	1.2	0.02	0.52	2.8	—
En	6.9	2.9	2.7	4.0	6.0	3.7
Fs	3.9	2.5	4.1	1.4	3.8	1.0
Mt	4.1	2.6	0.75	3.7	3.4	4.1
Il	1.4	0.89	0.77	1.3	1.4	0.80
Ap	0.74	—	0.40	0.67	0.48	0.50
D.I.	55	62	65	74	62	67
A/CNK**	0.85	0.95	0.97	0.94	0.83	1.02

Note: EC = granodiorite from Ecstall pluton, British Columbia; from Woodsworth et al. (1983, Table 4). AK-1 = tonalite from pluton at Moth Bay, SE Alaska; microprobe analysis of fused glass provided by John Clemens; Fe<sup>3+</sup>/Fe<sup>2+</sup> arbitrarily set to same as for Ecstall granodiorite. 7-30-5 = tonalite, Round Valley, Idaho. 7435 = Mount Aetna Quartz Monzonite porphyry, Colorado. M547-1 = Trapper Tonalite, Montana. IVP = Uphill Creek Granodiorite, Montana.

\* Rapid rock analyses performed in USGS laboratories by L. Artis, K. Coates, and H. Smith.

\*\* A/CNK = molar Al<sub>2</sub>O<sub>3</sub>/(CaO + Na<sub>2</sub>O + K<sub>2</sub>O).

Ma for the plutons to be exhumed. Even if the uplift and erosion rate was as high as 1.5 mm/yr, no more than 3 km (1 kbar) of over-burden could have been removed during this time interval (Priestley Toulmin III, 1984, oral comm.). For the Pioneer batholith, the estimate is based on the presence of a biotite + cordierite + andalusite mineral assemblage in the contact aureole and on stratigraphic reconstruction (~3–4 km) for the time of intrusion (Zen, 1987). Snee (1982) showed that <sup>40</sup>Ar/<sup>39</sup>Ar age spectra on hornblendes and biotites from the Pioneer batholith represent cooling ages and that regional cooling rates defined by these ages support shallow levels of emplacement (high cooling rates) for rocks from the eastern Pioneer Mountains, which include those described in this study.

In contrast, the Round Valley pluton, the Moth Bay pluton, and the Ecstall pluton are all located within the Mesozoic accreted terranes of the western Cordillera and are thus within the mobile belt; these three groups of plutons contain magmatic epidote (Zen and Hammarstrom, 1984a). On the basis of the stability field of magmatic epidote (Naney, 1983) and, for the Ecstall pluton, the nature of the metamorphic mineral assemblages in the surrounding country rocks, the final consolidation pressures for these plutons are estimated to be no less than

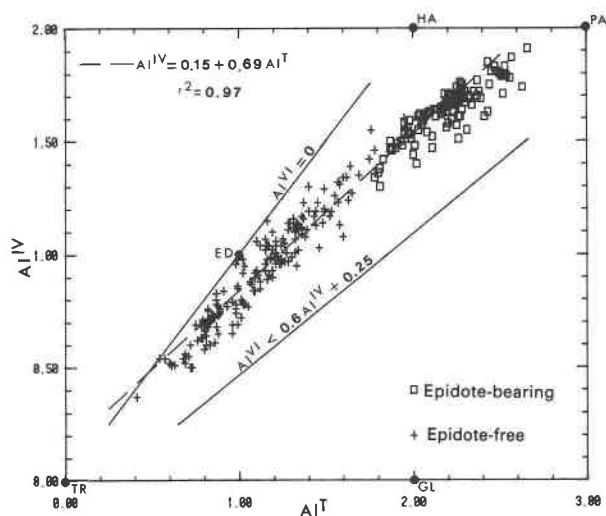


Fig. 1. Plot of Al<sup>IV</sup> versus Al<sup>IV</sup> in hornblende from five calc-alkalic intrusive complexes. Hornblende compositions from low-pressure rocks (Mount Princeton and Pioneer batholiths) are shown as crosses; compositions from high-pressure plutons in southeast Alaska, Idaho, and British Columbia are shown as squares. Ideal endmember amphibole compositions are tremolite (TR), edenite (ED), glaucophane (GL), hastingsite (HA), and pargasite (PA). The solid lines mark the limiting condition for Al<sup>VI</sup> = 0 and Leake's (1971) proposed limit for maximum possible Al<sup>VI</sup> in calcic amphibole. The equation for a linear regression on the data set (dashed line) is given in the upper left corner. All figures represent cations based on the 23-oxygen anhydrous formula unit; total Fe treated as FeO unless stated otherwise. Analytical uncertainty on Al<sup>IV</sup>, based on replicate analyses of a standard, is ±0.03 cations.

8 kbar (Zen and Hammarstrom, 1984a; Crawford and Hollister, 1982). Furthermore, these rocks are commonly gneissic, foliated, associated with migmatites, and lacking in field evidence for shallow emplacement such as associated volcanic rocks,miarolitic cavities, and widespread hydrothermal activity.

Thus, the five intrusive complexes can be separated into two groups on the basis of the presence or absence of magmatic epidote and the tectonic setting of emplacement: A high-pressure group (about 8 kbar) and a low-pressure group (1–2 kbar).

A basic assumption we make in this study is that the estimated final consolidation pressure for each pluton corresponds to the pressure of crystallization of its hornblende, at least for rims in equilibrium with quartz, so that the mineral chemistry of hornblende can be used, in conjunction with the pressure estimates, to calibrate a hornblende geobarometer.

#### Hornblende in the six calc-alkalic plutons

Hornblende is the only amphibole present in these plutons; it occurs as euhedral to subhedral phenocrysts and as inclusions in zoned magmatic feldspars. In thin section, the hornblende shows no exsolution textures but does show zoning and patchiness within single grains. Pleochroic color varies from rock to rock, depending on min-

Table 4. Representative microprobe analyses of hornblendes from five plutonic complexes

	Epidote bearing			Epidote free	
	EC1 426*	AK-1 313*	7-30-5 398*	7435 53*	IVP 249*
SiO <sub>2</sub>	41.93	41.91	41.59	46.65	47.12
Al <sub>2</sub> O <sub>3</sub>	12.53	11.46	13.39	6.69	7.60
FeO	18.19	19.40	19.84	15.74	13.48
MgO	9.56	8.87	7.51	12.74	13.58
CaO	11.15	10.85	12.21	11.68	12.15
Na <sub>2</sub> O	1.86	2.03	1.52	1.42	1.32
K <sub>2</sub> O	1.58	1.60	1.48	0.87	0.50
TiO <sub>2</sub>	0.88	1.27	0.69	1.14	0.93
MnO	0.55	0.87	0.47	0.90	0.74
Cl	0.25	0.19	0	0.03	0.13
F	0.05	0.26	0.05	0.76	0.24
Subsum	98.62	98.71	98.75	98.62	97.79
-Cl = O	0.06	0.04	0	0.01	0.03
-F = O	0.02	0.11	0.02	0.32	0.10
Sum	98.54	98.56	98.73	98.29	97.66
H <sub>2</sub> O <sub>calc</sub> **	1.89	1.79	1.95	1.64	1.88
Sum	100.44	100.35	100.68	99.94	99.54
Cations calculated on the basis of 23 O, total Fe as FeO					
Si	6.34	6.39	6.31	6.96	6.95
Al <sup>iv</sup>	1.66	1.61	1.69	1.04	1.05
T sites	8.00	8.00	8.00	8.00	8.00
Al <sup>vi</sup>	0.58	0.45	0.71	0.13	0.28
Fe <sup>2+</sup>	2.17	2.38	2.52	1.91	1.63
Mg	2.16	2.02	1.70	2.83	2.99
Ti	0.10	0.15	0.08	0.13	0.10
Mn	0	0	0	0	0
M1,M2,M3	5.00	5.00	5.00	5.00	5.00
Mn	0.07	0.11	0.06	0.11	0.09
Fe <sup>2+</sup>	0.13	0.09	0	0.06	0.03
Mg	0	0	0	0	0
Ca	1.80	1.77	1.94	1.83	1.88
Na	0	0.02	0	0	0
M4 site	2.00	2.00	2.00	2.00	2.00
Ca†	0.01	0	0.05	0.04	0.05
Na	0.55	0.58	0.45	0.41	0.38
K	0.30	0.31	0.29	0.17	0.09
A site	0.85	0.89	0.74	0.58	0.47
Σ cations	15.86	15.89	15.78	15.61	15.52
Cl	0.06	0.05	0	0.01	0.03
F	0.02	0.13	0.02	0.36	0.11
OH <sub>calc</sub> **	1.91	1.83	1.98	1.63	1.86
Fe/(Fe + Mg)	0.52	0.55	0.60	0.41	0.36

\* Analysis number in Hammarstrom's (1984) data tabulation; samples are those described in Table 3 except EC1.

\*\* H<sub>2</sub>O calculation is based on the assumption of full OH site occupancy (OH = 2.00 - F - Cl) and is only used as a check on analysis sums.

† Excess Ca after M4 site is filled; disappears if calculation assumes reasonable amounts of Fe<sup>3+</sup>.

eral association and hornblende chemistry. Hornblendes in the low-pressure plutons exhibit normal brownish-green to straw-yellow pleochroic colors, whereas hornblendes in epidote-bearing plutons are usually blue green to straw yellow. Partially resorbed hornblende is present in many rocks. The lack of subsolidus alteration is demonstrated by the fact that for some rocks of the Pioneer batholith and Mount Princeton batholith at least, <sup>40</sup>Ar/<sup>39</sup>Ar spectra for hornblende yield intrusive ages (John Sutter, oral comm.; Snee, 1982), indicating that Ar loss has not been a problem and so the hornblende probably has retained its cationic components. In parts of the Mount Princeton

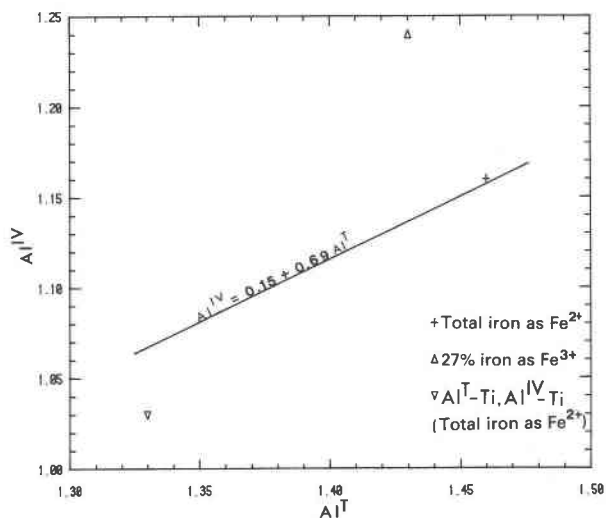


Fig. 2. The effect of including Fe<sup>3+</sup> estimates and Ti substitutions on the relative positions of points on the Al<sup>T</sup> versus Al<sup>iv</sup> plot. This is an enlargement of part of Fig. 1. The data point illustrated is for hornblende from a sample of granodiorite from the Pioneer batholith for which a complete chemical analysis on separated hornblende gave Fe<sup>3+</sup>/(Fe<sup>3+</sup> + Fe<sup>2+</sup>) = 0.27.

batholith, the hornblende has been altered to actinolite riddled with opaques: such obviously altered compositions are excluded from this study.

The range of hornblende compositions exhibited by the five intrusive complexes is given in Figure 1, which is a plot of total Al (Al<sup>T</sup>) versus tetrahedral Al (Al<sup>iv</sup>), calculated on the basis of 23 oxygens for the anhydrous formula and total Fe as FeO. All hornblendes reported show textural evidence of coexistence with quartz. For this reason, only rim compositions for hornblende from the Pioneer batholith are included in Figure 1, whereas both rim and core analyses are represented in the other data sets where (1) the overall abundance of quartz in the rocks studied or (2) the presence of quartz inclusions in hornblende suggests that most hornblende crystallized from a SiO<sub>2</sub>-saturated magma.

Al<sup>iv</sup> is calculated as the difference between 8.0 cations (full tetrahedral occupancy) and the number of Si cations; increasing values of Al<sup>iv</sup> in Figure 1 thus represent decreasing Si content. Figure 1 also shows, for reference, a line representing no Al<sup>iv</sup> content (Al<sup>T</sup> = Al<sup>iv</sup>) and locations of some endmember amphiboles.

A plot of Al<sup>T</sup> versus Al<sup>iv</sup> is easy to use for microprobe data compared to other possible amphibole plots and minimizes the problems of cumulative errors due to assignment of cations to sites and to possible site vacancies. Cumulative errors arise in standard computation of amphibole site occupancy in the order T < (M1, M2, M3) < M4 < A. Values for A-site occupancy are thus especially sensitive to errors in the analysis and to the site-partitioning scheme employed.

Figure 1 is remarkable in that hornblendes from all five intrusive complexes, ranging from magnesiohornblendes

and edenites to pargasites, plot along a single, tightly clustered trend. Since Al and Si are two major components in hornblende and  $Al^{iv} = 8 - Si$ , a strong correlation between  $Al^T$  and  $Al^{iv}$  is expected. That the points fall on a single line, however, is a surprise and permits an attempt to use the data as the basis for a geobarometer. The least-squares-fitted straight line for all data points is  $Al^{iv} = 0.15 + 0.69Al^T$ ,  $r^2 = 0.97$  ( $n = 272$ ). Hornblendes from the Mount Princeton batholith and Pioneer batholith have maximum  $Al^T$  contents of  $\leq 1.8$  cations, whereas the epidote-bearing complexes all have hornblendes with  $\sim 1.8$  or more Al cations per formula unit. Separate least-squares-fitted equations for the high-pressure (epidote-bearing) and low-pressure (epidote-free) sample segments are given in Table 2. The flatter slope for hornblendes from epidote-bearing rocks reflects a higher proportion of  $Al^{vi}$ . Leake (1971) reviewed 1500 analyses of aluminous and edenitic hornblendes and found that igneous hornblendes have lower  $Al^{vi}$  contents than metamorphic hornblendes. His proposed limit for  $Al^{vi}$  in calcic amphiboles,  $Al^{vi} = 0.6Al^{iv} + 0.25$ , is plotted on the figures along with a line representing minimum  $Al^{vi}$  content ( $Al^{vi} = 0$ ) to show that our data follow a definite trend within a field of "permissible" compositions. Representative hornblende analyses from each complex are listed in Table 4. Hornblendes from the epidote-bearing rock complexes are enriched in Al, Fe, and K relative to hornblendes from the epidote-free rocks.

#### Effects of $Fe^{3+}$ content and Ti substitutions on Al in hornblende

Obviously, Fe in hornblende is not all  $Fe^{2+}$ . However, the main separation of the data points in the plot is quite insensitive to variations in the oxidation state of Fe. An increase in  $Fe_2O_3$  will decrease the calculated content of each cation per 23 oxygens, resulting in decreased cation contents for Si,  $Al^T$ , and  $Al^{vi}$  and increased contents of  $Al^{iv}$ . Therefore, the net effect is a shift in points on an  $Al^T$  versus  $Al^{iv}$  plot diagonally toward the  $Al^{vi} = 0$  reference line. Figure 2 shows the effect of different treatment of  $Fe^{2+}$ - $Fe^{3+}$  relations on the  $Al^T$  versus  $Al^{iv}$  plot, for a selected group of data. The shifts in the plotted positions of analyses are nearly normal to the trend of the data points as a whole and do not affect the grouping of data according to the plutons. Chemical analyses of hornblende separates reveal that  $Fe^{3+}/(Fe^{2+} + Fe^{3+})$  ranges from 0.22 to 0.25 for Mount Princeton hornblende and is 0.27 for Pioneer hornblende. Although direct  $Fe^{3+}$  determinations are not available for hornblendes from the other rocks, estimates of  $Fe^{3+}$  based on stoichiometric considerations, such as those calculated by RECOMP (Spear and Kimball, 1984), are lower than or overlap values for the epidote-free rocks of this study.

Another possible influence on the distribution of points on Figure 1 is the nature of Ti substitution. Substitution of Ti for Mg in the M1, M2, and M3 sites can be accommodated by glaucophane-type,  $2Na(M4) + Ti \rightarrow 2Ca(M4) + Mg$ , or tschermakite-type,  $Ti + 2Al^{iv} \rightarrow Mg +$

$2Si$ , substitutions. If both substitutions occur, the net effect is the "NATAL" substitution described by Papike et al. (1974):  $Na(M4) + Ti + Al^{iv} \rightarrow Ca(M4) + Mg + Si$ . Thus, assuming no other type of Ti substitution, incorporation of Ti will be accompanied by an increase in  $Al^{iv}$  and consequently in  $Al^T$ . If the effect of Ti substitution is removed, the points in Figure 1 will be shifted along a line of slope +1 toward the origin, roughly parallel to the main trend of the data points in Figure 2. The effect of Ti substitution, however, is small; it is least for hornblende from the high-pressure plutons, and so a correction for Ti would tend to further separate the data for the two groups of plutons. The relatively constant Ti content throughout the data set may reflect effective buffering of Ti activity by the mineral assemblage, assuming an invariant system.

#### COMPARISON OF HORNBLENDES FROM LOW- AND HIGH-PRESSURE PLUTONS

Figure 3 shows the range of compositions from single thin sections of two rocks: sample IVP, from the Uphill Creek granodiorite of the low-pressure Pioneer batholith, and sample AK-1, from the high-pressure tonalite pluton at Moth Bay, southeast Alaska. Whole-rock analyses for both rocks are given in Table 3. The plots of Figures 3a-3d were chosen to demonstrate the possible types of Al substitution schemes in amphiboles. Arrows indicate the direction of substitution leading to relevant endmember amphibole composition from an ideal tremolite formula (A site = 0,  $Al^T = 0$ ). These plots illustrate several points that generally apply to the entire set: (1) None of the amphiboles encountered in this study are chemically homogeneous on the scale of a single thin section. (2) There is no compositional overlap on these plots between samples from low- and high-pressure environments. (3) Hornblende in AK-1 is distinctly higher in  $Al^{iv}$ ,  $Al^{vi}$ , and A-site occupancy, lower in Mg, and indistinguishable in Ti content from sample IVP. (4) High Al contents in AK-1 relative to IVP are not due to differences in amount of Ti substitutions. (5) Higher A-site occupancy (Fig. 3a) is caused by a pargasitic, rather than an edenitic coupled substitution. (6) Hornblendes from the high-pressure rocks have lower  $Mg/(Mg + Fe)$  than those from low-pressure rocks, as shown in Figure 3d; in the whole data set, there is some overlap in  $Mg/(Mg + Fe)$  between low- and high-pressure samples, but high-pressure hornblendes are *always* more aluminous.

Czarnanske and Wones (1973) suggested that high  $Al^{iv}$  balances high Fe contents in amphiboles to maintain a good fit between tetrahedral and octahedral layers. Correlation of Fe/Mg with Al in hornblende has been explained on crystal-chemical grounds:  $Al^{vi}$  is strongly ordered at the M2 site (Hawthorne, 1981) and readily displaces Mg from that site. In calcic amphiboles,  $Fe^{2+}$  prefers the M1 and M3 sites over the M2 site. Robinson et al. (1982, p. 69-76) discussed the compositional gap between actinolites and hornblendes observed in metamorphic rocks and showed that higher Fe/Mg in hornblendes relative to coexisting actinolites results from dis-

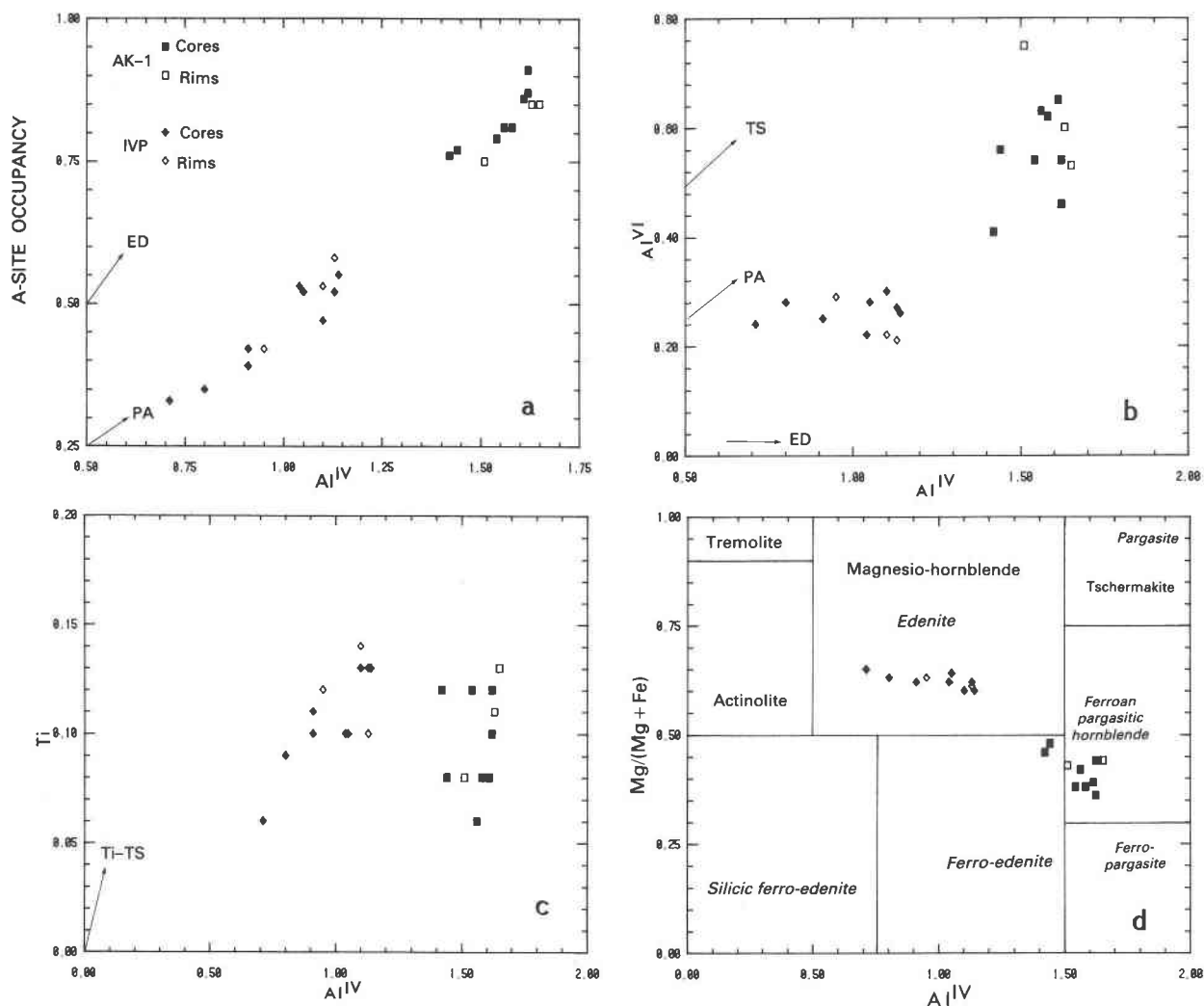


Fig. 3. Range of compositions for hornblendes from single thin sections of granodiorite from the Pioneer batholith (IVP) and tonalite from the pluton at Moth Bay (AK-1). Symbols are as follows: diamonds—analyses from sample IVP, a granodiorite from the Pioneer batholith, Montana; squares—analyses from sample AK-1, a tonalite pluton at Moth Bay, southeast Alaska. Filled symbols represent cores; open symbols represent rims. (a-c) Plots illustrating the principal substitutions involving Al<sup>iv</sup> which lead to the ideal endmembers (TS = tschermakite, see Fig. 1 for others) indicated by the arrows. (d) Modified version of the IMA-recommended plot for naming calcic amphiboles. Field names in slanted type refer to nomenclature for  $(Na + K)_A \geq 0.50$ ,  $Fe^{3+} \pm Al^{vi}$  whereas those in straight type refer to nomenclature for  $(Na + K)_A < 0.50$ ;  $Ti < 0.50$  in both cases.

placement of Mg from the M2 site by Al<sup>vi</sup>. In our data from low-pressure settings, there is no gap in compositions indicative of a solvus analogous to that between tremolite and pargasite observed in experiments by Oba (1980).

#### EFFECTS OF SiO<sub>2</sub> ACTIVITY ON HORNBLENDE COMPOSITION

Cawthorn (1976), among others, has shown that amphiboles become more SiO<sub>2</sub> rich as they crystallize from increasingly SiO<sub>2</sub>-rich melts. However, once quartz begins to crystallize, the SiO<sub>2</sub> activity ( $a_{SiO_2}$ ) = 1, and subsequent amphibole crystallization will not be affected by changes in SiO<sub>2</sub> activity.

All analyses in Figure 1 are interpreted to be in equilibrium with quartz. Hornblendes in the high-pressure

plutons contain quartz inclusions, implying SiO<sub>2</sub> saturation at the time of hornblende growth; thus, the high Al<sup>iv</sup> content of these hornblendes cannot be caused by low  $a_{SiO_2}$  in the melt. We find no textural evidence in our samples to support the interpretation of quartz inclusions in hornblende as reaction products from pyroxene replacement by hornblende, as suggested by Grissom (1984) and Peters (1984) for rocks of the Coast Plutonic Complex.

Analyses on a hornblende grain from the Ecstall pluton (Fig. 4) showed no systematic changes in composition due to proximity to a quartz inclusion. Hornblendes in the two low-pressure complexes are generally free of quartz inclusions but are commonly in contact with quartz. The most Al-rich compositions observed for Pioneer hornblendes occur in cores of hornblendes away from quartz



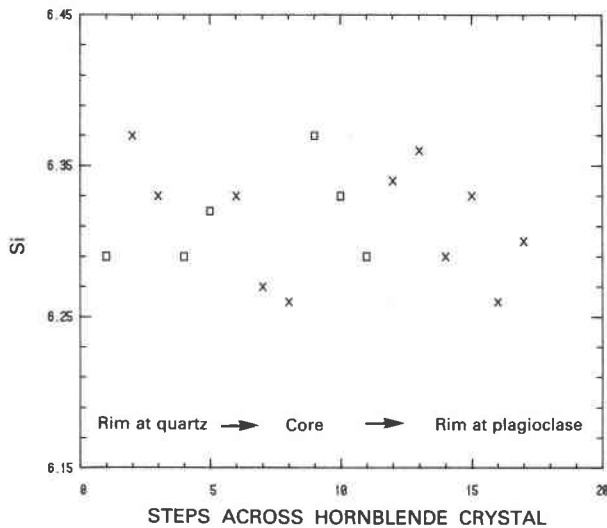


Fig. 4. Variation in Si content of hornblende across a single grain that contains a number of anhedral quartz inclusions (from the Ecstall pluton). Numbers on the  $x$  axis represent points across the crystal from a rim at quartz to a rim at plagioclase, but the points are not equally spaced. □ = points adjacent to quartz; × = points away from quartz inclusions. Analytical uncertainty on Si, based on replicate analyses of a standard, is  $\pm 0.02$  cations.

contacts in the more mafic rocks (quartz diorite, tonalite). The entire range of hornblende compositions encountered in Pioneer rocks is reported in Table 2, to demonstrate the effects of  $\text{SiO}_2$  saturation on hornblende composition, but only rim compositions are included in Figure 1. The high-Al, low-Si core portions of hornblendes in quartz diorites and tonalites may have grown before quartz began to crystallize and are presumed to reflect low values of  $a_{\text{SiO}_2}$  in the melt. However, other causes, such as higher pressure and/or temperature of early crystallization, cannot be excluded.

Figure 5 shows the range of Si content in hornblende as a function of whole-rock  $\text{SiO}_2$  content for Pioneer rocks. Hornblendes in rocks having  $< 60\%$   $\text{SiO}_2$  tend to exhibit a much wider range in Si content than hornblendes from more  $\text{SiO}_2$ -rich rocks. Maximum Si content in hornblende does not generally increase with rock  $\text{SiO}_2$  content, but minimum Si content in hornblende (i.e., maximum  $\text{Al}^{\text{IV}}$  content) jumps from  $\sim 6.3$  to  $> 6.5$  between 57 and 60% bulk-rock  $\text{SiO}_2$  content.

#### Other natural hornblendes

Hornblende compositions from the following six plutonic complexes were compiled from the literature: the Dinkey Creek intrusives, Sierra Nevada batholith, California; the Teutonia batholith, Mojave Desert, California; the Hardwick pluton, Massachusetts; the Inner Zone batholith of southwest Japan; the Finnmarka Complex, Norway; and the Pliny Range pluton, New Hampshire. Most of these have ranges in bulk-rock composition and Al saturation similar to those of the complexes discussed above. Estimated values of intensive parameters given by various authors are noted in Table 5, along with ranges

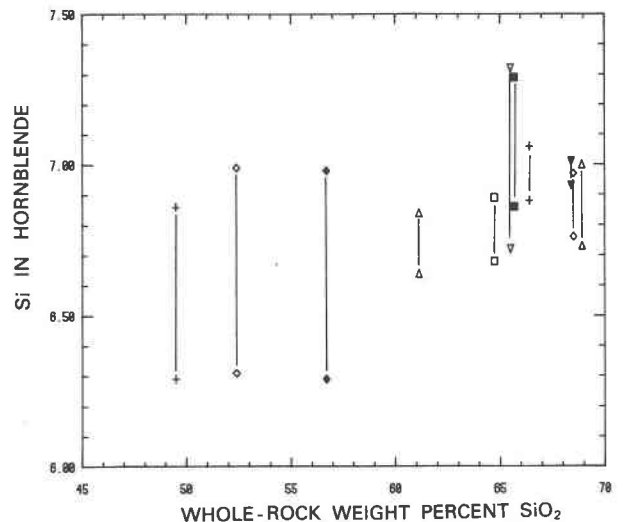


Fig. 5. Range of Si (cations per 23 oxygens) in hornblende as a function of whole-rock  $\text{SiO}_2$  content for selected rocks from the Pioneer batholith, Montana. Each symbol represents a separate rock. Lines connect minimum and maximum values of Si observed for hornblende from a single rock.

of Al and Ti contents in hornblendes. The published hornblende analyses (all microprobe data) were recalculated on the basis of 23 oxygens; total Fe was calculated as FeO for comparison with our data set. Volcanic rocks were omitted because of the difficulty in estimating pressures during crystallization. Hornblendes from rocks described in Table 5 plot along a trend (Fig. 6) similar to that shown for hornblendes in Table 2:  $\text{Al}^{\text{IV}} = 0.19 + 0.67\text{Al}^{\text{T}}, r^2 = 0.88$  ( $n = 107$ ). No hornblende with  $\text{Al}^{\text{T}} > 2.14$  was observed from these complexes. The Finnmarka Complex, with estimated emplacement pressures of 1 kbar or less, has hornblendes with maximum  $\text{Al}^{\text{T}} = 1.3$ , whereas hornblendes from the Inner Zone batholith, where pressures are estimated at 1 to 5 kbar, have  $\text{Al}^{\text{T}}$  ranging from 0.61 to 2.02, with most values  $> 1.3$ .  $\text{Fe}^{3+}$  contents estimated by Czamanske and Wones (1973) for Finnmarka and by Czamanske et al. (1977) for Pliny Range exceed  $\text{Al}^{\text{VI}}$ , which is negligible. The Hardwick pluton is part of the Acadian New Hampshire Plutonic Suite; it is a composite tonalite in the Merrimack synclinorium of central Massachusetts. Shearer (1983) described hornblende in the Hardwick pluton as a primary magmatic phase that re-equilibrated at or slightly above solidus temperatures. Pressure estimates of 5–7 kbar are given by Tracy et al. (1976) for the metamorphism that accompanied emplacement of syntectonic plutons of the region.

Inspection of Table 5 reveals (1) that maximum  $\text{Al}^{\text{T}}$  content in hornblende for a plutonic complex tends to increase with increasing estimated pressure of emplacement, (2) that no simple relationship exists between Al in hornblende and bulk-rock Al content as expressed by the A/(CNK) values, and (3) that the content of Ti is not distinct for any of the complexes. This last observation suggests that hornblende crystallization temperatures among the complexes were not grossly different, assuming



Table 5. Data for other plutonic complexes

	Finnmarka complex	Pliny complex	Dinkey Creek intrusives	Teutonia batholith	Inner Zone batholith	Hardwick pluton
Location	Oslo, Norway	Pliny Range, New Hampshire	Sierra Nevada batholith, California	Mojave Desert, California	SW Japan	Central Massachusetts
Dominant rock types	monzonite granodiorite	syenite, diorite, quartz monzonite, granite	quartz diorite, tonalite, granodiorite	gabbro, granodiorite, monzodiorite, granite	quartz diorite, tonalite, granite, granodiorite, quartz monzodiorite	tonalite
Rock chemistry						
SiO <sub>2</sub> (%)	~57–63	~50–72	57–70	49–72	53–68	52–66
A/CNK*	0.70–0.94	0.73–1.06	0.92–1.13	0.42–1.06	0.70–1.10	0.73–1.24
Assemblage	amphibole + biotite + plagioclase + K-feldspar + quartz + sphene ± magnetite ± ilmenite ± pyroxene					
T estimates	>720°C	540–740°C	~720°C	630–975°C	600–950°C	500–700°C
Basis for estimates	mineral stabilities	mineral stabilities, feldspar geothermometry	Fe-Ti oxide stabilities	hornblende-clinopyroxene and feldspar stabilities	ilmenite, biotite stabilities	feldspar geothermometry
f <sub>O<sub>2</sub></sub> estimates	NB-HM	variable, same range as Finnmarka	10 <sup>-15</sup> to 10 <sup>-10</sup>	>NB	10 <sup>-18</sup> to ~10 <sup>-12</sup>	NB-HM
P estimates	<1 kbar	2–3 kbar	2.5 kbar	<2 kbar (epizonal)	1–5 kbar	6.3 kbar
Basis for estimates	phase equilibria of rock compositions, fluid inclusions	regional metamorphism, presence of miorolites, rock compositions	metamorphism of roof pendant, erosion estimate	nature of intrusions, phase equilibria of rock compositions	no volcanics, field relations with high-grade metamorphics, sphalerite geobarometry	geobarometry on cordierite + garnet + aluminum silicate + quartz assemblage in pelitic rocks
Hornblende (cations per 23 oxygens)						
Al <sup>T</sup>	0.65–1.30	0.90–1.93	1.18–1.76	0.84–1.45	1.58**	0.62–2.04
Al <sup>vi</sup>	0.09–0.17	0.02–0.42	0.28–0.38	0.10–0.29	0.29**	0.16–0.55
Ti	0.06–0.21	0.04–0.44	0.09–0.17	0.06–0.18	0.19**	0.01–0.31
No. of analyses	8	17	6	6	1**	20
References	Czamanske, 1965; Czamanske and Wones, 1973	Czamanske et al., 1977	Guy, 1980	Beckerman et al., 1982	Czamanske et al., 1981	Shearer, 1983; Tracy et al., 1976

\* Molar Al<sub>2</sub>O<sub>3</sub>/(CaO + Na<sub>2</sub>O + K<sub>2</sub>O). \*\* Quartz-free gabbro.

similar  $f_{O_2}$ , bulk composition, and presence of a Ti-buffering phase such as sphene for all the complexes. Temperature estimates from geothermometers (Table 5) for crystallization of all these complexes largely overlap, with the exception of the low temperatures from feldspar geothermometry on the Hardwick pluton.

### Hornblendes in phase-equilibrium experiments

Hornblendes produced in seven sets of phase-equilibrium experiments that used natural rock compositions ranging from basalt to granodiorite at a variety of conditions fall along a line on an Al<sup>T</sup>-Al<sup>vi</sup> plot (Fig. 7a) having a slope similar to slopes found for natural hornblendes: Al<sup>vi</sup> = -0.23 + 0.87Al<sup>T</sup>,  $r^2 = 0.92$  ( $n = 80$ ). The synthetic hornblendes were not buffered by the same mineral assemblage as described previously for natural hornblendes in plutonic rocks.

Data from experiments provide an opportunity to investigate compositional dependence of hornblende on in-

tensive parameters and to independently examine the pressure dependence of Al content of hornblende. Helz (1982) summarized these relationships in detail and concluded that SiO<sub>2</sub> and Al<sub>2</sub>O<sub>3</sub> in hornblende do not vary systematically with bulk composition and that when pressure alone is considered, Al<sup>vi</sup> content in hornblende increases with increasing pressure.

The most Al-rich amphiboles are found in high-temperature and/or high-pressure runs, regardless of bulk-composition. Figure 7b shows that Al<sup>vi</sup> in these amphiboles increases with increasing run temperature in a fairly linear fashion and suggests that a 100-deg temperature increase can account for an increase in Al<sup>vi</sup> on the order of ~0.3 cations. Nabelek and Lindsley (1985) proposed an equation for using Al<sup>vi</sup> in amphibole as a geothermometer for some mafic rocks. Their equation includes a pressure term and is based on a subset of the experimental data considered in this study. Ti in these synthesized amphiboles also increases with increasing run temperature for a given  $f_{O_2}$  buffer, Ti and Al<sup>vi</sup> are positively correlated,

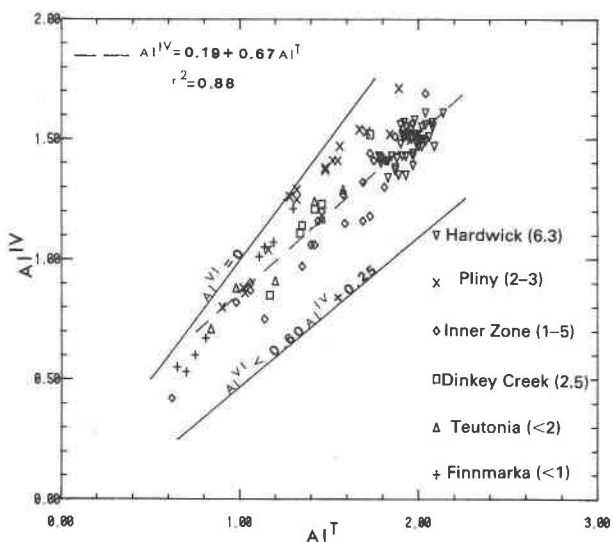


Fig. 6. Plot of  $Al^T$  versus  $Al^{IV}$  in hornblendes from six other plutonic complexes. Estimated pressures of emplacement are given in parentheses (in kilobars). Reference lines are the same as in Fig. 1. See Table 5 for data sources.

and  $Al^{VI}$  shows no correlation with temperature. In Figure 7c,  $Al^T$  is plotted against pressure for the restricted temperature range of 800 to 900°C. Most of these amphibole-bearing charges contain pyroxene and lack quartz and are, therefore, not comparable to the mineral assemblages observed in granodiorites and tonalites.

Despite the range in other intensive variables present in the data set, no hornblendes formed at 3 kbar or less have  $Al^T > 2.1$ , and all hornblendes formed at 8 kbar or more have  $Al^T > 2.0$ . This break at ~5 kbar agrees well with the break in  $Al^T$  observed for natural hornblendes. An error allowance of  $\pm 0.3$  in  $Al^T$  for the 100-deg temperature interval still results in a separation between low- and high-pressure points.

#### Discussion of the data set

The differences in amphibole composition noted for the high-pressure, epidote-bearing and the low-pressure epidote-free rocks do not simply reflect temperature differences for the following reasons: (1) Ti contents for both groups of hornblendes overlap; (2) the range of plagioclase compositions for both groups overlaps; and (3) at any crustal pressure, the crystallization temperature interval for these compositionally similar rocks should overlap, i.e., all of the high- $Al$  hornblende cannot have formed at temperatures 200 deg greater than all of the low- $Al$  hornblende (compare, for example, the  $P$ - $T$  diagrams of Wyllie, 1977, and Naney, 1983).

Similarly, one must ask if differences in  $f_{O_2}$  can account for the observed compositional differences in hornblendes. Both Helz (1973) and Spear (1981) investigated the effects of varying  $f_{O_2}$  on amphibole composition at controlled  $T$  and  $P$  for olivine basalt and olivine tholeiite bulk compositions, respectively. At 5 kbar, 725°C, Helz

reported 1.58  $Al^T$  in hornblende run at HM, and 1.62  $Al^T$  in hornblende run at QFM. Spear (1981) investigated a more aluminous bulk composition at 1 kbar and found hornblende with  $Al^T = 1.00$  in HM runs at 750°C and hornblende with  $Al^T = 1.3$  in QFM runs at the same  $T$  and  $P$ . These experiments imply that differences in  $Al^T$  resulting from  $f_{O_2}$  differences are small to negligible compared to differences that can be ascribed to  $P$  (at similar  $T$ ).

Czamanske et al. (1981) found no significant differences in  $Al^{IV}$  or  $Al^T$  for hornblendes from ilmenite and magnetite series rocks of the Inner Zone batholith. Hornblendes in ilmenite series rocks, with emplacement estimated at 4–5 kbar, do, however, have higher  $Al^{VI}$  contents which the authors suggested might be related to higher pressures of crystallization. Despite the range of oxygen fugacities represented by the Japanese hornblendes, none of them have  $Al$  contents as high as those found in epidote-bearing rocks.

### HORNBLLENDE AS A GEOBAROMETER

#### Pressure- $Al^T$ correlations

The correlation between  $Al^T$  in hornblende and pressure of formation was examined by fitting simple regression curves to the data for plutonic assemblages. More sophisticated statistical treatments of the data were not considered because of the inability to precisely quantify many of the variables inherent in the data set, lack of equal variances among data sets for different pressures, and a desire to keep to a simple, and therefore easily applied model. In order to minimize temperature and bulk compositional effects on hornblende composition, the data input was restricted to hornblende from the plutonic complexes described in Tables 2 and 5 (which have similar bulk chemistry, mineral assemblages, and estimated temperature and oxygen fugacity ranges) to examine the relationship between  $Al^T$  in hornblende and estimated pressure of emplacement. Neither rim-core relations nor quartz-hornblende textures are described in most of the studies drawn from the literature. The data are shown in Figure 8 along with several simple regression curves that are described in Table 6. The best fit (highest  $r^2$ , lowest standard error of the estimate) is given by the linear regression  $P = -3.92 + 5.03Al^T$ ,  $r^2 = 0.80$  ( $n = 379$ ). This line intersects all the data sets and is easy to use, but it predicts negative pressures for  $Al^T < 0.79$  and has no upper limit ( $Al^T$  cannot exceed 4 for amphibole). The limiting condition,  $Al^T = 0.79$  for nonnegative pressures, is interesting because it is near Leake's (1971) proposed limit for  $Al^T$  in truly igneous amphiboles ( $Al^T \geq 0.5$ ). The  $Y = AX^B$  form of the regression fits the data reasonably well but misses the data for intermediate pressures completely. The form  $Y = Ae^{BX}$  is asymptotic to both axes but also fails to intersect some pressure groups.

Despite the problems with a linear fit, we prefer to use it to describe the data set for low- to moderate-pressure regimes because it has the highest  $r^2$  and lowest standard error on the estimate. Pressure estimates based on the

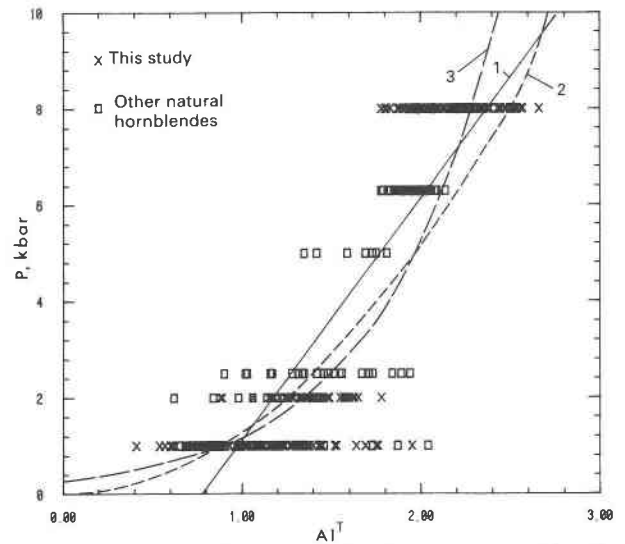
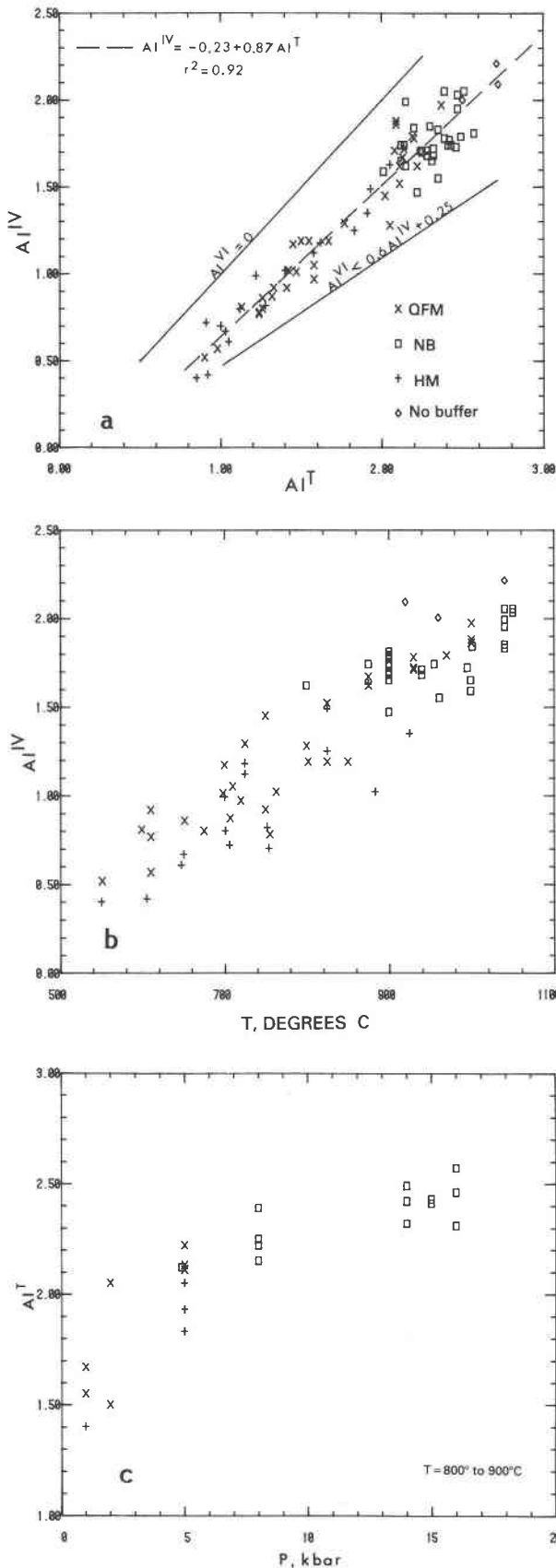


Fig. 8. Pressure estimates (kilobars) versus observed  $Al^T$  in natural hornblende. All our data (x) plotted at pressures as given in Table 2. Other data (□) as given in Table 5. Number of data points ( $n$ ) = 379. Solid line:  $Y = AX + B$  (Eq. 1). Short dashes:  $Y = AX^B$  (Eq. 2). Long dashes:  $Y = Ae^{BX}$  (Eq. 3).

various equations described above overlap within the limits of resolution of the equations ( $\pm 2$ – $3$  kbar). Better calibration is needed, especially at the high-pressure end of the data set, as the various equations diverge the most at high  $Al^T$  values.

#### Tests of the equations

Results of applying the three regression equations to our microprobe data on hornblendes from the magmatic epidote-bearing pluton at Bushy Point, Alaska (Zen and Hammarstrom, 1984b), and the Tenpeak pluton, north-central Washington (Cater, 1982; Hammarstrom, unpub. data), are tabulated in Table 7a, along with pressure estimates for hornblendes from other plutonic complexes and migmatitic gneisses. Pressures of 8 kbar or more are expected for these plutons on the basis of grossular-rich garnet compositions and on the stability of magmatic epidote. Equation 1 of Table 6 predicts  $P = 8.5$  kbar for the Bushy Point pluton and 8.2 kbar for the Tenpeak pluton, on the basis of mean  $Al^T$  values from 12 analyses on two samples from Bushy Point and 28 analyses on three samples from the Tenpeak pluton.

Pressures predicted for hornblendes in granodiorites and adamellites from Kyushu, Japan, are in excellent agree-

Fig. 7. (a) Plot of  $Al^T$  versus  $Al^{IV}$  for hornblende from phase-equilibria experiments. Different symbols represent various oxygen buffers used. Same reference lines as in Fig. 1. Sources for data are Allen and Boettcher (1983), Green and Ringwood (1968), Naney (1983), Helz (1979), Holloway and Burnham (1972), Helz (1973), Sykes (1979), and Spear (1981). (b–c) Plots of selected compositional parameters as functions of intensive variables for the experimental data as in (a). Same symbols as in (a).

Table 6. Regression equations relating pressure and Al<sup>T</sup> in hornblende

	Eq. 1	Eq. 2	Eq. 3
Form	$Y = A + BX$	$Y = AX^B$	$Y = Ae^{Bx}$
A	-3.92	1.27	0.26
B	5.03	2.01	1.48
r <sup>2</sup>	0.80	0.77	0.76
Residual error	1.86	2.11	2.22

Note: X = Al<sup>T</sup> in hornblende (cations per 23 O) and Y = pressure in kilobars; n = 379.

ment with depths of emplacement for the plutons of 1–3 kbar estimated by Tsusue et al. (1984). Estimates for the Red Lake and Eagle Peak plutons in the Sierra Nevada are high by 1 or 2 kbar compared to the  $1 \pm 0.5$  kbar depth proposed by Noyes et al. (1983). Similarly, pressures of 2–4 kbar predicted from hornblende for the Sparta quartz diorite seem a bit high for a subvolcanic complex (Brooks and Vallier, 1978). Our pressure estimates are based on microprobe data from the literature. However, these studies generally report a single averaged analysis for a rock, so compositional ranges within grains, within rocks, and as a function of buffering by other minerals in the rock are unknown. If analyses from hornblendes that are *not* in equilibrium with quartz are used, high Al<sup>T</sup> contents may be expected; this result is most likely to be a problem in the more mafic rocks, such as quartz diorites.

Pressure estimates for the Wooley Creek batholith in the Klamath Mountains are from Barnes (1983) and Barnes et al. (1986) and are based on contact metamorphic mineral assemblages and field relations. Excellent agreement is obtained between pressures predicted by Equation 1 (Table 6) and pressure estimates from metamorphic mineral assemblages associated with the Kasiks sill (Selverstone and Hollister, 1980; Kenah and Hollister, 1983; Hollister, written comm.), the Carlson Creek pluton (Grisom, 1984), and the Ponder pluton (Peters, 1984), which are all part of the Coast Plutonic Complex of British Columbia and Alaska. Hornblendes from migmatites in British Columbia reported by Lappin and Hollister (1980) yield reasonable pressures, whereas hornblendes from migmatites in the Front Range of Colorado reported by Olsen (1984) yield pressures that are higher than expected.

Predicted pressures of 0.6 to 5.2 kbar for central Sierra Nevada hornblendes encompass Evernden and Kistler's (1970) estimate of 0.8 to 3 kbar based on age-elevation relations in Cathedral Peak-type plutons and overlap the  $P = 2.5$  kbar obtained by Guy (1980) for the Dinkey Creek intrusives (see Table 5).

The rest of this discussion (Tables 7b, 7c) concerns tests of the equations as pressure predictors for calcic amphiboles in high-pressure metamorphic rocks, and for synthetic amphiboles. *None* of these rocks contains appropriate mineral assemblages for comparison with the data used to formulate the regression equations; many of these rocks contain pyroxene and/or garnet and lack quartz.

Amphiboles in lower-crustal and upper-mantle xenoliths, eclogites, and charnockites provide examples of well-

documented high-pressure occurrences. Dawson and Smith (1982) reviewed upper-mantle amphiboles that occur in xenoliths, as megacrysts, and in peridotites. As they pointed out, pressures inferred from anhydrous silicate assemblages may significantly overestimate depths of amphibole crystallization because the amphibole may not be in equilibrium with the mineral assemblages used for geobarometry. Pargasites and kaersutites characterize these assemblages; K-richterites reported for peridotites and amphiboles having Ca < 1.6 were omitted from the compilation summarized in Table 7b. Pressure estimates from the literature range from 6 to 20 kbar and are generally based on garnet-pyroxene geobarometry. These high-pressure hornblendes show (1) higher Al<sup>iv</sup> and Al<sup>T</sup> contents than observed in hornblende from plutonic rocks, (2) variable Ti content (Ti is high for lower-crustal xenoliths and is comparable to plutonic hornblende for charnockites and eclogites), (3) higher contents of Na in the M4 site than plutonic hornblende, and (4) more scatter in Al<sup>iv</sup> and Al<sup>T</sup> than plutonic hornblende, which may be a consequence of different buffering mineral assemblages. Tests on amphiboles from charnockites and granulites predict reasonable agreement over a broad pressure range (Raith et al., 1983; Janardhan et al., 1983; and revisions of those estimates provided to us by R. Newton). Results of applying regression equations to such data are included to illustrate the point that, to our knowledge, all *calcic* amphiboles from very high pressure occurrences have high Al contents. However, the regression equations may fail for pressures above 10 kbar or for inappropriate mineral assemblages.

Finally, Table 7c shows the results of applying the equations to predict pressures for some hornblendes synthesized in experiments. Plyusnina (1982) synthesized hornblendes from gels (approximately tholeiitic in composition), documented continuous changes in amphibole and plagioclase compositions with temperature and pressure, and proposed an experimental geothermobarometer for plagioclase-hornblende assemblages in amphibolites ( $P = 2-8$  kbar,  $T = 450-650^\circ\text{C}$ ). Although quartz is present and  $T$ , composition, and  $f_{\text{O}_2}$  were constant for the runs reported in Table 7c, Plyusnina reported a considerable range of amphibole compositions at each pressure. The average Al<sup>T</sup> at each pressure is used as input for the equations, and reasonable agreement between predicted pressures and actual run pressures is obtained. The predictive equations (which are based on natural hornblende data only) were also applied to other hornblendes from experiments on basalts at varying oxygen fugacities. Again, pressures predicted from hornblende compositions are generally within  $\pm 1$  kbar of actual run pressures.

#### Effects of mineral assemblage

Mean Al<sup>T</sup> = 2.48 for hornblende analyses from epidote- and garnet-bearing tonalite in the Bushy Point pluton (Table 7a) whereas mean Al<sup>T</sup> = 2.12 for hornblende from the garnet-free Moth Bay pluton, which was used to calibrate

Table 7. Tests of the method\*

Geologic occurrence	Range of Al <sup>T</sup> in amphibole	P from literature	P from regression equations			References and comments	
			1	2	3		
a. Hornblende analyses from intrusive rocks and migmatites							
Pluton at Bushy Point	2.29–2.66	>8	7.6–9.5	6.8–9.1	8.1–14.0	This study	
Tenpeak pluton	2.00–2.93	>8	6.1–10.8	5.1–11.1	5.2–20.8	This study	
Northern Kyushu, Japan	1.26–1.27 (east)	1	2.4–2.5	2.0	1.7–1.8	Tsusue et al., 1984; granodiorite and adamellite	
	0.90–1.21 (central)	↓	0.6–2.2	1.0–1.9	1.0–1.6		
	1.27–1.35 (west)	3	2.5–2.9	2.0–2.3	1.8–2.0		
Red Lake pluton, central Sierra Nevada	1.17–1.41	1.0 ± 0.5	2.0–3.2	1.7–2.5	1.4–2.2	Noyes et al., 1983; granodiorite and granite	
Eagle Peak pluton, central Sierra Nevada	1.46–1.61	1.0 ± 0.5	3.4–4.2	2.7–3.3	2.3–2.9	Noyes et al., 1983	
Sparta quartz diorite, Oregon	1.20–1.57	subvolcanic	2.1–4.0	1.8–3.2	1.6–2.8	Phelps, 1979	
Wooley Creek batholith, Klamath Mountains, California	1.76 (cumulate)	7.5	4.9	4.0	3.7	Barnes, 1983; see also Barnes et al., 1986	
	1.61 (tonalite)	↓	4.2	3.3	2.9		
	1.50 (granodiorite)	↓	3.6	2.9	2.5		
	1.12 (granite)	2	1.7	1.6	1.4		
Kasiks sill, British Columbia	1.61	4.7	4.2	3.3	2.9	Hollister, 1983 (written comm.)	
Ponder pluton	1.3	2–3	2.6	2.2	1.8	Grissom et al., 1985	
Carlson Creek pluton	1.9	4–5	5.6	4.6	4.5	Grissom et al., 1985	
Central Gneiss Complex, British Columbia, migmatitic gneiss	1.87–2.21 (gneiss)	6–8	5.5–7.2	4.5–6.3	4.3–7.2	Lappin and Hollister, 1980	
	1.83 (tonalite)	6–8	5.3	4.3	4.1		
Front Range, Colorado, migmatites	2.04–2.13	3.5 ± 1	6.3–6.8	5.3–5.8	5.5–6.4	Olsen, 1984	
Sierra Nevada batholith, California	0.91–1.81**	0.8–3	0.6–5.2	1.0–4.2	3.9	Dodge et al., 1968; mafic and granitic rocks. Evernden and Kistler, 1970	
Guadalupe Complex, California	1.34–1.73**	4–5	2.8–4.8	2.3–3.8	2.0–3.5	Best and Mercy, 1967; P estimates from contact assemblage	
b. Microprobe analyses of calcic amphiboles from xenoliths and high-grade metamorphic rocks							
Megacrysts in xenoliths	2.01–2.57	upper mantle	6.2–9.0	5.2–8.5	5.3–12.2	Dawson and Smith, 1982	
Mafic to ultramafic xenoliths	2.09–2.81	lower crust or upper mantle	6.6–10.2	5.6–10.2	6.0–17.4	Schulze and Helmstaedt, 1979; Kay and Kay, 1983; Best, 1975; Conrad and Kay, 1984; Wass and Hollis, 1983	
Charnockite and granulite, India	1.75 (metabasite)	4.5–5.0	4.8	3.9	3.6	Janardhan et al., 1983; Raith et al., 1983; R. Newton, 1984, written comm.	
	1.81 (acid gneiss)	4.5–5.0	5.2	4.2	3.2		
	1.84 (charnockite)	4.5–5.0	5.3	4.4	4.1		
	2.29 (charnockite)	7.0	7.6	6.8	8.1		
	2.18 (charnockite)	7.2	7.1	6.1	6.8		
	2.50 (charnockite)	7.2	8.6	8.1	11.0		
2.56 (charnockite)	6–9	9.0	8.5	12.0			
c. Microprobe analyses of amphiboles from experiments							
T	f <sub>02</sub> buffer	Al <sup>T</sup>	Run P	P from regression equations			References
				1	2	3	
560	NB	1.32 (1.18, 1.45)	2	2.7	2.2	1.9	Plyusnina, 1982
550, 580	NB	1.73 (1.16–2.16)	4	4.8	3.8	3.5	Plyusnina, 1982
550	NB	1.93 (1.90, 1.95)	6	5.8	4.8	4.7	Plyusnina, 1982
754	QFM	1.33	1	2.8	2.2	1.9	Spear, 1981
750	HM	1.00	1	1.1	1.3	1.2	Spear, 1981
763	QFM	1.42	3	3.2	2.6	2.2	Spear, 1981
752	HM	1.28	3	2.5	2.1	1.8	Spear, 1981
710	QFM	1.58	5	4.0	3.2	2.8	Spear, 1981
690	HM	1.47	5	3.5	2.8	2.4	Spear, 1981
725	QFM	1.62	5	4.2	3.4	3.0	Helz, 1973
725	HM	1.58	5	4.0	3.2	2.8	Helz, 1973

\* Al<sup>T</sup> = total Al per 23-oxygen amphibole formula unit with total Fe calculated as FeO; all pressures in kilobars.

\*\* Complete chemical analyses rather than microprobe analyses reported in these studies; analyses recalculated to a 23-oxygen formula basis with total Fe as FeO and H<sub>2</sub>O omitted.

the geobarometer at 8 kbar. Similarly, mean Al<sup>T</sup> = 2.41 for hornblendes from the magmatic epidote-bearing Tenpeak pluton. Garnet is reported in the Tenpeak, and potassium feldspar is rare to absent (Art Ford, 1985, written comm.). The effects of differences in mineral assemblages

in equilibrium with hornblende on the validity of the geobarometer are not clear. Zen and Hammarstrom (1984a, 1984b) proposed pressures on the order of 13 kbar or more for the almandine-grossular garnets in the Bushy Point pluton; these garnets are usually sheathed in pla-

gioclase and are not in textural equilibrium with hornblende. The more aluminous hornblendes in these garnet-bearing rocks may reflect relatively higher pressures of formation for these plutons than for the other epidote-bearing plutons considered in this study, but the possibility of buffering of Al content of hornblende by a coexisting Al-bearing garnet at high pressures cannot be ruled out. Such a buffering effect may account for the fact that  $Al^T$  in amphiboles produced in phase-equilibria experiments increases with increasing run pressure up to 10 kbar (Fig. 7c), then levels off at higher pressures, where Al-bearing garnets are sometimes observed in the phase assemblage (Allen et al., 1975).

Peters (1984) and Grissom (1984) tested a preliminary formulation of our pressure-correlation equation by analyzing hornblendes from the Ponder pluton, a granodiorite, and from the Carlson Creek pluton, a tonalite, and found that pressures from hornblende agreed to  $\pm 1$  kbar with independent pressure estimates from metamorphic assemblages in country rocks. Peters et al. (1985) found that while  $Al^{vi}$  and  $Al^T$  increase from core to rim in both plutons, the  $Al^{iv}$  zoning pattern varies with the presence or absence of potassium feldspar.

The proposed correlation should be applied to calc-alkalic rocks that have the mineral assemblage plagioclase (andesine-oligoclase) + K-feldspar + quartz + hornblende + biotite + sphene + magnetite (or ilmenite). Such a mineral assemblage should adequately buffer the system so that variation of the bulk composition should be largely expressed by modal proportions of the minerals rather than by the compositions of the individual phases. In particular, because the  $Al^{iv}$  content of hornblende is negatively correlated with the Si content, hornblende chosen to estimate the pressure should coexist with quartz to eliminate  $a_{SiO_2}$  as a variable. Enough analyses must be obtained to ascertain the true range of amphibole composition in a given rock or pluton, including core-to-rim variations, variation in rim composition as a function of adjacent minerals, and subsolidus alteration.

Late-stage (probably near-solidus) alteration by oxidation results in the growth of more magnesian, less aluminous amphibole. In the most altered samples of the Mount Princeton quartz monzonite, hornblendes are patchy. Changes in composition from  $Al^T = 1.08$  and  $Fe/(Fe + Mg) = 0.40$  in darker-colored areas to  $Al^T = 0.80$  and  $Fe/(Fe + Mg) = 0.35$  in pale-colored parts of the same grain are associated with small, rounded magnetite grains and suggest that alteration by oxidation has occurred. Equation 1 of Table 6 predicts  $P = 1.5$  kbar for part of the grain, 0.1 kbar for the other. Care must be used in screening analyses for pressure estimates; alteration will yield anomalously low pressure estimates (although alteration through vapor-phase saturation may itself be a hallmark of shallow emplacement), whereas lack of equilibrium with quartz may yield anomalously high pressure estimates.

Use of hornblende for geobarometry presumes that hornblende preserves its magmatic composition. Use of

the concentration of Al, one of the less- if not the least-mobile elements in a crystalline environment, helps to insure meaningful results (in addition to the other advantages cited earlier). It is possible to make rough estimates of the likelihood that the Al in a given hornblende has not been exchanged under subsolidus conditions. For a given value of the diffusion coefficient, the rate of growth depends on the square root of time,  $t$ . Suppose  $t = 3 \times 10^{13}$  s ( $10^6$  yr);  $t^{1/2} \approx 6 \times 10^6$ . A 1-mm crystal would be completely "worked over" in the allotted time if the diffusion coefficient is no larger than about  $10^{-6}$  m<sup>2</sup>/s. This value is certainly orders of magnitude greater than a reasonable diffusion coefficient for Al in hornblende even at magmatic temperatures for granodiorite and granite (by analogy with data for Al in diopside, Freer, 1981). Therefore, even if the crystals were considerably smaller, hornblende ought to retain magmatic Al contents for reasonably large and slow-cooling plutons.

## DISCUSSION

The data for natural hornblende follow a single, tightly clustered trend when the  $Al^T$  is plotted against  $Al^{iv}$  ( $= 8 - Si$ ). The partitioning of Al into the two coordination sites follows a simple relation; in the magma, Al behaved as a single component with a single activity coefficient. The activity of Al in calc-alkalic magmas clearly varied with pressure in a manner so different from that of Si that a geobarometer is implied. Since hornblende contains measurable quantities of all the major rock-forming components, unlike other major plutonic minerals, study of hornblende compositional variation provides information on changes of the activities of these components in the magma and thus provides a basis for use of hornblende as a monitor of magma history.

Metamorphic hornblendes show equally large variations in Al content, although  $Al^T$  versus  $Al^{iv}$  plots appear to follow different trends, implying different substitution schemes. Hornblendes from high-grade metamorphic rocks have higher  $Al^{vi}$  and Na(M4) contents than the plutonic hornblendes. (See, for example, the fields for calcic and sodic-calcic amphiboles from low-, medium-, and high-pressure mafic schist assemblages outlined by Laird and Albee, 1981, Fig. 12.) The reasons for these compositional differences are not clear; lower metamorphic temperature may be a significant factor. Most metamorphic rocks do not have mineral assemblages or bulk compositions that correspond to those characteristic of calc-alkalic igneous rocks, but many do contain quartz. Study of the variation in Al content of metamorphic hornblende coexisting with specified mineral assemblages in contact aureoles could eventually provide a calibration between the two groups of rocks, as equal pressure could be assumed.

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