

## Calcic and Alkali Amphiboles from the Golden Horn Batholith, North Cascades, Washington

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### Abstract

Five optically and chemically different amphiboles in calc-alkaline and alkaline granites of the Golden Horn Batholith represent both the calcic and alkali amphibole groups. A gradation in the hornblende-ferrohastingsite solid solution series developed as, with falling temperature, the Fe/Mg ratio of the calc-alkaline magma increased and oxygen fugacity decreased. The alkali amphibole group in the alkaline granite includes riebeckite, osannite, and an additional amphibole with a natural arfvedsonite composition, but a crossite optic orientation. The alkali amphiboles are in a solid solution series between natural arfvedsonite,  $\text{Na}_2\text{Ca}_{0.5}\text{Fe}^{++}_{2.5}\text{Fe}^{+++}_{1.5}\text{Al}_{1.5}\text{Si}_{7.5}\text{O}_{22}(\text{OH})_2$  (Miyashiro, 1957) and end-member arfvedsonite,  $\text{Na}_2\text{Fe}^{++}_4\text{Fe}^{+++}\text{Si}_5\text{O}_{22}(\text{OH})_2$  (Sundius, 1946). The solid solution series progressed from earlier-formed arfvedsonite through osannite to riebeckite as temperature fell, oxygen fugacity rose, and Ca content of the magma decreased. Rare crystals, zoned from corroded ferrohastingsite cores to arfvedsonite rims, indicate a short period of calcic amphibole formation followed by a discontinuous reaction preceding the alkali amphibole crystallization.

### Introduction

Misch (1965) described four optically different amphiboles, including both the calcic and alkali amphibole groups, in the Golden Horn Batholith. In addition, Stull (1969, 1971) found an amphibole with a crossite optic orientation, but an arfvedsonite composition, not generally known to occur in igneous rocks. These five amphiboles represent a composite example of several interpretations (Billings, 1928; Holm, 1971) and experimentally-based theories (Ernst, 1962, 1968) on the relationships within and between the calcic and alkali amphibole groups. This paper describes: (1) the characteristics of the five amphiboles from the Golden Horn Batholith; (2) the relationships within and between the calcic and alkali amphibole groups of the batholith; (3) the relationship between observed features in the Golden Horn amphiboles and previous experimental work; and (4) the significance of amphibole chemistry in interpretation of the magmatic environment.

### Geology

Misch (1965, 1966) recognized three rock types within the Eocene Golden Horn Batholith: (1) alkaline granite bearing sodic amphiboles; (2) one-feld-

spar biotite granite and (3) two-feldspar biotite granites. These rocks contain four optically different amphiboles (Misch, 1965):

- (1) riebeckite and osannite, in rare cases associated with late-formed aegirine-augite, in the alkaline granite;
- (2) ferrohastingsitic hornblende or ferrohastingsite in the one-feldspar biotite granite;
- (3) common hornblende in the two-feldspar biotite granite.

In addition, Stull (1969) identified an amphibole with a crossite optical orientation, but an arfvedsonite composition, in the alkaline granite.

Mapping by Misch (personal communication) and Stull (1969) indicates that the alkaline granite reached the most shallow level of intrusion and may have crystallized at pressures between 0.5 and 1.0 kbar. The biotite-bearing granites probably began crystallization of euhedral amphibole and plagioclase at deeper levels and later intruded to nearly the same level as the alkaline granite. This is supported by the experiments of Piwinskii and Wyllie (1968) and Piwinskii (1968) which indicate that amphibole will form early in the crystallization sequence only if  $P_{\text{fluid}}$  is greater than 1.0 kilobar.

### Petrography

Common hornblende passes through ferrohastingsitic hornblende to ferrohastingsite with the optic axial angle decreasing as the two-feldspar biotite granite grades into the one-feldspar biotite granite (Table 1). Similarly, the associated biotite is more iron-rich in the one-feldspar biotite granite (Stull, 1969).

The alkali amphiboles have three different optic orientations (Table 1). The arfvedsonite (A-1, A-2, A-3) has an optic orientation similar to crossite with its optic axial plane perpendicular to (010) and  $Y$  being close to  $c$ . This mineral is referred to as arfvedsonite rather than crossite because it occurs in the low pressure environment of a shallow pluton and is chemically identical to Miyashiro's (1957) natural arfvedsonite. The optic axial plane of osannite (O-1) is perpendicular to (010) and close to (100) with a small angle between  $X$  and  $c$ . Although osannite has been dropped as a proper term by Deer, Howie, and Zussman (1963), it is retained here to indicate an amphibole that is optically identical to Tröger's (1959) osannite and chemically different from other alkali amphiboles including arfvedsonite and riebeckite. The riebeckite (R-1) has its optic axial plane parallel to (010) as properly defined by Tröger (1959).

Texturally, the alkali amphiboles have three modes of occurrence: (1) early-formed euhedral crystals; (2) large grains partially recrystallized to finer-grained alkali amphibole and associated with fluorite, magnetite, zircon, apatite, carbonate, and minor biotite; and (3) large, late-formed crystals, locally two or three inches long, in miarolitic cavities and

other openings. The euhedral form of the arfvedsonite suggests that it was the first amphibole to crystallize while the subhedral osannite formed slightly later. The riebeckite formed last and is the only amphibole in miarolitic cavities. Thus, these amphiboles did not form as coexisting minerals, but crystallized in a sequence that involved three different optical orientations and a continuous change in chemical character.

Discussion of the relationship between calcic and alkali amphiboles began when Billings (1928) postulated that crystallization of ferrohastingsite could be followed by riebeckite. Borley and Frost (1963) later published one analysis of an intermediate member as evidence of a solid solution series between these minerals. Holm (1971) recently described an amphibole that is zoned from a hastingsite core through a uniaxial zone to an alkali amphibole rim as evidence for a similar solid solution series. Holm's alkali amphibole is close to the rare mboziite ( $\text{Na}_2\text{CaFe}^{++}_3\text{Fe}^{+++}_2\text{Al}_2\text{Si}_6\text{O}_{22}(\text{OH})_2$ ) originally described by Brock, Gellatly, and Von Knorring (1964), and is chemically much different from the Golden Horn alkali amphiboles. Zoned crystals in Golden Horn granite provide evidence of the relationship between ferrohastingsite and a more alkali amphibole of arfvedsonite composition. The rare zoned crystals contain ferrohastingsite cores [O.A.P. = (010);  $2V = -10^\circ$ ] and arfvedsonite rims [O.A.P.  $\perp$  (010),  $Z = b$ ,  $Y:c = 18 - 21^\circ$ ,  $2V = -70^\circ$ ] with the transition from ferrohastingsite to arfvedsonite occurring over a narrow zone where the optic axial angle is zero. The outer edge of the uniaxial zone is partially resorbed, suggesting

TABLE 1. Optical Data

	H-1	F-2	F-3	F-4	A-1	A-2	A-3	O-1	R-1
orientation	$Y=b$	$Y=b$	$Y=b$	$Y=b$	$Z=b$	$Z=b$	$Z=b$	$Z=b$	$Y=b$
optic plane	(010)	(010)	(010)	(010)	$\perp(010)$	$\perp(010)$	$\perp(010)$	$\perp(010)$	(010)
$X:c$	--	--	--	--	--	--	--	$3^\circ$	$5-18^\circ$
$Y:c$	--	--	--	--	$18^\circ$	$20^\circ$	$21^\circ$	--	--
$Z:c$	$15^\circ$	$13^\circ$	$14^\circ$	$16^\circ$	--	--	--	--	--
$\gamma-\alpha$	0.023	0.021	0.019	0.019	0.011	0.010	0.012	0.013	0.013
$2V_x$	$75^\circ$	$50^\circ$	$40^\circ$	$30^\circ$	$60^\circ$	$70^\circ$	$70^\circ$	$80^\circ$	$95^\circ$
dispersion	none	none	none	none	$r>v$ strong	$r>v$ strong	$r>v$ strong	$r>v$ strong	$r<v$ strong
absorption	$Y>Z>X$	$Z>Y>X$	$Y>Z>X$	$Y>Z>X$	$Z>Y>X$	$Z>Y>X$	$Z>Y>X$	$Z>X>Y$	$X=Y>Z$
$X$	tan	tan	tan	tan	tan	brown	brown	dark blue	dark blue
$Y$	green-brown	brown	green-brown	dark brown	blue	blue	blue	tan	dark blue
$Z$	green	dark green	green	dark green	dark blue	dark blue	dark blue	opaque	yellow

H-1 - Common hornblende in two feldspar-biotite granite  
 F-2, F-3, F-4 - Ferrohastingsite in one feldspar-biotite granite  
 A-1, A-2, A-3 - Arfvedsonite in alkaline granite  
 O-1 - Osannite in alkaline granite  
 R-1 - Riebeckite in alkaline granite

that a discontinuous reaction normally separates ferrohastingsite and alkali amphibole crystallization as postulated by Billings (1928).

### Materials and Procedures

Nine specimens selected for mineral separation and chemical analysis are presented on Tables 2 and 3. Mineral separations of better than 98 percent purity were done with a combination of the Frantz isodynamic separator and heavy liquids including methylene iodide and Clerici. The amphibole analyses were done with wet chemical techniques by Dr. Ken-Ichiro Aoki. Li and Zn were determined with atomic absorption. Na, K, and total iron were repeated by atomic absorption as a check on the wet chemical analysis and no significant error was found. The alkali amphibole structural formulae were calculated on the basis of 23 oxygens per formula unit, and not 24, due to the difficulty in water determination and the absence of fluorine in the analysis. The calcic amphiboles probably contain little fluorine and were calculated on the basis of 24 oxygens. Optic axial angles were measured directly in thin-section with a 5-axis universal stage.

### Amphibole Chemistry

#### *Hornblende and Ferrohastingsite*

The optically observed gradation from hornblende to ferrohastingsite is marked by a sharp increase in FeO and a decrease in MgO (Table 2) resulting in a change of the Fe<sup>++</sup>/Mg ratio from 1.10 to 14.30. In addition, the Fe<sup>++</sup>/Fe<sup>+++</sup> ratio increases from 3.79 in hornblende to 7.50 in ferrohastingsite. Billings (1928) observed a similar relationship and concluded that the variation from magnesianhastingsite to ferrohastingsite is a continuous reaction series that is terminated at lower temperature with a discontinuous reaction and crystallization of riebeckite. In the Golden Horn case, common hornblende grades into ferrohastingsite in a traverse from the two feldspar-biotite granite toward the sodic amphibole-bearing alkaline granite.

The hornblende and ferrohastingsite are low in Al, reflecting the low Al content of the Golden Horn granite. The maximum amount of Al calculated to be in the tetrahedral site is 1.29 atoms. Silica and Al remain fairly constant from common hornblende to ferrohastingsite; only sample F-3 has relatively higher Si and lower Al. Compared with previously published analyses (Buddington and Leonard, 1953;

Borley, 1963) the percent Al<sub>2</sub>O<sub>3</sub> is very low and the SiO<sub>2</sub> is slightly high.

The transition from common hornblende to ferrohastingsite parallels the chemical trends within the batholith. A pronounced increase in Fe<sup>++</sup> relative to Mg of the amphiboles parallels the sharp increase of the Fe/Mg ratio in the biotite-bearing granite. The increase of Na plus K relative to Ca is much less pronounced in the amphiboles than in the granites, whereas the amount of Ca in amphiboles F-3 and F-4 is very high relative to the rock compositions. Progressively more reducing conditions accompanied by increase in the Fe<sup>++</sup>/Mg ratio of the magma caused the gradual change in optic and chemical properties from hornblende to ferrohastingsite.

TABLE 2. Composition of Calcic Amphiboles

	Weight Per Cent				
	H-1	F-2	F-3	F-4	
SiO <sub>2</sub>	43.92	41.25	43.49	42.11	
TiO <sub>2</sub>	1.71	1.79	1.33	1.57	
Al <sub>2</sub> O <sub>3</sub>	7.19	6.87	3.84	6.64	
Fe <sub>2</sub> O <sub>3</sub>	5.01	4.83	5.70	4.01	
FeO	17.16	26.77	29.44	26.97	
Li <sub>2</sub> O	0.01	0.01	0.03	0.01	
MnO	0.66	0.64	0.49	0.67	
MgO	8.81	2.99	1.19	3.18	
ZnO	0.08	0.18	0.31	0.17	
CaO	10.60	9.15	8.34	9.31	
Na <sub>2</sub> O	2.01	1.96	2.13	2.26	
K <sub>2</sub> O	0.79	1.05	0.95	1.00	
H <sub>2</sub> O <sup>+</sup>	1.56	1.75	1.86	1.68	
H <sub>2</sub> O <sup>-</sup>	0.27	0.72	1.01	0.29	
TOTAL	99.78	99.96	100.11	99.87	
	Cations per 24 (O, OH)				
Z {	Si	6.72	6.55	6.94	6.72
	Al	1.28	1.29	0.72	1.25
Y {	Al	0.02	-	-	-
	Ti	0.20	0.21	0.16	0.19
	Fe <sup>+++</sup>	0.58	0.58	0.68	0.48
	Fe <sup>++</sup>	2.20	3.56	3.93	3.60
	Li	0.002	0.002	0.007	0.002
	Mg	2.01	0.71	0.28	0.76
	Zn	0.01	0.02	0.04	0.02
	Mn	0.08	0.09	0.07	0.09
X+A {	Ca	1.74	1.56	1.42	1.59
	Na	0.60	0.60	0.66	0.70
	K	0.15	0.21	0.19	0.20
OH		1.87	2.62	3.06	2.10
Total X+A		2.49	2.37	2.27	2.49
Total Y		5.10	5.17	5.17	5.14
Total Z		8.00	7.84	7.66	7.97
Fe <sup>++</sup> /Fe <sup>+++</sup>		3.79	6.14	5.78	7.50

H-1 - Hornblende from two feldspar-biotite granite.  
 F-2, F-3, F-4 - Ferrohastingsite from one feldspar-biotite granite. Numbers represent position in traverse from the two feldspar-biotite granite across the one feldspar-biotite granite.

TABLE 3. Composition of Alkali Amphiboles from the Alkaline Granite

	Weight Per Cent					
	A-1	A-2	A-3	O-1	R-1	
SiO <sub>2</sub>	44.85	46.24	47.14	47.13	47.72	
TiO <sub>2</sub>	1.30	1.29	1.11	1.18	1.24	
Al <sub>2</sub> O <sub>3</sub>	1.75	1.66	1.64	1.84	1.29	
Fe <sub>2</sub> O <sub>3</sub>	10.07	10.75	10.89	11.56	13.18	
FeO	26.50	24.85	24.36	23.70	21.99	
Li <sub>2</sub> O	0.09	0.14	0.13	0.16	0.28	
MnO	0.45	0.39	0.37	0.34	0.43	
MgO	0.58	0.60	0.05	0.20	0.22	
ZnO	0.38	0.40	0.39	0.39	0.42	
CaO	3.95	3.82	4.20	3.20	1.82	
Na <sub>2</sub> O	5.24	5.94	5.82	6.01	7.24	
K <sub>2</sub> O	1.04	1.05	1.06	1.04	1.15	
H <sub>2</sub> O <sup>+</sup>	2.08	2.03	2.64	2.18	2.53	
H <sub>2</sub> O <sup>-</sup>	1.70	0.82	0.34	0.67	0.27	
TOTAL	99.98	99.98	100.14	99.60	99.78	
	Cations per 23 (O, OH)					
Z	{ Si	7.37	7.46	7.57	7.57	7.63
	{ Al	0.34	0.32	0.31	0.35	0.24
	{ Fe <sup>+++</sup>	0.29	0.22	0.12	0.08	0.13
Y	{ Al	-	-	-	-	-
	{ Ti	0.16	0.16	0.13	0.14	0.15
	{ Fe <sup>+++</sup>	0.96	1.09	1.20	1.32	1.46
	{ Fe <sup>++</sup>	3.64	3.36	3.27	3.19	2.94
	{ Li	0.02	0.03	0.03	0.04	0.06
	{ Mg	0.14	0.14	0.01	0.05	0.05
	{ Zn	0.04	0.05	0.05	0.05	0.05
	{ Mn	0.06	0.05	0.05	0.05	0.06
X+A	{ Ca	0.70	0.66	0.72	0.55	0.31
	{ Na	1.67	1.86	1.81	1.87	2.25
	{ K	0.22	0.22	0.22	0.21	0.24
OH		2.00	2.00	2.00	2.00	2.00
Total X+A		2.59	2.74	2.75	2.63	2.80
Total Y		5.02	4.88	4.74	4.84	4.77
Total Z		8.00	8.00	8.00	8.00	8.00
Fe <sup>++</sup> /Fe <sup>+++</sup>		2.91	2.56	2.48	2.28	1.85
A-1, A-2, A-3	- Arfvedsonite					
O-1	- Osannite					
R-1	- Riebeckite					

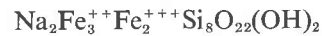
### Alkali Amphiboles

The chemical data (Table 3) indicate a systematic relationship between the alkali amphiboles corresponding to their different optic orientations and crystallization sequence. This is first evident in the uniform increase of Fe<sup>+++</sup> (and decrease of Fe<sup>++</sup>) from arfvedsonite through osannite to riebeckite (Fig. 1). The possibility that the oxidation state of iron is related to the optic orientation of the amphibole is consistent with the experimental observation of Ernst (1962, 1968) that riebeckite is stable under relatively oxidizing conditions at temperatures as low as 500°C. Higher temperatures and more

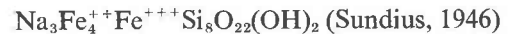
reducing conditions cause the amphibole composition to shift toward arfvedsonite at the more ferrous end of the riebeckite-arfvedsonite solid solution series. Furthermore, in Ernst's experiments the Si proportion of the amphibole diminished as the iron became more reduced. The experimental work of Ernst (1962) and the chemical variation and crystallization sequence of the Golden Horn amphiboles both indicate that amphibole composition is a function of oxygen fugacity and temperature as well as of magma composition.

The influence of magma composition and time of crystallization on crystal chemistry will be illustrated by comparing the Golden Horn amphiboles with the following formulae:

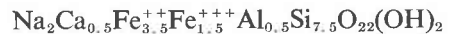
riebeckite



arfvedsonite (end-member)



arfvedsonite (natural)



(Miyashiro, 1957)

None of the Golden Horn alkali amphiboles approaches the theoretical end-member riebeckite composition. Instead, the Ca content ranges between 0.72 atoms in the earlier formed arfvedsonite and 0.31 in the later formed riebeckite, with osannite being intermediate. Figure 2 shows that Ca + Al decreases as Na + K + Si increases in the series from arfvedsonite to osannite to riebeckite. The Golden Horn arfvedsonites are very similar to natural arfvedsonite, and the osannite and riebeckite lie almost exactly on a line between Miyashiro's natural arfvedsonite and Sundius's end-member arfvedsonite with the riebeckite being closest to the end-member arfvedsonite. A systematic relationship between arfvedsonite, osannite, and riebeckite results from the substitution of Fe<sup>+++</sup>Si for CaFe<sup>++</sup>Al. A less important substitution is NaSi for CaAl. The arfvedsonite incorporated much of the available Ca thereby leaving riebeckite to crystallize in a Ca-poor environment as oxygen fugacity rose.

### Summary

Field, petrographic, and geochemical examination of the Golden Horn amphiboles indicates that the crystallization sequence with decreasing tem-

perature was hornblende  $\rightarrow$  ferrohastingsite  $\rightarrow$  discontinuous reaction  $\rightarrow$  arfvedsonite  $\rightarrow$  osannite  $\rightarrow$  riebeckite. A similar, although not identical, sequence was recognized by Aoki (1964) in alkaline volcanic rocks of Japan. The sequence is closely paralleled by changes in the host rock chemistry. The change from hornblende to ferrohastingsite is accompanied by increase in the Fe/Mg and  $(\text{Na} + \text{K})/(\text{Na} + \text{K} + \text{Ca})$  ratios of the bulk rocks and associated biotite and feldspar. Changes in the oxidation state of iron indicate that oxygen fugacity was decreasing as the calcic amphiboles crystallized.

The alkali amphiboles of the Golden Horn granite represent an example of the stability relationships of the riebeckite-arfvedsonite solid solution series experimentally observed by Ernst (1962). Crystallization proceeded from arfvedsonite to riebeckite with  $\text{Fe}^{++}\text{Si}$  substituting for  $\text{CaFe}^{++}\text{Al}$  as temperature fell, oxygen fugacity rose, and the Ca content of the melt decreased.

Zoned crystals in the alkaline granite provide the most rigorous evidence of the relationship between the calcic and alkali amphiboles of the Golden Horn Batholith. Crystals containing ferrohastingsite cores but arfvedsonite rims indicate that crystallization of a calcic amphibole preceded, at least for a short time, the alkali amphiboles. The irregular ferrohastingsite core boundary and the scarcity of ferrohastingsite crystals in the alkaline granite suggest that the calcic and alkali amphiboles are related through a discontinuous reaction as originally proposed by Billings (1928). Decreased temperature, rising oxygen fugacity, and a transition of the magma composition

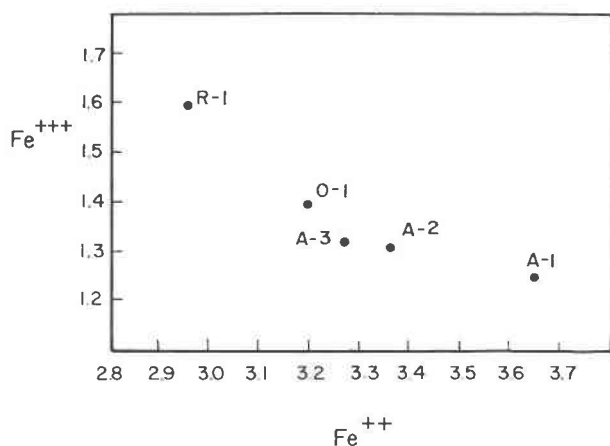


FIG. 1. The ferrous and ferric iron content of riebeckite (R-1), osannite (O-1), and arfvedsonite (A-1, 2, 3).

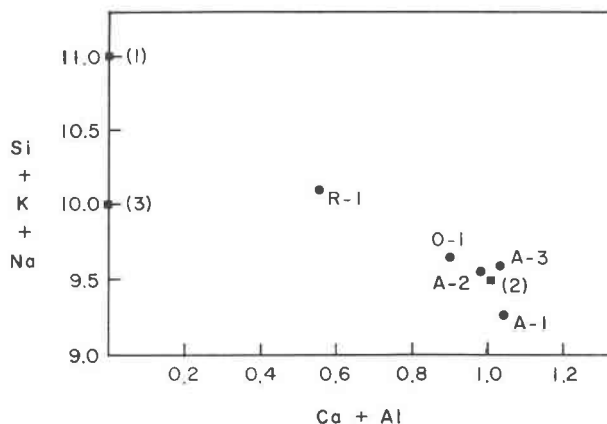


FIG. 2. The variation of  $\text{Ca} + \text{Al}$  with  $\text{Na} + \text{K} + \text{Si}$  in the Golden Horn alkali amphiboles compared with: (1) theoretical arfvedsonite (Sundius, 1946), (2) natural arfvedsonite (Miyashiro, 1957), and (3) theoretical riebeckite.

from peraluminous to alkaline, with the atomic ratio of  $(\text{Na} + \text{K})/\text{Al}$  becoming greater than one, probably caused ferrohastingsite to cease crystallization and arfvedsonite to begin.

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