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Coexisting Actinolite and Hornblende from West-Central New Hampshire

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Abstract

The coexistence of actinolite and hornblende and of cummingtonite and hornblende has been observed in garnet-zone and staurolite-kyanite-zone amphibolites of the Ammonoosuc Volcanics near Hanover, New Hampshire. The amphibole pairs occur both as separate grains and as intergrowths with boundaries that are optically and chemically sharp, but geometrically irregular; no exsolution lamellae were observed. Electron microprobe analyses of the amphibole pairs show that: (1) hornblende is richer in Al, Na, K, Ti, and Fe^{$3+$}, and has a lower Mg/(Mg + Fe) ratio than the coexisting actinolite; (2) hornblende is enriched in Al, Na, and Ca, but has a similar Mg/(Mg + Fe) ratio to that of the coexisting cummingtonite. Recalculations show that the A site is nearly empty in actinolites and typically one-third to one-half full in coexisting hornblendes. Single-crystal precession photos of actinolite-hornblende crystals show that the lattices of the two phases share a common b-axis, but are rotated relative to one another by two to three degrees.

Introduction

The existence of a miscibility gap in the calcic amphiboles has been substantiated only recently (Hallimond, 1943; Shido and Miyashiro, 1959; Klein, 1969), probably because of the optical similarity between coexisting actinolite and hornblende. In addition, the two amphiboles are commonly found together in a patchy intergrowth that may be easily overlooked, rather than as single grains showing exsolution lamellae. Klein (1969) and Cooper and Lovering (1970) have presented analyses for nine actinolite-hornblende pairs from a variety of localities. Although their analyses reveal several important features of the miscibility gap, there are too many variables involved to explain the variety of compositions observed, This report presents new analyses of actinolite-hornblende pairs from several rocks of a single metamorphic grade. It is a first step toward a more detailed understanding of the calcic amphibole miscibility gap.

Geologic Setting of Amphibole Pairs

All the rocks studied were collected from the Middle Ordovician Ammonoosuc Volcanics in westcentral New Hampshire. Specifically, all the amphibole pairs are from cliff-like outcrops on the west flank of the Mascoma gneiss dome, primarily in the Mascoma Quadrangle (location map in Brady, 1970). Details of the local geology may be found in Chapman (1939), Hadley (1942), and Naylor (1968). The sample localities closely straddle a staurolite isograd, samples coming mainly from the staurolite-kyanite zone (Thompson et al., 1968).

The Ammonoosuc Volcanics consist primarily of fine-grained amphibolites with subordinate felsic layers. The formation is believed to be volcanic in origin. Billings (1937, p.476) has described remnant volcanic structures in the less-metamorphosed Ammonoosucs of the Littleton-Moosilauke area, and Chapman (1939, p. 135) found structures resembling those of volcanic tuffs and breccias in the area studied here. The amphibolites contain hornblende and plagioclase as the principal minerals, though epidote is locally abundant. Estimated modes of the polished sections used in electron probe microanalysis are given in Table 1.

Coexisting amphiboles were distinguished primarily on the basis of color and pleochroism. The hornblende is pleochroic deep green to blue-green. Coexisting actinolite is nearly colorless, slightly pleochroic from pale blue-green to green. When more iron-rich, the amphiboles are more highly colored. A difference in birefringence commonly accompanies the color difference. the actinolite having the higher order interference colors. Where cummingtonite coexists

TABLE 1. Estimated Modes of Samples Containing Amphibole Pairs*

	$C-2$		Ğ		$C-6$		17	
hornblende	32		28		77		18	
actinolite			15		6			
cummingtonite	--		--				13	
epidote	40		47				ᅲ	
chlorite					10			
plagioclase		20 (An_{50})	3	(An_{30})	2	$(\text{An}_{1.5})$		16 (An_{25})
quartz			4		3		45	
sphene	1		3					
rutile			tr.**		2			
zircon			$m = 1$				tr.	
biotite			--				tr.	
ilmenite							1	

with hornblende, its colorless crystals and optically positive character permit easy identification.

Contacts between the coexisting amphiboles are optically sharp but may have an irregular shape (Fig. 1A). In some occurrences the two amphiboles appear to form a single crystal that is a "patchwork" of the two phases (see Klein, 1969, Figure 1). Actinolite-hornblende pairs commonly exhibit this

FIG. 1. A. Photomicrograph of sample #C2 in plain-polarized light showing actinolite-hornblende pairs. The grass-green hornblende is the darker mineral. The very pale green actinolite looks colorless in the photo. Note the optically sharp contacts between the two amphiboles which occur both as separate grains and as "patchy" intergrowths. The high relief mineral is epidote; the low relief mineral is plagioclase. The field of view is 0.8×0.8 mm.

relationship. Cummingtonite-hornblende intergrowths are more regular, the two phases sharing ends or sides of the same crystal (Figure 1B). Extinction of both phases of these two-amphibole crystals appears simultaneous. Whether or not these textures are consistent with the postulate of chemical equilibrium between the two amphiboles is an important question which has been addressed by others (Klein, 1969; Stout, 1971). I believe that the presence of a sharp optical and chemical discontinuity between two homogeneous amphiboles is certainly consistent with equilibrium and strong evidence in favor of a miscibility gap. No exsolution lamellae were observed in any of the amphiboles.

Chemical Analyses

Amphibole pairs were analyzed in polished carboncoated thin sections on an Applied Research Laboratories (EMX) microprobe. Nine element analyses were obtained using the methods of Bence and Albee (1968) and Albee and Ray (1970) for an accelerating potential of 15 kV. Seven element analyses were obtained using the standard curve method as described by Klein (1968) for an accelerating potential of 20 kV. The analyses listed in Table 2 have been

FIG. 1. B. Photomicrograph of sample #17 in plain-polarized light showing cummingtonite-hornblende pairs in a quartz matrix. The hornblende is pale green; the cummingtonite is colorless. Note particularly the crystals which have hornblende at both ends with cummingtonite between. The field of view is 0.6×0.6 mm.

unit numbers.
(see text) in terms of least units cited for the stations there are approximately in the distinct of the section of the section of the section of the column of
https://www.terminitedized figures represent one half the difference between the greatest lower bound and least

selected as representative of the 35 amphibole pairs actually analyzed.

The chemical analyses have been recalculated utilizing chemical and stoichiometric constraints to give upper and lower bounds for the value of each cation in the formula unit. This type of recalculation is necessitated by the indeterminacy of the valence state of iron in electron microprobe analyses. It is common practice simply to consider all the iron to be ferrous. However, it is perhaps more useful to use the knowledge available concerning a mineral's structure and stoichiometry to determine maximum and minimum values for ferric iron, between which the "real" value lies. The range of values so determined decreases in size as the iron-content of the mineral increases. A more complete discussion of this type of recalculation procedure is given in Stout (1972).

All recalculations used the following assumptions:

(1) there are 23 oxygens exclusive of $H₂O$ per half unit cell; (2) the analyses are complete except for H_2O ; (3) there are no vacancies except in the A site. One additional assumption is sufficient to specify the ferricferrous ratio. For the calcic amphiboles the most useful numbers are provided by assumptions concerning the occupancy of the $M(4)$ site. Usually, the tightest brackets are obtained by assuming that there is no sodium or potassium in the $M(4)$ site (minimum $Fe³⁺$) or that there is only calcium and sodium in the $M(4)$ site (maximum Fe^{3+}). In some cases, the assumption of all ferrous iron (minimum Fe³⁺) or the assumption that there is only potassium in the A site (maximum Fe^{3+}) will provide more restrictive bounds. Results of computer calculations are presented in Table 2 along with the chemical analyses. The numbers given are the mean of the greatest lower bound and least upper bound recalculated for each analysis. The numbers in parentheses are half the difference of the two bounds in terms of the least units cited for the value to the immediate left.

For the actinolite-hornblende pairs the analyses show the following element fractionations: (1) Actinolite is richer in Mg and Si and very poor in Al, Na, and K. (2) Hornblende is richer in Al, Na, K, Ti, and Fe³⁺ (calc) and has a lower Mg/(Mg + Fe) ratio. The cation sums below the recalculations show that in actinolites the A site is nearly, but not quite, empty. The coexisting hornblendes typically yield values for the A site one-third to one-half full.

The actinolite varies from an ideal two-component composition (tremolite-ferroactinolite) principally utilizing the tschermakite $[A]^{VI}A]^{IV} \rightleftharpoons (Mg, Fe^{2+})^{VI}Si^{IV}$ and edenite $[Na⁴Al^{iv} \rightleftharpoons \Box⁴Si^{iv}]$ substitutions (Fig. 2). 1 The hornblende, however, has a significant

 $\frac{1}{1}$ The superscripts refer to coordination number. A refers to the \vec{A} site, which has 10-12-fold coordination. $M(4)$ refers to the $M(4)$ site, which has 6-8-fold coordination. \Box means a vacancy.

Edenite

 $[(Fe³⁺)^{VI} \rightleftharpoons Al^{VI}]$ substitution as well, tending toward a magnesio-hastingsite composition. A few of the hornblendes also have a noticeable glaucophane component $[Na^{M(4)}Al^{VI} \rightleftharpoons Ca^{M(4)}(Mg, Fe^{2+})^{VI}].$ Interestingly, Figure 3 shows that alkali content is higher for amphiboles with a higher $Mg/(Mg + Fe)$ ratio, particularly for hornblendes. The tschermakite component also increases with the $Mg/(Mg + Fe)$ ratio. However; these effects may be an accident of bulk composition. In general, the compositions observed are consistent with the trends discussed by Robinson, Ross, and Jaffe (1971a) for the calcic amphiboles and related anthophyllite-gedrite series.

Analyses of coexisting cummingtonite and hornblende show ,element fractionations consistent with the results of'more detailed work by others (Klein, 1968; Robinson and Jaffe, 1969, with wet analyses of two pairs). (l) Cummingtonite is richer in Mg, Mn, Fe, and Si. (2) Hornblende is richer in Ca, Al, and Na and has a slightly higher $Mg/(Mg + Fe)$

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mates the end-member system tremolite-edenite-tschermakite. The ordinate is the calculated A site occupancy per half unit cell (total number of cations minus 15.00), The abscissa is the calculated quantity of tetrahedral aluminum (8.00 minus the number of silicon atoms per half unit cell) minus the calculated Λ site occupancy. These axes were chosen to make the end-member system orthogonal. The dashed tie lines are for pairs for which potassium was not determined.

ratio. Totals for the cummingtonite analyses are low, perhaps because of the standard curves used. The cummingtonite-hornblende data is presented primarily because these pairs were found within meters of the actinolite-hornblende pairs (Fig. 4).

X-ray Crystallography

Selected actinolite-hornblende crystals from several samples were examined using single-crystal precession and rotation X-ray techniques to determine: (l) the possible presence of amphiboles with a primitive monoclinic lattice, (2) the unit-cell parameters of the coexisting phases, and (3) the relative orientations of the amphibole lattices in "two-phase" crystals. Crystals were picked from samples within millimeters of the area of the analyzed amphibole pairs. The "two-phase" crystals chosen, although part green, part colorless, appeared otherwise structurally continuous.

Of the crystals photographed, none showed violations of $C2/m$ symmetry. There did appear to be a consistent relationship in the orientations of the

FIG. 3. Compositions of the actinolite-hornblende pairs are shown here in a comparison of alkali content and magnesium to magnesium plus total iron ratio. The hornblendes have a significant alkali content lacking in coexisting actinolites. The alkali content of the hornblendes is greater for hornblendes with a higher magnesium to magnesium plus total iron ratio. The dashed tie lines are for pairs for which potassium was not determined.

TABLE 3. Unit Cell Parameters for "Two-Phase" Actinolite-Hornblende Crystals*

		Crystal C-2b	Crystal C-6b		
		Hornblende Actinolite	Hornblende Actinolite		
(λ) α	9.84	9.86	9.83	9.81	
(A) Ъ	18.12	$n.d.**$	18.07	n.d.	
(A) \mathcal{C}	5.30	5.28	5.30	5.30	
$B =$	104°52"	104°48'	104°55'	104°54"	
Space group	C2/m	C2/m	C2/m	C2/m	

* Results of measurements of precession photos. Cell edges are precise to \pm 0.5 percent, angular values to \pm 3'. $** n.d. = not determined$

lattices of the actinolite-hornblende intergrowths. Three different actinolite-hornblende crystals showed lattices orientated within 2 to 3 degrees of rotation $(+)$ or $-)$ about a common b axis. The b axes were very nearly, though not exactly, coincident. The unit cells of the two phases are quite similar (Table 3).

The orientation of exsolution lamellae in horn' blende-cummingtonite crystals has been discussed by Robinson et al (1971b). These authors conclude that the two amphibole phases are interfaced along planes of "best fit" of the two lattices. The irregular intergrowths of actinolite and hornblende observed in this study, while possibly formed by a different process, clearly are not interfaced on a plane of best fit. The intergrowths do show a close relative lattice orientation, however. It follows that one of the amphibole phases (or a homogeneous parent) pre-dates the other and that the orientation of the second amphibole lattice was controlled by that of the first. The mechanism of this structural control may involve a nucleation or a replacement process.

The textures presented by patchy, two-amphibole crystals are most intriguing. Why are the contacts between the two phases commonly so irregular? Such boundaries seem to be far from representing a minimization of the potential energy of the interface. It may be that the assemblages themselves are a long way from being equilibrium assemblages. On the other hand, it may be that the potential energy per unit area of interface is very low, due to the similarity of the unit cells of the phases involved. If so, then there is little to be gained, thermodynamically, by a more regular geometry. Evidence in favor of the latter view may be found in an examination of the list of amphibole pairs that exhibit the irregular intergrowth texture. This list includes actinolitehornblende, glaucophane-hornblende, actinolite-glaucophane (Klein, 1969), and anthophyllite-gedrite

Ftc. 4. Compositions of both actinolite-hornblende pairs and cummingtonite-hornblende pairs are shown on a portion of the triangle CaO- $(A1_2O_3-Na_2O)$, MgO, FeO + MnO, which is the plagioclase projection of Robinson and Jaffe (1969). This diagram conveniently shows relationships between the compositions of the cummingtonite-hornblende pairs (found within meters of actinolite-hornblende sample $#G$) and the compositions of the actinolite-hornblende pairs.

(Stout, 1971)². Noticeably absent from the list are pairs consisting of cummingtonite and a calcic or sodic amphibole, calcic amphibole-orthoamphibole pairs, or sodic amphibole-orthoamphibole pairs. These are the combinations which have the greatest contrast in structure and, therefore, possibly the greatest potential energy reduction due to a regulated interface geometry.

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