

FOUR COEXISTING AMPHIBOLES FROM TELEMARK, NORWAY

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ABSTRACT

A mineral assemblage that includes anthophyllite, gedrite, cummingtonite, and hornblende in an amphibolite from southern Norway is described and its petrologic significance discussed. All four amphiboles occur as discrete, optically homogeneous grains within the same bulk compositional layer. Every combination of two and three amphiboles exhibits mutual grain contacts suggesting on the basis of texture alone that the phases have equilibrated.

Electron microprobe analyses coupled with single crystal diffraction studies indicate that each of the four amphiboles is chemically homogeneous as well. This strongly suggests that the phases responded fully to the prevailing external conditions of the staurolite-sillimanite zone and that the effect of possible retrograde metamorphism is negligible. As the stability fields of the four amphiboles overlap, four possible sets of three coexisting amphiboles may be anticipated for appropriate bulk compositions and external conditions.

INTRODUCTION

During the last several years there has been considerable interest in the compositions of coexisting amphiboles, that is, those which occur in mutual contact and presumably have equilibrated under fixed external conditions. Amphibole pairs described by several authors include combinations of cummingtonite, anthophyllite, gedrite, and calcic amphiboles. Perhaps the first description of three coexisting amphiboles is given by Milton (1961) of an assemblage that includes cummingtonite, gedrite, and hornblende. More recently, Robinson (*et al.*, 1969) describes the equilibrium coexistence of cummingtonite, anthophyllite, and hornblende and includes electron microprobe analyses of the coexisting phases. Stout (1970a) gives a preliminary account of two additional three-amphibole assemblages, namely cummingtonite-anthophyllite-gedrite, and anthophyllite-gedrite-hornblende. The latter two assemblages complete the four possible three-phase combinations of cummingtonite, anthophyllite, gedrite, and hornblende. Each of the assemblages listed above is stable in the field of sillimanite, and each includes quartz, plagioclase feldspar, ilmenite or magnetite or both.

Any of the above assemblages may be described, as a first approximation, in the system $\text{Al}_2\text{O}_3\text{-MgO-FeO-Fe}_2\text{O}_3\text{-MnO-SiO}_2\text{-CaO-Na}_2\text{O-H}_2\text{O}$. In an environment in which P , T , and $\mu_{\text{H}_2\text{O}}$ are externally controlled, the maximum number of phases for an arbitrary choice of these variables is

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eight. If only those assemblages that include quartz are considered and $\text{FeO} + \text{MnO}$ is treated as a composite component, the maximum number of phases in a typical three-amphibole assemblage is still in accordance with the restrictions imposed by the phase rule. These considerations suggest that, in the absence of quartz or feldspar, or both, four amphiboles may coexist stably under arbitrary external conditions within the stability field of sillimanite. Such an assemblage was found in amphibolite-facies rocks from the Precambrian basement of south-central Norway just west of Nisser in Telemark ($59^{\circ}09'36''$ lat., $98^{\circ}20'00''$ long.).

DESCRIPTION OF SPECIMENS

The specimens (101A, 101AB) described here were collected from the marginal part of a metagabbro sill. Coarse amphiboles are conspicuous in the surrounding rocks, all of which have been subjected to at least two episodes of folding and regional metamorphism to the staurolite-sillimanite zone. There is little or no evidence for extensive retrograde metamorphism, nor is there evidence for cataclasis or late stage plutonism that perhaps would obscure the phase equilibria recorded from the earlier metamorphism.

The phases in specimen 101A and their relative abundance are biotite (38 modal percent), anthophyllite (26 percent), gedrite (12 percent), cummingtonite (11 percent), hornblende (10 percent), and a sulfide, probably pyrite (3 percent). Approximately the same percentages of phases are present in specimen 101AB. Trace amounts of ilmenite and a colorless chlorite interleaved with the biotite are the only additional phases. Adjacent compositional layers contain two- and three-amphibole assemblages with or without quartz and feldspar depending on the bulk composition of the rocks (Stout, 1970b).

Electron microprobe analyses of each of the four amphiboles in assemblage 101A and of coexisting anthophyllite and gedrite in assemblage 101AB are presented in Table 1. The polished sections used in the analytical work contain circular areas one millimeter in diameter in which all four amphiboles occur as discrete optically, homogeneous grains. Although all four were never observed in mutual contact, all other combinations of two and three amphiboles exhibiting mutual contacts were observed (Figure 1). Anthophyllite and gedrite are the only phases that may be readily mistaken on the basis of their optical properties alone, but with high magnification and an intense light source, the pleochroism of the latter distinguishes it from the colorless anthophyllite (Table 4). Optically, the colorless cummingtonite is easily recognized by its inclined extinction. The calcic amphibole has the absorption of typical blue-green hornblende.

ANALYTICAL PROCEDURE

The analyses in Table 1 were determined using standard curves made up of homogeneous, naturally occurring amphiboles for which wet chemical analyses were available. The standards are the same as those described by Klein (1969), and the analyses were done on the same instrument. An accelerating potential of 20 kV was used with a sample current of 0.03 microAmperes. X-ray counts were recorded over the time necessary to accumulate 10,000 counts on a beam current monitor. This minimizes drift due both to filament warpage and instability in the high voltage supply. During the several hours required to complete the analyses, no drift was detected.

Continuous scans across grain contacts for $K\alpha$ of seven elements were made using an X-Y recorder. This was done in order to define chemically the immiscibility of anthophyllite and gedrite and to graphically portray their compositional differences.

TABLE 1. ELECTRON MICROPROBE ANALYSES OF COEXISTING AMPHIBOLES, ANTHOPHYLLITE (101A), GEDRITE (101A), CUMMINGTONITE (101A), AND HORNBLLENDE (101A), WITH ANALYSES FOR COEXISTING ANTHOPHYLLITE (101AB) AND GEDRITE (101AB) FROM AN ADJACENT SPECIMEN

	Anthophyllite 101A				Gedrite 101A				Cummingtonite 101A			
	Wt. %	σ	σ/\sqrt{N}	No. of det.	Wt. %	σ	σ/\sqrt{N}	No. of det.	Wt. %	σ	σ/\sqrt{N}	No. of det.
SiO ₂	52.4	0.4	1.9	40	45.0	0.4	2.0	40	53.5	0.4	1.3	40
Al ₂ O ₃	3.1	0.2	1.6	40	11.7	0.4	2.5	40	1.5	0.3	1.6	40
FeO*	21.4	0.2	1.0	20	21.8	0.2	1.0	20	22.0	0.2	1.4	20
MnO	0.5	0.04	1.0	30	0.5	0.03	1.1	30	0.5	0.04	1.0	30
MgO	18.3	0.4	1.4	20	15.8	0.6	1.9	20	18.5	0.2	1.0	20
CaO	0.3	0.03	1.4	20	0.7	0.1	1.7	20	0.6	0.1	3.6	20
Na ₂ O	0.4	0.03	1.3	30	1.7	0.03	1.3	20	0.2	0.02	1.1	20
Total	96.4				97.2				96.8			
	Hornblende 101A				Anthophyllite 101AB				Gedrite 101AB			
SiO ₂	43.2	0.4	1.7	40	52.4	0.4	1.6	20	44.3	0.4	1.1	20
Al ₂ O ₃	15.7	0.2	1.2	40	3.4	0.1	4.3	100	12.6	0.1	2.0	100
FeO*	15.8	0.2	1.3	20	21.7	0.1	1.0	100	21.8	0.1	1.1	100
MnO	0.3	0.03	1.0	30	0.5	0.03	1.0	20	0.6	0.03	1.0	20
MgO	9.7	0.2	1.0	20	18.3	0.1	1.7	100	15.8	0.1	1.9	100
CaO	10.3	0.1	1.0	20	0.3	0.05	1.7	20	0.8	0.04	1.0	20
Na ₂ O	2.2	0.03	1.2	30	0.4	0.02	2.5	20	1.8	0.02	1.8	20
Total	97.2				96.4				97.6			

FeO* denotes total Fe as FeO. σ is one standard deviation, and \bar{N} is the arithmetic mean of X-ray counts. The number of determinations for each element in each phase is given in the fifth column in each analysis. A single determination constitutes 10,000 counts on a beam current monitor recorded over a time interval of approximately 15 seconds.

RECALCULATION OF ANALYSES

Analyses of the two gedrites are recalculated on the basis of both 15 cations (excluding Na) and 23 oxygen atoms (Tables 2 and 3). Because the ferric component is not known in a microprobe analysis, the "all ferrous" assumption, *i.e.*, that obtained by treating total Fe as FeO in the 23 oxygen recalculation, may not be the best approximation for these phases. Both gedrites, when recalculated on the basis of 23 oxygen atoms, have octahedral sums excluding Na in excess of 7. On that basis, Ca would be assigned to the "A" site contrary to its usual assignment to the M(4) site (see Ghose, 1961, for nomenclature). A more plausible alternative is to consider a recalculation in which some fraction of the total Fe is in the ferric state. This would effectively reduce the sum of cations normally thought to occupy M sites such that Ca may be accommodated in M(4) rather than in the "A" site.

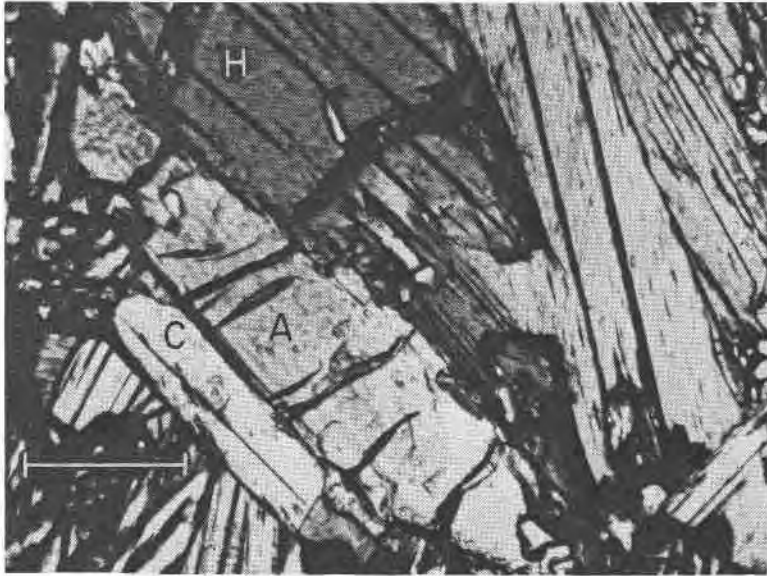


FIG. 1. Coexisting anthophyllite (A), cummingtonite (C) and hornblende (H) in specimen 101A. Coarse gedrite is one millimeter away. Length of bar is 0.25 mm.

TABLE 2. RECALCULATION OF AMPHIBOLE ANALYSES ON THE BASIS OF FIXED NUMBERS OF CATIONS (15 CATIONS EXCLUDING NA FOR THE NON-CALCIC AMPHIBOLES AND 13 CATIONS EXCLUDING CA AND NA FOR HORNBLENDE 101A)

	Antho 101A	Ged 101A	Cum 101A	Hnbl 101A	Antho 101AB	Ged 101AB
Si	7.71	6.64	7.85	6.27	7.66	6.50
Al	0.29	1.36	0.15	1.73	0.34	1.50
Al	0.25	0.67	0.11	0.95	0.25	0.68
Fe	2.63	2.68	2.69	1.91	2.65	2.67
Mn	0.05	0.06	0.06	0.04	0.06	0.07
Mg	4.02	3.48	4.05	2.10	3.99	3.45
Ca	0.05	0.11	0.09	1.60	0.05	0.13
Na(M4)	—	—	—	0.40	—	—
Na(A)	0.11	0.49	0.05	0.22	0.11	0.50
$\frac{100Mg}{Mg+Fe}$	60.4	56.3	60.1	52.2	60.0	56.5
$\frac{1}{2}Al$	3.9	14.1	1.9	25.1	4.2	15.1
Fe	38.0	37.5	39.2	39.1	38.3	37.0
Mg	58.1	48.4	58.9	35.8	57.5	47.9

TABLE 3. RECALCULATION OF AMPHIBOLE ANALYSES ON THE BASIS OF 23 OXYGEN ATOMS

	Antho 101A	Ged 101A	Cum 101A	Hnbl 101A	Antho 101AB	Ged 101AB
Si	7.70	6.67	7.84	6.40	7.66	6.54
Al	0.30	1.33	0.16	1.60	0.34	1.46
Al	0.23	0.71	0.10	1.14	0.25	0.74
Fe	2.62	2.70	2.69	1.95	2.65	2.69
Mn	0.06	0.06	0.06	0.04	0.06	0.08
Mg	4.01	3.49	5.05	2.14	3.99	3.48
Ca	0.05	0.11	0.09	1.64	0.05	0.13
Na(<i>M</i> 4)	0.03	—	0.01	0.09	—	—
Na(<i>A</i>)	0.08	0.49	0.05	0.54	0.11	0.50

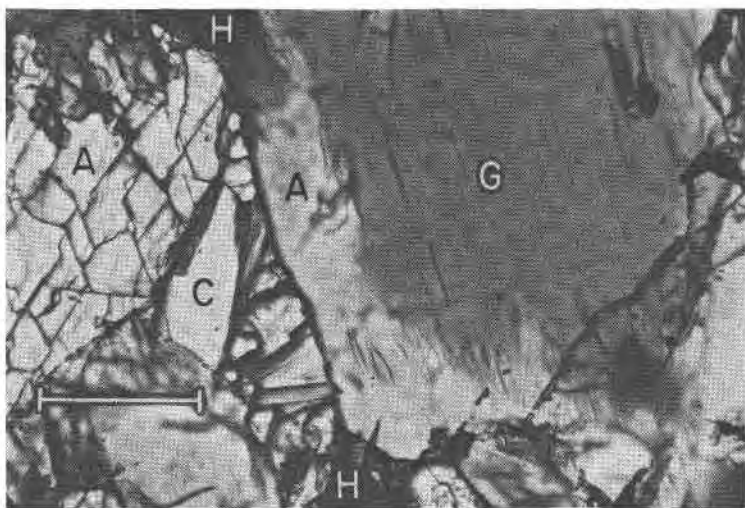


FIG. 2. Composite orthoamphibole grain coexisting with homogeneous cummingtonite (C), hornblende (H), and anthophyllite (A) in specimen 101A. Length of bar is 0.25 mm.

In contrast, similar recalculations of two anthophyllite analyses and one cummingtonite analysis (Tables 2 and 3) indicate that the "all ferrous" assumption is quite reasonable for these phases. The sum of cations in *M* sites is close or equal to 7 based on 23 oxygen atoms, and the oxygen totals based on the cation recalculations are close to 23.

Hornblende 101A is recalculated with the additional assumption that

the $M(4)$ site is occupied solely by Ca and perhaps Na. This assumption is not strictly valid because the hornblende coexists with both anthophyllite and cummingtonite and thus must be saturated with respect to those components. Accordingly, Fe^{+2} and Mg are probably present in $M(4)$, but as their exact amounts in this site are not known, the 13 cation recalculation must be considered as only an approximation to the actual cell contents. Similarly, the recalculation based on 23 oxygen atoms (Table 3) remains an approximation as well since the important ferric component is neglected.

HOMOGENEITY OF THE ANALYZED PHASES

There is little evidence for chemical inhomogeneities in the analyzed phases. Standard deviations and homogeneity factors¹ (Boyd, 1967) were calculated from the $K\alpha$ intensities for each of the elements listed in oxide form in Table 1. The normal operating procedure is to record X-ray counts twice at a minimum of 10 localities on each phase, but as 20 determinations may not represent a statistical sampling, 100 determinations were made for Fe, Mg, and Al in anthophyllite 101AB and gedrite 101AB as a check for homogeneity. A homogeneity factor less than three indicates that at the 99 percent confidence level, individual counts may be expected to lie within ± 3 standard counting errors. If higher values are taken to indicate chemical inhomogeneity, only Al in anthophyllite 101AB shows any degree of inhomogeneity. The remaining values of σ/\sqrt{N} provide no evidence for inhomogeneity of the other elements. The low values of σ/\sqrt{N} in general suggest that the phases are homogeneous relative to the chemical differences between them.

SINGLE CRYSTAL DIFFRACTION DATA

Cummingtonite. Precession photographs were taken of three cummingtonite crystals handpicked from specimen 101A. The diffraction pattern of each is consistent with a C2/m space group which is normal for most cummingtonites. None of the primitive reflections such as those described by Ross (*et al.*, 1968), Robinson (*et al.*, 1969), and others in other multi-amphibole assemblages were recognized. Only optically homogeneous crystals were selected for study, and, of these, none show diffraction that can be attributed to exsolution of hornblende. The ratio 100 Mg/(Mg+Fe) for cummingtonite 101A is 60.1, compared to 64.6 (Robinson, *et al.*, 1969) and higher values (e.g., Kisch, 1969) for primitive cummingtonites low in Ca and Mn.

¹ σ/\sqrt{N} where σ = one standard deviation; N = arithmetic mean of X-ray counts.

Anthophyllite and gedrite. Precession photos were taken of single, homogeneous crystals of both anthophyllite and gedrite, as well as single grains that are visible mixtures of these phases. Both phases have a diffraction pattern consistent with either $Pnma$ or $Pn2_1a$ space groups depending on whether the structure is centrosymmetric or not. Optically homogeneous grains of each phase, whether in oil immersion mounts (Figure 3) or in the thin section, show no uniformly distributed lamellae such as those described by Robinson (*et al.*, 1970) parallel to $\{010\}$ of anthophyllites thought to have formed by exsolution. However, some contacts between anthophyllite and gedrite in heterogeneous grains of this study do have that orientation.

Better resolution in the back reflection region was obtained with an equi-inclination Weissenburg apparatus. Lattice parameters (Table 4) measured from back reflection Weissenburg photographs are accurate to $\pm 0.01 \text{ \AA}$. Values for a and c are identical for the two phases. The values of b , however, differ significantly, being smaller for gedrite. Their values differ only slightly from those reported by Robinson (*et al.*, 1969) in mixed crystals thought to be of exsolution origin.

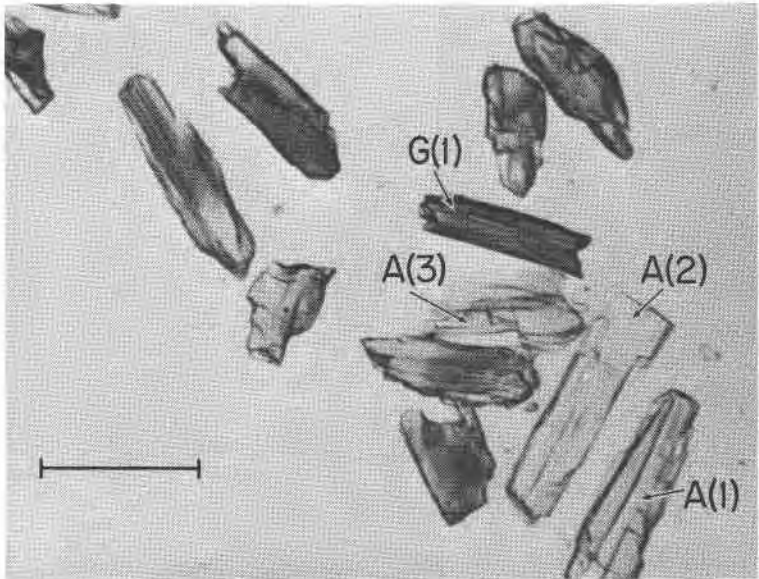


FIG. 3. Oil immersion mount of orthoamphibole grains from specimen 101A. Grains A(1), A(2) and A(3) are homogeneous anthophyllite. Grain G(1) is gedrite with a thin lamella of anthophyllite. The other grains are composite, made up of various proportions of the orthorhombic phases. Length of bar is 0.5 mm.

TABLE 4. OPTICAL PROPERTIES AND LATTICE PARAMETERS OF COEXISTING AMPHIBOLES

	Anthophyllite 101A	Gedrite 101A	Cummingtonite 101A	Hornblende 101A
α	1.648 ± .002	1.655 ± .062	—	—
β	1.650 ± .002	1.662 ± .002	—	—
γ	1.663 ± .002	1.668 ± .002	1.667 ± .002	1.670 ± .002
$\gamma - \alpha$	0.015 ± .004	0.013 ± .004	—	—
$c \wedge Z$			21°	17°
$2V\gamma$	75–80°	75–80°	—	110–115°
(estimate)				
dispersion	$r < v$, moderate	$r < v$, moderate	—	—
a , Å	18.58	18.58	9.40	—
b , Å	18.08	17.89	18.08	—
c , Å	5.30	5.30	5.30	—
			($\beta = 102^\circ 10'$)	
Absorption				
X	colorless	colorless	colorless	straw
Y	colorless	pale greenish yellow	colorless	green
Z	colorless	greenish gray	colorless	bluish green

TEXTURAL EVIDENCE FOR EQUILIBRATION

Without exception, the intercrystalline contracts between all phases in the two specimens studied are sharp and well defined (Fig. 1). Two-phase contacts made up one homogeneous amphibole grain in contact with another, but different amphibole, are commonly observed. Three-amphibole contacts where three amphiboles are in mutual contact at a point are less common but observed for the four combinations possible. Four-amphibole contacts were not observed, probably because of the mechanical difficulty of observing any four phases in mutual contact in a plane.

Single grains of cummingtonite and hornblende are optically homogeneous in nearly all cases. The few exceptions are restricted to irregular shaped areas of cummingtonite in hornblende and, similarly, areas of hornblende in cummingtonite. Many of the ortho-amphibole grains, however are, composite, being made up of various proportions of anthophyllite and gedrite. In some cases gedrite may seem to rim anthophyllite, but just the reverse relationship is equally common (Fig. 2). Intergrowths of the two phases parallel to the prismatic zone are also common, as are tiny blebs of one phase in larger, homogeneous parts of the other phase.

Some of the textures seen here, particularly those *within* individual grains of orthoamphibole, could be interpreted as due to intracrystalline exsolution of two phases. Such a phenomenon could conceivably have taken place after the primary crystallization, perhaps during cooling, and thus not be related to the pressure and temperature of the primary metamorphism. Textural relationships between coexisting amphiboles in high-grade metamorphic rocks described by Ross (*et al.*, 1969) and others are similar to those in pyroxenes from large, ultramafic bodies, such as the Skaergaard, Bushveld, and Stillwater, for which there is ample evidence for slow cooling through a temperature interval below the solidus. Pyroxenes that exsolve under such conditions typically show a regularly repeated pattern of the exsolved phase and host which is uniform throughout any given grain. Prior to exsolution it seems likely that each pyroxene grain in a given rock would have a fixed composition identical to that of its immediate neighbors, implying that after exsolution, the volume ratio of exsolved phase to host would remain constant from one grain to the next. These features, taken as indicative of sub-solidus exsolution, are not observed in the anthophyllite-gedrite intergrowths reported in this paper. To the contrary, composite grains made up of the two phases exhibit the complete range of volume ratios from optically homogeneous anthophyllite to optically homogeneous gedrite. This is shown by an oil immersion mount of single orthamphibole grains from specimen 101A (Figure 3). Furthermore, the electron probe analyses give no indication that the composition of a small volume of anthophyllite, for example, in a large grain of gedrite is any different than the composition of a discrete, homogeneous grain of anthophyllite. Thus, it appears that phases in this study that are optically homogeneous are chemically homogeneous as well.

Intracrystalline segregation of two exsolved phases due to post-metamorphic or post-igneous recrystallization has been pointed out by Ross (*et al.*, 1969) as a possible explanation of the irregular distribution of exsolved phases in single grains of metamorphic amphibole. It seems unlikely that such a process operated in the case of the ortho-amphiboles discussed here, as the criterium that the volume ratios of exsolved phase to host remain constant from grain to grain for a given bulk composition is not satisfied.

CHEMICAL EVIDENCE FOR EQUILIBRATION

The invariant composition of each amphibole throughout the examined sections is taken to represent equilibration of these phases under the same external conditions. In addition to the analyses presented in Table 2, counting rates of Fe⁻, Mg⁻, and AlK α for phases about several

two- and three-amphibole contacts were examined in section 101A. Regardless of the specific amphibole in mutual contact with cummingtonite, for example, the counting rates for these elements do not change within the standard deviations of the determinations. Similarly, the compositions of each of the other amphiboles are constant regardless of the compositions of adjacent phases. Such chemical homogeneity, implies that each phase responded fully to the prevailing temperature and pressure of metamorphism. In the absence of a special "point" contact where all four amphiboles are in mutual contact, this approach seems the best suited one to insure that the analyzed phases are in equilibrium.

Calculated distribution coefficients (Stout, 1970b) for the six amphibole pairs which comprise the four-amphibole assemblage indicate that the fractionation of Fe and Mg between any two coexisting amphiboles is the same as between members of corresponding pairs from surrounding rocks. The latter pairs include coexisting cummingtonite and hornblende, anthophyllite and hornblende, and anthophyllite and cummingtonite. If a disequilibrium hypothesis is proposed, then it would follow that the partitioning of Fe and Mg between coexisting phases is insensitive to temperature change. As this seems unlikely, I favor the interpretation that the four amphiboles in question did in fact equilibrate, most probably at the thermal peak of metamorphism.

Anthophyllite-gedrite miscibility gap. One method that may be employed to recognize gaps in solid solution in a mineral series is to look for chemical discontinuities in the graphical representation of available analyses. In the case of the "anthophyllite series," this has been done by Sundius (1932) and Rabbitt (1948). The method suffers not only from difficulties in sampling all of the compositional space in which anthophyllite and gedrite are stable, but also because no regard is usually taken of the possible changes in the compositional fields of these phases brought about by changes in metamorphic grade. The latter consideration, in principle, is enough to completely camouflage gaps in solid solution if the analyses are chosen at random. Thus Rabbitt (1948) concluded that the name gedrite should be discarded and that all members of the series be referred to as anthophyllite. More recently Robinson *et al.* (1969) showed on the basis of single crystal diffraction studies that anthophyllite and gedrite are distinct phases, occurring together as exsolution products. The results of the present study show as well that anthophyllite and gedrite are distinct phases and may occur as an equilibrium pair in a suitable environment.

Figure 4 illustrates the fractionation of major elements between coexisting anthophyllite and gedrite in section 101A. Continuous micro-

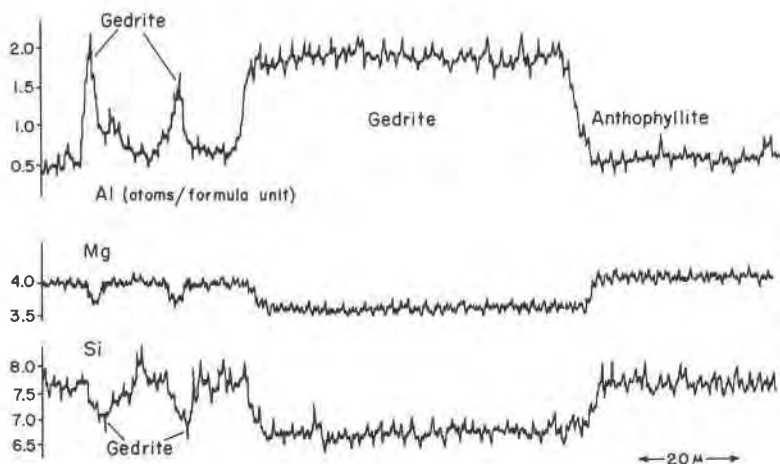


FIG. 4A. Continuous electron microprobe scan for AlK α , MgK α and SiK α across a composite orthoamphibole grain similar to that shown in Figure 2.

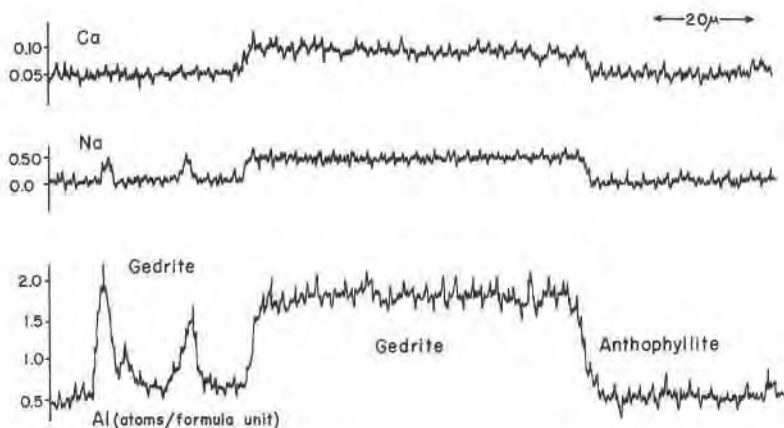


FIG. 4B. Same as in Figure 4A but for CaK α and NaK α with SiK α again for comparison.

probe scans were made across a single, composite grain similar to the one shown in Figure 2. Al, Na, and Ca are concentrated in the gedrite, whereas Si and Mg are concentrated in the anthophyllite. A similar scan for Fe K α is not shown because Fe is present in equal amounts in both phases, as shown by the analyses in Table 1. The gedrite, however, has a lower Mg/(Mg+Fe) than the anthophyllite, indicating that the intracrystalline partitioning with respect to this ratio is a function of relative Mg contents alone.

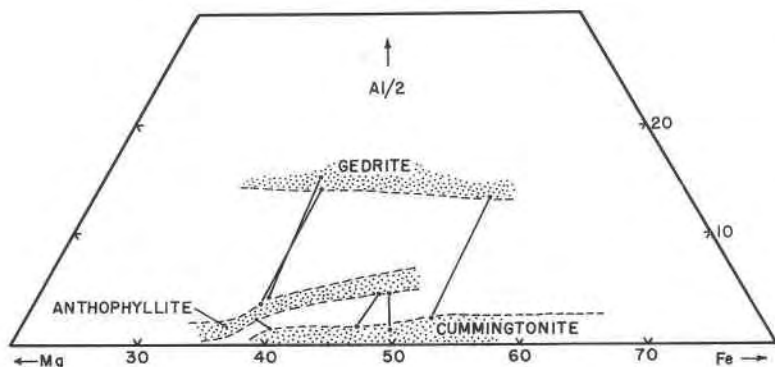


FIG. 5. Amphibole quadrilateral illustrating the generalized compositional fields of anthophyllite and gedrite irrespective of metamorphic grade. Anthophyllite-cummingtonite pairs from Klein (1969) and a gedrite-cummingtonite pair from Eskola (1936) are shown in addition to 101A and 101AB of this study.

The approximate compositional fields of anthophyllite, gedrite, and cummingtonite are shown in the amphibole quadrilateral of Figure 5 without regard to metamorphic grade. In this representation the least aluminous gedrites generally coexist with less aluminous phases such as cummingtonite or anthophyllite, again depending on the bulk compositions considered. The single gedrite-cummingtonite tieline from Eskola (1936) shown in Figure 5 places a restriction on the compositional field of anthophyllite, namely that it does not extend to more Fe-rich compositions than those represented by the tieline. Anthophyllite-cummingtonite pairs, taken from Klein (1969) are also shown in addition to the anthophyllite-gedrite pairs in this report.

The chemical extent of the miscibility gap in the anthophyllite-gedrite series can be rigorously determined only by examining the compositions of anthophyllite-gedrite pairs for a variety of bulk compositions in rocks recrystallized under the same external conditions. The coexistence of four amphiboles without feldspar or quartz implies that the stabilities of each of the amphiboles overlap. Thus, four different three-amphibole assemblages could occur in rocks bearing feldspar \pm quartz at some particular set of external conditions. Two of those assemblages, namely anthophyllite-gedrite-cummingtonite and anthophyllite-gedrite-hornblende, may provide data on the limiting compositions of the anthophyllite solid solution for feldspar-bearing assemblages. A preliminary investigation of three-amphibole assemblages in the Telemark area (Stout, 1970a) suggests that the composition of the anthophyllite in the former assemblage is the most Fe-rich possible.

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