

TABLE 1. RECENT REPORTS OF EXSOLUTION IN AMPHIBOLES

Reference	Rock type	Host amphiboles	Exsolved amphiboles
Seitsaari (1952) p. 11	amphibolite, Perniö, Finland	hornblende	cummingtonite on ($\bar{1}01$)
Asklund, Brown, and Smith (1962)	amphibolite, Sköthagen, Sweden	hornblende cummingtonite	cummingtonite on ($\bar{1}01$) hornblende on ($\bar{1}01$)
Vernon (1962)	amphibolite, NW Queensland, Australia	hornblende cummingtonite	cummingtonite on ($\bar{1}01$) hornblende on ($\bar{1}01$)
Robinson (1963)	amphibolites, Orange area, Mass., N. Hampshire	hornblende cummingtonite	cummingtonite on (100), ($\bar{1}01$) hornblende on (100), ($\bar{1}01$)
Binns (1965)	amphibolites, gneisses, New South Wales, Australia	hornblende	cummingtonite on ($\bar{1}01$), (100)
Boriani and Minutti (1965)	cummingtonite amphibolite, Novara, Italy	cummingtonite	hornblende on ($\bar{1}01$)
Bown (1965)	talc schist, Gouverneur, N. Y.	tremolite	$P2_1/m$ clinoamphibole on ($\bar{1}01$)
Callegari (1966)	quartz-biotite amphibolite, Adamello, Italy	hornblende cummingtonite	cummingtonite on (100), ($\bar{1}01$) hornblende on (100), ($\bar{1}01$)
Jaffe, Robinson, and Klein (1968)	metamorphosed volcanic rocks, Mass., N. H.	hornblende cummingtonite	cummingtonite on (100), ($\bar{1}01$) hornblende on (100), ($\bar{1}01$)
Ross, Smith, and Ashton (1968)	talc schists, Gouverneur, N. Y.	tremolite	primitive manganooan cummingtonite on ($\bar{1}01$)
Ross, Papike, and Weiblen (1968)	talc schists, Gouverneur, N. Y.	tremolite primitive manganooan cummingtonite	primitive manganooan cummingtonite on ($\bar{1}01$) tremolite on ($\bar{1}01$)
Robinson, Jaffe, Klein, and Ross (1969)	metamorphosed volcanic rocks, Orange area, Mass.	hornblende, primitive cummingtonite anthophyllite	primitive cummingtonite on (100), ($\bar{1}01$) hornblende on (100), ($\bar{1}01$) gedrite on (100, 010)

key amphiboles reported on in this study: tremolite (G-21, Table 2), primitive manganooan cummingtonite (No. 115046, Table 2), C-centered manganooan cummingtonite (No. 2, Table 8). The details of the crystal structures are given by Papike, Ross, and Clark (1969).

CLINOAMPHIBOLES FROM THE GOUVERNEUR MINING DISTRICT

General geology and sample description. The talc-amphibole schists of the Gouverneur mining district occur as lenses and pods associated with marbles, amphibolites, and gneisses of the Precambrian Grenville Series, St. Lawrence County, New York. Engel and Engel (1953, 1958, 1960, and 1962), and Engel (1962) have described in detail the petrology of these rocks, as well as evidences of progressive regional metamorphism along a 35 mile section of the major paragneiss of this area. Maximum temperatures of metamorphism increased from southwest to northeast,

reaching about 525°C near Emeryville, New York, and increasing to about 625°C near Colton. Metamorphic rocks associated with the paragneiss also show progressive metamorphism from amphibole-biotite-rich rocks of the amphibolite facies at Emeryville to garnet-plagioclase-potassium feldspar-pyroxene-rich rocks of the lower granulite facies at Colton.

Our specimens are from collections (Ross, Smith, and Ashton, 1968) made at the Arnold pit (Fowler, New York, samples G-16, G-21) and Wight talc mine (Fowler, New York, sample G-24) and from the International Talc Company collections (Talcville, New York, sample G-FT-1) and collections obtained by Segeler (1961, U.S.N.M. Nos. 115046 and 115545) from the International Talc mines, Talcville, New York. All of the locations are situated within about three miles of Emeryville and so a maximum temperature of metamorphism of 525°C is suggested.

Tremolite from the Arnold pit, Wight mine, and the In-

cummingtonite. X-ray photographs (Fig. 2B) of crystals similar to that shown in Figure 1B indicate that the manganian cummingtonite host contains about 10 percent tremolite. Although the unit-cell dimensions of the host and lamellar manganian cummingtonites are similar to those expected for Mg-cummingtonites, the space group symmetry is $P2_1/m$, and not $C2/m$ as usually expected for cummingtonite. Bown (1965) was the first to recognize this $P2_1/m$ variety of clinoamphibole as lamellae in tremolite from the Wight mine. He suggested it be called "clinoanthophyllite," in allusion to an expected high magnesium content.

In the precession photographs of the $h0l$ net of tremolite (G-24) and manganian cummingtonite (115545), Figures 2A and 2B, respectively, there appear two sets of spots representing the lattices of the host and lamellar phases. The stronger set of reflections in each photograph is due to the host; the weaker set is due to the lamellae. A comparison of Figure 2A with Figure 2B illustrates the reciprocal relationship of the pattern intensities; the strong set of reflections in one figure appears as the weak set in the other. A rough estimate of the relative amounts of host and lamellar phases (Table 2) is obtained by evaluating the relative intensities of the two diffraction patterns registered in the $h0l$ X-ray photograph. Most amphibole crystals from the Gouverneur localities contain an estimated 5 to 15 percent of second amphibole phase. Samples 115046 (primitive manganian cummingtonite host) and G-21 (tremolite host), however, contain very small amounts of lamellar phase; and some crystals show none—at least none that we can resolve optically or with X-ray techniques. Unit-cell parameters, space group symmetry, and approximate amounts of host and included phase for each crystal examined are given in Table 2.¹

¹ The accuracy of the unit-cell parameters varies depending on the character of the pattern but is generally within ± 0.2 to ± 0.5 percent of the values given in Table 2 and also in Tables 4, 6, and 8. The estimates of relative amounts of host and lamellar phase in each grain given in these tables are only semi-quantitative.

Figure 2B also clearly demonstrates the violation of $C2/m$ symmetry expected for cummingtonite. The strongest reflections showing this are 502, 304, $30\bar{4}$, $10\bar{4}$, and 704. These same reflections appear in Figure 2A but are much weaker because very little lamellar primitive cummingtonite is present. We here classify cummingtonites which have $P2_1/m$ space group symmetry as "primitive" cummingtonites in allusion to the primitive lattice. Cummingtonites with $C2/m$ space group symmetry are designated "C-centered" cummingtonites. Further reasons for these designations are given below.

Chemical composition. Chemical analyses and partial analyses for some of the amphibole specimens described here have been given by Ross, Smith, and Ashton (1968) and Ross, Papike, and Weiblen (1968). An electron-microprobe scan for manganese and calcium along the prism axis of a grain of tremolite (sample G-24) similar to the one shown in Figure 1A reveals that the lamellae are high in Mn and low in Ca relative to the host tremolite. When the tremolite grains from specimen G-24 were excited by the unfocused electron beam of the electron probe, the tremolite host luminesced golden yellow and the included lamellae luminesced dark red. A few of the host crystals of this sample luminesced dark red and the included ($\bar{1}01$) lamellae luminesced gold yellow. Probe scans across such grains gave patterns reciprocal to those given by the host tremolite grains, in that the lamellae are rich in Ca and the host is rich in Mn. Partial quantitative analyses of this sample by electron microprobe (Ross, Papike, and Weiblen, 1968) indicate the composition $\text{Ca}_{1.49}\text{Mn}_{0.29}\text{Fe}_{0.04}\text{Mg}_{5.18}\text{Si}_8\text{O}_{22}(\text{OH},\text{F})_2$ for the yellow-luminescing host tremolite, and $\text{Ca}_{0.24}\text{Mn}_{0.88}\text{Fe}_{0.08}\text{Mg}_{5.81}\text{Si}_8\text{O}_{22}(\text{OH},\text{F})_2$ for the red-luminescing host manganian cummingtonite. The narrowness of the included lamellae prevents quantitative estimation of their chemical composition, but it is probable that the compositions of the lamellae are similar to those of the host grains having similar luminescence color.

It is clear that the schist from the area of the Wight mine contains two primary clinoamphibole phases: tremo-

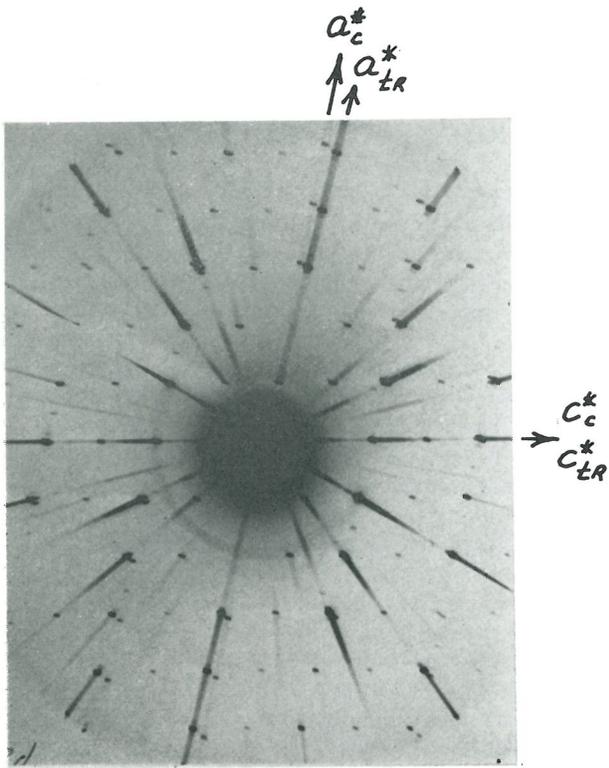
←

FIG. 1. Exsolution in amphiboles from Gouverneur, New York. A. Large tremolite crystal containing thin (approximately one μ wide) lamellae of primitive manganian cummingtonite oriented parallel to ($\bar{1}01$) of the host tremolite. Thin section, crossed polarizers, sample G-24, Arnold pit.

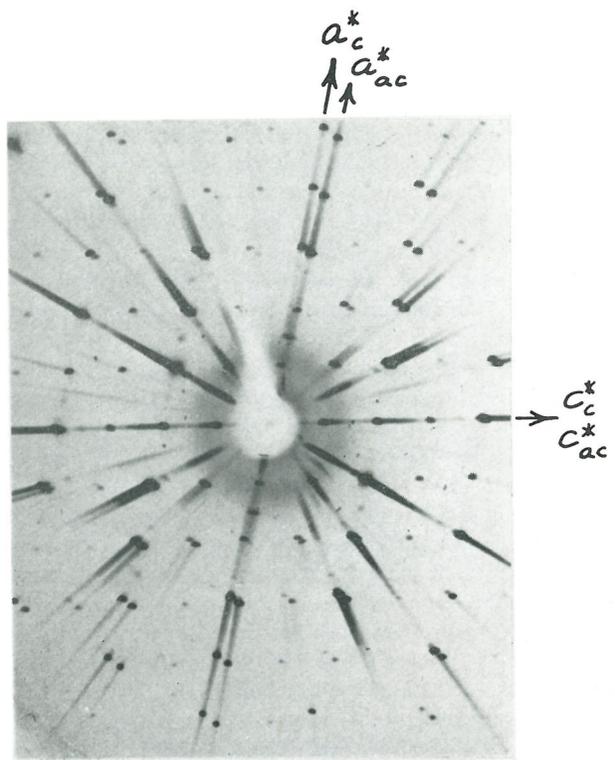
B. Large primitive manganian cummingtonite grain containing tremolite lamellae approximately 0.4 μ thick oriented parallel to ($\bar{1}01$) of the host cummingtonite. Oil mount, crossed polarizers, sample G-FT-1, International No. 3 mine, Talcville.

C. Luminescence photomicrograph taken in the electron probe of primitive manganian cummingtonite grains. The host areas, appearing as shades of gray, luminesce grayish-yellow; the included tremolite lamellae, appearing as white bands, luminesce golden-yellow. Bordering the tremolite lamellae are zones which luminesce dark-red (very dark areas). Polished section, sample 115545, Talcville.

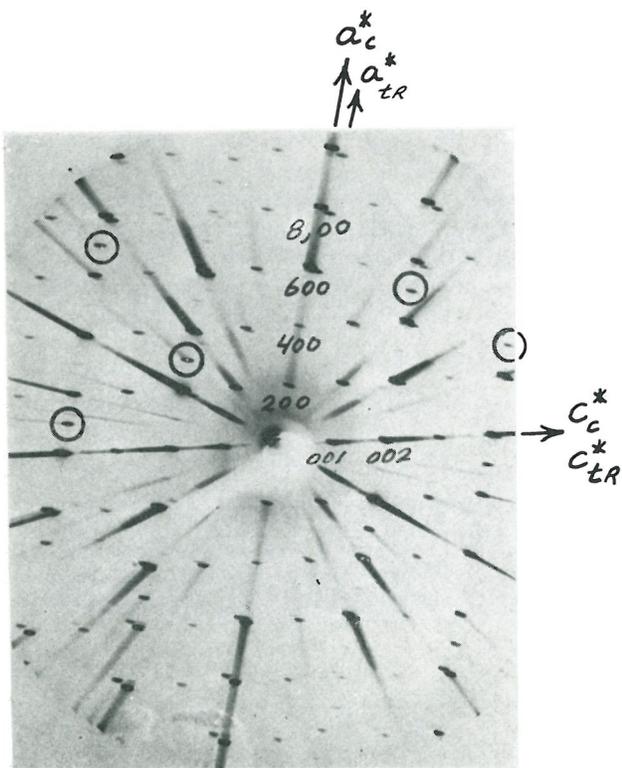
D. Large tremolite grain which has been heated to 750°C at 10 kbar for 6 days. The ($\bar{1}01$) lamellae of primitive manganian cummingtonite no longer appear as continuous striae but as beads which line up along the trace of the old ($\bar{1}01$) lamellae. Oil mount, crossed polarizers, sample G-24, Arnold pit.



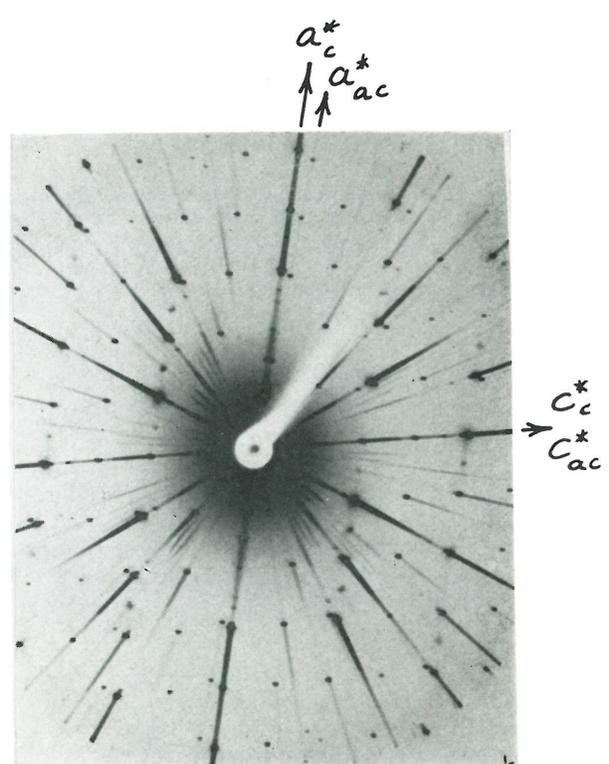
A



B



C



D

lite and manganian cummingtonite. As the schists cooled from a temperature of approximately 525°C, the two host phases exsolved primitive manganian cummingtonite and tremolite. Host manganian cummingtonite from sample G-24 was not examined by X-ray techniques for it could only be identified by its luminescent properties in the electron probe. Our experience with other samples strongly suggests, however, that this cummingtonite possesses $P2_1/m$ symmetry and has unit-cell dimensions similar to those of the manganian cummingtonite lamellae that are included within the associated host tremolite.

Tremolite in sample G-21 is much more nearly homogeneous than that in the G-24 specimen; only a few dark-red luminescing cummingtonite lamellae were seen during examination with the electron probe. The X-ray photographs of some crystals (Table 2) showed a trace of included cummingtonite; others showed none. The crystal structure, determined on a tremolite crystal (sample G-21) that contains no included phase, is reported by Papike, Ross, and Clark (1969, this volume). Wet chemical analysis (Table 3) of 5 grams of carefully picked tremolite gives the composition $(\text{Na,K})_{0.12}\text{Ca}_{1.86}\text{Mg}_{4.97}(\text{Mn,Fe}^{2+},\text{Ti,Al})_{0.16}\text{Si}_{7.95}\text{O}_{22}(\text{OH,F})_2$. This is very close to the theoretical tremolite end-member composition $\text{Ca}_2\text{Mg}_5\text{Si}_8\text{O}_{22}(\text{OH})_2$. Very little exsolution is, therefore, expected. Two other excellent wet chemical analyses of tremolites (samples 9 and 15) collected by Engel (1962) from the old Arnold mine (now the Arnold pit) are also reported in Table 3. Sample 9 is deficient in calcium (1.68 atoms per formula unit) whereas sample 15 is close to end-member composition.

Segeler (1961) has described a very manganese-rich cummingtonite-like clinoamphibole collected from the International Talc mines, samples of which are now at the U. S. National Museum (Nos. 115046, 115545). Similar material was also obtained in 1967 from the International Talc Company collections (G-FT-1). Optical, X-ray, and

electron-probe examination of these clinoamphiboles show that they are very similar to the primitive cummingtonites found in sample G-24. The luminescent properties are particularly revealing as shown in the electron-probe luminescence photomicrograph (Figure 1C) of a polished section of sample 115545. The host cummingtonite (shades of gray in the photograph) luminesces a grayish-yellow; the included tremolite lamellae (white bands in the photograph) luminesce golden-yellow. Bordering the tremolite lamellae are zones that luminesce dark-red. Ross, Papike, and Weiblen (1968) give evidence that the dark-red luminescing areas are slightly enriched in Mn and depleted in Ca relative to the grayish-yellow luminescing host areas, which have the composition $\text{Ca}_{0.38}\text{Mn}_{1.09}\text{Fe}_{0.08}\text{Mg}_{5.45}\text{Si}_8\text{O}_{22}(\text{OH,F})_2$. The apparent chemical differences among the three areas (grayish-yellow cummingtonite host, golden-yellow tremolite lamellae, and dark-red border zones) are interpreted as being due to a diffusion front, calcium ions moving toward, and manganese away from the tremolite lamellae. Appreciable unmixing probably has occurred only within the border zones and tremolite lamellae. It appears that the composition given above for the grayish-yellow luminescing host is essentially that of the original unmixed primitive cummingtonite.

The amphibole crystals in sample 115046 are much more homogeneous; only a few lamellae of tremolite appear in the dark-red luminescing primitive cummingtonite. X-ray examination (Table 2) shows that some crystals (Nos. 4, 6A, 6B) from this sample consist entirely of cummingtonite. Other crystals (Nos. 1, 2, 3, 5) contain about 5 percent included tremolite lamellae oriented parallel to (101) of the host. A complete chemical analysis accomplished with the electron probe (Table 3) gives the following composition for the red luminescing host areas: $\text{Na}_{0.06}\text{Ca}_{0.36}\text{Mn}_{0.96}\text{Fe}_{0.01}\text{Mg}_{5.57}\text{Al}_{0.01}\text{Si}_{8.02}\text{O}_{22}(\text{OH,F})_2$. The crystal structure of this primitive cummingtonite (crystal 6A) is reported by Papike, Ross, and Clark (1969, this volume).

←

FIG. 2. Buerger precession X-ray photographs of amphibole crystals. A. The $h0l$ net of a tremolite grain, sample G-24, crystal 1. The strong set of reflections marked by axes a^*_{tr} and c^*_{tr} are due to the tremolite host. The weaker set of reflections marked by axes a^*_c and c^*_c are due to the primitive manganian cummingtonite lamellae. Mo $K\alpha$ radiation, 70 hr exposure.

B. The $h0l$ net of a primitive manganian cummingtonite grain, sample 115545, crystal 1. The strong set of reflections marked by axes a^*_c and c^*_c are due to the cummingtonite host. Some of the stronger reflections violating the $C2/m$ space-group symmetry are circled (i.e., 502, 304, 302, 104, and 704). The weaker set of reflections marked by axes a^*_{tr} and c^*_{tr} are due to the tremolite lamellae. Mo $K\alpha$ radiation, 60 hr exposure.

C. The $h0l$ net of an actinolite grain, sample HJ-163-60, crystal 1. A photomicrograph of this crystal is shown in Fig. 3A. Two sets of reflections of nearly equal intensity are apparent. One set of reflections, marked by the axes a^*_{ac} and c^*_{ac} , is due to actinolite; the other set, marked by the axes a^*_c and c^*_c , is due to the primitive cummingtonite. The reflections violating $C2/m$ symmetry are weak and slightly diffuse (e.g., 502, 302, 502). All cummingtonite lamellae are oriented parallel to (101) of actinolite. The very sharp spots out of register with the amphibole patterns are due to magnetite. Mo $K\alpha$ radiation, 72 hr exposure.

D. The $h0l$ net of a primitive cummingtonite host grain, sample HJ-182-60, crystal 2B. Two sets of reflections of unequal intensity appear. The strong set, marked by the axes a^*_c and c^*_c is due to cummingtonite; the much weaker set, marked by the axes a^*_{ac} and c^*_{ac} , is due to actinolite. All actinolite is oriented parallel to (101) of the host. The reflections violating the $C2/m$ symmetry are fairly strong (e.g., 102, 302, 104). Some are diffuse, especially the 502 and 704 which have the shape of an "X". Mo $K\alpha$ radiation, 80 hr exposure.

TABLE 3. CHEMICAL ANALYSES OF TREMOLITES AND PRIMITIVE MANGANOAN CUMMINGTONITE FROM THE GOUVERNEUR, N. Y. AREA

	Wt %				Atomic ratios ^e					
	9 ^a	15 ^b	G-21 ^c	115046 ^d	9	15	G-21	115046		
SiO ₂	57.83	57.76	58.90	58.31	Si 7.915	7.897	8.001	7.947	Si 8.023	
Al ₂ O ₃	0.57	0.64	0.56	0.06	Al 0.092	8.028	0.104	0.089		
Fe ₂ O ₃	0.20	0.00	—	—	Fe ³⁺ 0.021					
MgO	24.25	24.73	24.74	27.17	Mg 4.945	5.038	4.974		Mg 4.990	
FeO	0.11	0.14	0.22	0.13	Fe ²⁺ 0.012	5.285	5.066	0.025	Al 0.010	
TiO ₂	0.03	0.05	0.02	—	Ti 0.003	0.005	0.002			
MnO	2.81	0.06	0.42	8.24	Mn ²⁺ 0.325	0.007	0.048		Mg 0.582	
CaO	11.42	13.27	13.00	2.46	Ca 1.675	1.944	1.861		Ca 0.363	
K ₂ O	0.14	0.09	0.10	—	K 0.025	1.744	2.033	0.017	Mn 0.961	
Na ₂ O	0.17	0.28	0.40	0.22	Na 0.044	0.073	0.105		Na 0.058	
H ₂ O ⁻	0.06	0.13	—	—					Fe 0.015	
H ₂ O ⁺	2.09	2.38	1.19	—	OH 1.907	1.820	2.170	1.882	1.071	1.872
F	0.41	0.26	0.30	—	F 0.178	0.180	0.113	0.118	0.128	0.128
Cl	0.01	0.02	—	—	Cl 0.002		0.005			
CO ₂	0.00	—	0.10	—						
Σ	100.10	99.81	99.95	96.59						
O=F+Cl	0.17	0.11	0.13	—						
Σ	99.93	99.70	99.82	—						

^a Tremolite, Arnold No. 3 mine, 11th level, Fowler, N. Y. (Engel, 1962, p. 309).

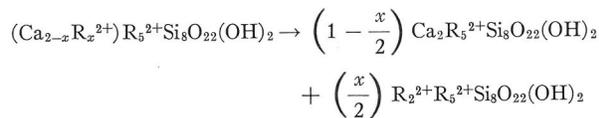
^b Tremolite, Arnold mine, 14th level, Fowler, N. Y. (Engel, 1962, p. 309).

^c Tremolite, Arnold pit, Fowler, N. Y. (Ross, Smith, and Ashton, 1968), J. J. Fahey analyst.

^d Primitive manganoan cummingtonite, International Talc mine, No. 3, Talcville, N. Y. (U.S.N.M. 115046). Electron microprobe analysis, A. T. Anderson, Jr., Analyst.

^e Number of ions on the basis of O=22 and (OH)=2.00-F-Cl.

Exsolution in the Gouverneur clin amphiboles. It is clear that the large $M(4)$ cation positions, which ideally contain two calcium atoms per tremolite formula unit, can, at least at high temperatures, accommodate significant amounts of smaller divalent cations such as Mn^{2+} , Fe^{2+} , and Mg . As the rocks cool, the small cations in $M(4)$ find themselves in an unstable atomic configuration and a phase rich in these ions unmixes from the tremolite host. An idealized reaction (assuming complete unmixing with respect to calcium) is written:



where $R^{2+} = Mg + Fe + Mn$. Conversely, cummingtonites, either primitive or C -centered, may have $M(4)$ sites (ideally containing $2R^{2+}$) which are somewhat enriched in calcium. Such amphiboles on cooling may exsolve tremolite. The exsolution relationships proposed for these clin amphiboles is quite analogous to the augite-orthopyroxene-pigeonite relationships.

It should also be kept in mind, particularly because of its very important bearing on amphibole phase relations, as discussed in the last section of this paper, that to maintain *inter-* and *intracrystalline* equilibrium, the amphibole crystals during cooling must not only unmix a second amphibole

phase but also must undergo homogeneous exchange between the various crystallographic sites. In amphiboles the most important exchange energetically will take place between cations in the large $M(4)$ site and the small $M(1)$, $M(2)$, and $M(3)$ sites. Because of their large size, calcium and sodium must remain in the $M(4)$ site or unmix as calcic amphibole. Cations such as Mn^{2+} , Fe^{2+} , and Mg , however, are free to exchange among all the M sites. For crystal-chemical reasons we believe that as temperature drops the larger of these cations (Mn^{2+} and Fe^{2+}) will concentrate in the large $M(4)$ site if it is not filled with calcium and sodium; the smaller cations such as Mg will concentrate in the small $M(1)$, (2), and (3) sites. We believe that the structural state of the cummingtonites depends directly on the occupancy of the $M(4)$ site. Primitive cummingtonite will contain significant amounts of magnesium in $M(4)$ and C -centered cummingtonite will contain little or no magnesium in $M(4)$.

The small amounts of sodium and aluminum usually found in Gouverneur tremolites are accounted for by one or more of the following coupled substitution mechanisms:

- (1) $Na_x(Ca_{2-x}Na_x)R_5^{2+}Si_8O_{22}(OH)_2$
- (2) $(Ca_{2-x}Na_x)(R_{5-x}^{2+}Al_x)Si_8O_{22}(OH)_2$
- (3) $Na_xCa_2R_5^{2+}(Si_{8-x}Al_x)O_{22}(OH)_2$
- (4) $Ca_2(R_{5-x}^{2+}Al_x)(Si_{8-x}Al_x)O_{22}(OH)_2$

where, in mechanisms (1) and (3), x sodium atoms enter the normally vacant A site. We have no evidence that trem-

olites of any of the above types unmix a phase rich in sodium and/or aluminum.

The amount of unmixing that occurs in the Gouverneur amphiboles depends not only on the initial composition of the host phase but also on diffusion rates of the participating ions, which are controlled particularly by temperature. The cooling history of the rocks, including the presence or absence of subsequent metamorphic events, has a great effect on changes in diffusion rates. Chemical compositions of the host phases given above show that there is still considerable enrichment of calcium in the $M(4)$ site of primitive manganoan cummingtonite (samples G-24, 115545, and 115046), and a considerable deficiency of calcium in the $M(4)$ site of the G-24 tremolite.

Several heating experiments were attempted by M. Charles Gilbert (Geophysical Laboratory) and J. S. Huebner (U. S. Geol. Survey) in order to see if the amphibole intergrowths could be homogenized. Tremolite G-24 was subjected to three heating experiments (750°C, 10 kbar, 6 days; 750°C, 2 kbar, 10 days; 805°C, 1 kbar, 66 days); and a primitive manganoan cummingtonite, no. 115545, was used in one experiment (805°C, 1 kbar, 66 days). In all runs the amphibole grains were in contact with a hydrous vapor phase. X-ray precession photographs showed no significant changes in unit-cell parameters or intensity of the X-ray reflections of the host and lamellar amphibole phases in the heated samples (Table 2). Also, no change in the intensity or character of the reflections violating $C2/m$ space group symmetry were noted in the X-ray photographs of the primitive cummingtonite host (115545) or the primitive cummingtonite lamellae (G-24). There is, however, optical evidence for a partial recrystallization of the included lamellae in the tremolite heated to 750°C at 10 kbar for 6 days. Lamellae which appeared originally as straight continuous striations (Fig. 1A) now appear as discontinuous traces (Fig. 1D). The lamellae apparently have separated into beads which approximately line up along the old $(\bar{1}01)$ trace.

Cummingtonite nomenclature. When we first found that the $(\bar{1}01)$ lamellae in the G-24 tremolite, a sample which must be very similar to the one studied by Bown (1965), were rich in manganese, we thought that the presence of this element was partly or wholly responsible for the unusual symmetry. However, we later found that some cummingtonites with $P2_1/m$ symmetry contain little or no manganese. Also, we find that cummingtonites from the Wabush Iron Formation, Labrador (Klein, 1964), which contain a large amount of manganese, possess $C2/m$ space group symmetry (Table 8).

Our present data on chemical compositions of primitive cummingtonites from several localities indicate that the range of composition overlaps that of both anthophyllite and C -centered cummingtonite. The crystal structure of the primitive manganoan cummingtonite from Talcville (115046) bears similarities to both anthophyllite and C -

centered cummingtonite (Papike, Ross, and Clark, 1969, this volume). The configuration of the double chains of tetrahedra above and below the octahedral strip containing the $M(1)$, $M(2)$, and $M(3)$ cations resembles that found in anthophyllite (Finger, 1967). However, the manner in which the tetrahedral-octahedral strips are linked together three-dimensionally is very similar to that found in C -centered cummingtonite and gives a monoclinic lattice metrically close to that of $C2/m$ cummingtonite.

We believe that primitive cummingtonite is more closely related to C -centered cummingtonite than to anthophyllite; thus, we prefer not to use the term "clinoanthophyllite" as suggested by Bown (1965). Also, at present, we cannot distinguish some primitive cummingtonites from the C -centered variety by optical or chemical means. Therefore, if the space group has not been determined, the name cummingtonite should be used without the prefix " C -centered" or "primitive." We do not believe a new name should be given to the $P2_1/m$ phase.

Segeler (1961) referred to clinoamphiboles from Talcville (samples 115046, 115545) as "tirodite" in allusion to the manganese-rich clinoamphiboles from Tirodi, India. Mr. G. Rowbotham (Dep. Geol., Univ. Durham, writt. comm., Oct. 3, 1968) finds that the type tirodite is very sodic with 1.06 atoms per formula unit. The unit-cell parameters are: $a = 9.799$, $b = 17.994$, $c = 5.289\text{\AA}$, $\beta = 103.89^\circ$. The high sodium content, large a -dimension, and β angle clearly demonstrate that type tirodite is related to tremolite and richterite, and does not resemble the Talcville primitive cummingtonite. The name tirodite should not be applied to the Talcville specimens.

CLINOAMPHIBOLES FROM THE RUBY MOUNTAINS, MONTANA

General geology and sample descriptions. The Carter Creek iron deposit, located at the southwest end of the Ruby Mountain Range in Beaverhead and Madison Counties near Dillon, Montana, occurs in a Precambrian terrane of highly deformed metasedimentary rocks (Heinrich, 1960). Finely banded to gneissic iron formation units, 5 to 40 feet thick, are interbedded with diopsidic dolomite, amphibolite, sillimanite-potassium feldspar-garnet schist, and quartzite (James and Wier, 1961). Detailed mapping of the Precambrian rocks in this part of the Ruby Mountains by the U. S. Geological Survey is now in progress. The rocks are pre-Belt in age, and radiometric age determinations by Gilletti (1966) indicate that a major metamorphic event produced mineral assemblages typical of the upper amphibolite facies about 1.6 billion years ago. Temperatures of metamorphism of about 620°C are inferred from O^{18}/O^{16} in several quartz-magnetite mineral pairs (E. C. Perry, Jr., oral comm.). Retrograde effects, however are widespread.

The most common mineral assemblage in the iron formation is quartz-magnetite-amphibole. In more than 40 percent of the 30 thin sections studied, the amphibole is a blue riebeckite(?); in about 25 percent, actinolite coexists

with cummingtonite; and in about 15 percent, the amphibole is cummingtonite. Minor constituents include apatite, microcline, plagioclase, epidote, biotite, and chlorite.

The four samples used in the present study are from diamond drill cores. Modes of three of these samples, based on a minimum of 1100 points per thin section, one thin section per sample, are as follows:

- HJ-163-60 quartz 44%, magnetite 34%, amphibole 18%, apatite 2%, biotite and chlorite 1%. (DDH-9A-396 feet)
 HJ-182-60 quartz 46%, magnetite 31%, amphibole 20%, apatite 2%, biotite and chlorite 1%. (DDH-9A-395-405 feet)
 HJ-167-60 quartz 52%, magnetite 26%, amphibole 20%, apatite 2%, chlorite trace. (DDH 11-383 feet)

Amphibole as counted includes both actinolite and cummingtonite. No mode of sample HJ-162-60 (DDH 9A-383 feet) was made because the thin section includes portions of a biotite schist, but the major minerals are quartz, actinolite, cummingtonite, and magnetite.

Optical and X-ray studies. Two actinolite grains (sample HJ-163-60) which have been examined by single-crystal X-ray techniques, are shown in Figures 3A (crystal 1) and 3B (crystal J-2). Crystal 1 contains fourteen thick primitive cummingtonite lamellae and numerous thin lamellae, oriented parallel to $(\bar{1}01)$ of the actinolite. Crystal J-2 contains two very thick $(\bar{1}01)$ primitive cummingtonite lamellae as well as numerous thin primitive cummingtonite lamellae oriented parallel to (100) of the actinolite. These optical observations are confirmed by the X-ray photographs (Fig. 2C), which show that crystal 1 contains roughly 44 percent $(\bar{1}01)$ primitive cummingtonite and crystal J-2 contains about 30 percent $(\bar{1}01)$ and about 15 percent (100) primitive cummingtonite. Six other actinolite grains from this sample also were found to consist of actinolite and cummingtonite in approximately the same ratio, 55:45 (Table 4). The relative amounts of (100) and $(\bar{1}01)$ lamellae vary a great deal; for example, crystal J-1 contains only (100) lamellae, and crystal 1 only $(\bar{1}01)$ la-

mellae. Two crystals (nos. 3 and 4, Table 4) contain only lamellae of cummingtonite with $C2/m$ space group symmetry. $P2_1/m$ reflections are present in the X-ray patterns of the other six actinolite grains, but are very weak and in some patterns are slightly diffuse.

X-ray photographs of six host actinolite grains from sample HJ 182-60, very similar to that shown in Figure 2C, indicate (by measurement of relative intensities) that all contain about 45 percent actinolite¹ and 55 percent primitive cummingtonite (Table 4). The cummingtonite host grains of this sample contain 5 to 10 percent actinolite lamellae. The single-crystal pattern of a host primitive cummingtonite grain (sample HJ 182-60) is shown in Figure 2D. Two of the cummingtonite grains (crystals 2B, 3) possess $P2_1/m$ symmetry, and one (crystal 1B) possesses $C2/m$ symmetry. The relative amounts of host and lamellar phase in the actinolite and cummingtonite grains can be seen in Figure 3D, a composite photomicrograph showing the cummingtonite host on the right and the actinolite host on the left. It is apparent that the colorless cummingtonite lamellae in the actinolite make up more than 50 percent of the grain. The lower third of this grain is nearly all cummingtonite indicating an intragranular coalescence of the two amphibole phases.

Samples HJ-167-60 and HJ-162-60 differ from the two samples discussed above in that the actinolite grains contain only about 20 to 30 percent cummingtonite lamellae. The cummingtonite grains from these samples have not been examined by X-ray techniques, but optically they appear similar to those of HJ-180-60 and HJ-163-60 and probably contain 5 to 10 percent actinolite lamellae.

Opaque rod-like inclusions are present in many of the actinolite grains; for example, as shown in Figure 3A (crystal 1) and Figure 3C. The single crystal X-ray photo-

¹ Even though it appears that these grains contain more than 50 percent cummingtonite component the term "actinolite host" will be retained for they coexist with cummingtonite host crystals containing 10 percent actinolite component.

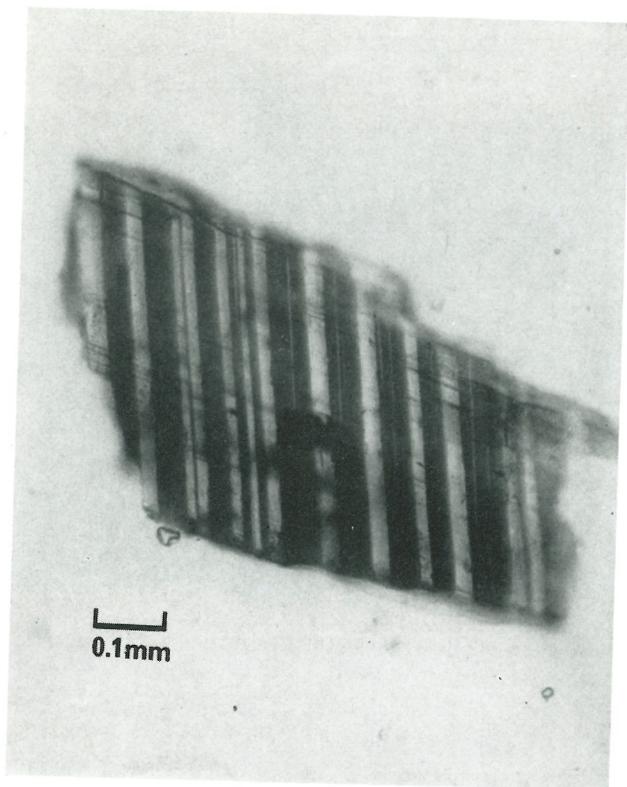
»»»→

FIG. 3 Exsolution in actinolite from Ruby Mountains, Montana. A. Pleochroic green actinolite (dark lamellae) makes up approximately 55% of the grain. The remainder of the grain is composed of colorless primitive cummingtonite lamellae oriented parallel to $(\bar{1}01)$ of actinolite. An X-ray photo of this grain is presented in Fig. 2C. Oil mount, uncrossed polarizers, sample HJ-163-60, crystal 1.

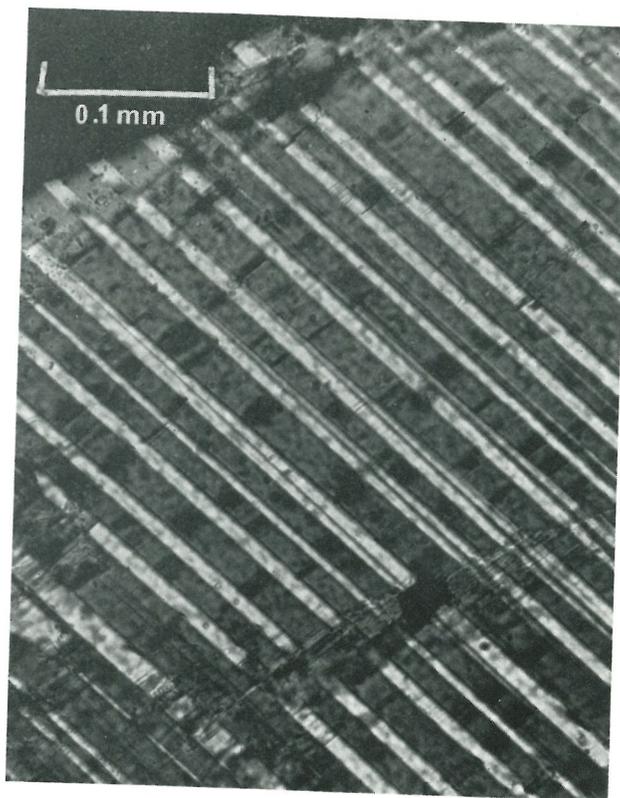
B. Actinolite grain (dark area) containing two large colorless primitive cummingtonite lamellae oriented parallel to $(\bar{1}01)$ of the actinolite and trending 12-6 o'clock. Thinner primitive cummingtonite lamellae are oriented parallel to (100) of the actinolite and trend 2-8 o'clock. Oil mount, uncrossed polarizers, sample HJ-163-60, crystal J-2.

C. Photomicrograph of a portion of an actinolite grain (dark lamellae) with primitive cummingtonite lamellae (light areas) oriented parallel to $(\bar{1}01)$ of the actinolite. The very dark rod-like inclusions (oriented 2-8 o'clock) in the actinolite lamellae are magnetite grains. Thin section, uncrossed polarizers, sample HJ-163-60.

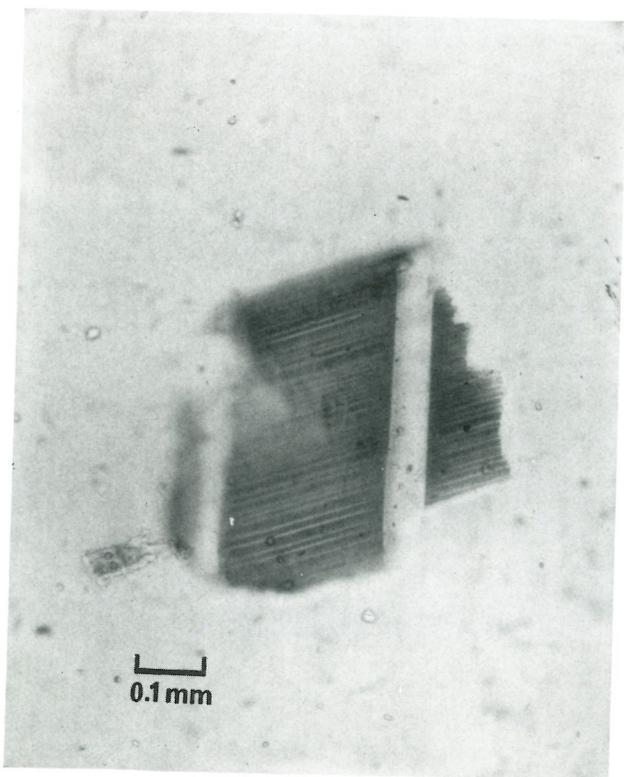
D. Composite photomicrograph showing actinolite host and cummingtonite host that coexist in the same sample. The grain on the left is actinolite (dark areas) containing about 55% primitive cummingtonite lamellae oriented parallel to $(\bar{1}01)$. The grain on the right is primitive cummingtonite containing about 10% actinolite lamellae (darker areas) oriented parallel to $(\bar{1}01)$ of the host. Oil mount, uncrossed polarizers, sample HJ-182-60.



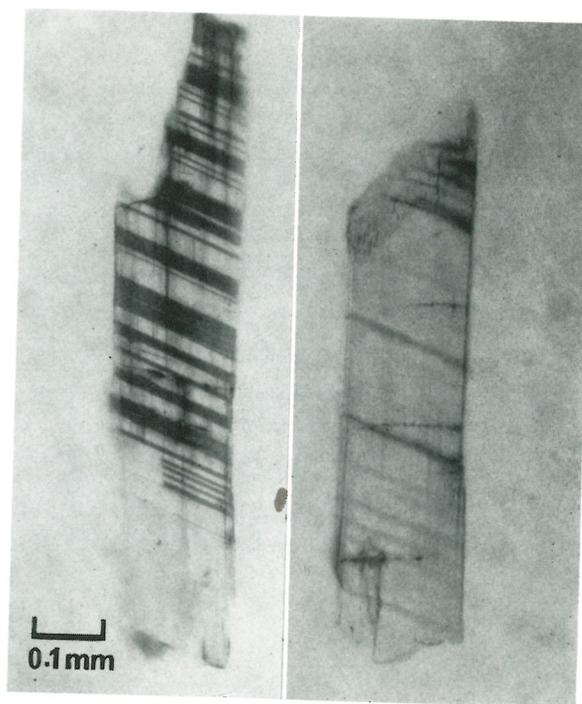
A



B



C



D

TABLE 4. CRYSTALLOGRAPHIC DATA FOR AMPHIBOLES FROM THE RUBY MOUNTAINS, SOUTHWEST MONTANA

Sample No.	Crystal No.	Amphibole Intergrowths	% of grain	a(Å)	b(Å)	c(Å)	β	Space Group	V(Å ³)
HJ 182-60, core 9A, 395-405 feet	1A	actinolite host	45	9.82	18.10	5.29	104°35'	C2/m	910.0
		P-cummingtonite on (101)	52	9.48	18.10	5.30	102°10'	P2 ₁ /m	889.0
		P-cummingtonite on (100)	3					P2 ₁ /m	
	2A	actinolite host	45					C2/m	
		P-cummingtonite on (101)	55					P2 ₁ /m	
	4	actinolite host	45					C2/m	
		P-cummingtonite on (100)	53					P2 ₁ /m	
		P-cummingtonite on (101)	2					P2 ₁ /m	
5, 6, 7	actinolite host	45					C2/m		
	P-cummingtonite on (100)	55					P2 ₁ /m		
2B	P-cummingtonite host	90	9.50	18.14	5.32	102°10'	P2 ₁ /m	896.2	
	actinolite on (101)	10	9.83	18.14	5.30	104°35'	C2/m	914.6	
3	P-cummingtonite host	95	9.51	18.17	5.32	102°5'	P2 ₁ /m	898.9	
	actinolite on (101)	5	9.84	18.17	5.29	104°30'	C2/m	915.7	
1B	C-cummingtonite host ^b	95	9.50	18.15	5.30	102°10'	C2/m	893.3	
	actinolite on (101)	5	9.82	18.10	5.29	104°35'	C2/m	910.0	
HJ 182-60 ^a powdered mixture		actinolite		9.839(3)	18.114(9)	5.285(6)	104°33'(3)		911.8(8)
		cummingtonite		9.509(16)	18.116(5)	5.323(2)	102°18'(2)		896.0(3)
HJ 162-60 core 9A, 383 feet	1	actinolite host	70	9.81	18.07	5.30	105°0'	C2/m	907.5
		C-cummingtonite on (100)	20	9.50	18.07	5.30	102°15'	C2/m	889.1
		C-cummingtonite on (101)	10					C2/m	
HJ 163-60, core 9A, 396 feet	J-1	actinolite host	55	9.84	18.11	5.28	104°28'	C2/m	906.8
		P-cummingtonite on (100)	45	9.49	18.11	5.30	102°0'	P2 ₁ /m	891.0
	J-2	actinolite host	55	9.82	18.05	5.29	104°35'	C2/m	907.4
		P-cummingtonite on (101)	30	9.51	18.05	5.32	102°5'	P2 ₁ /m	893.3
		P-cummingtonite on (100)	15					P2 ₁ /m	
	1	actinolite host	55	9.81	18.07	5.29	104°25'	C2/m	908.2
		P-cummingtonite on (101)	44	9.48	18.07	5.30	102°5'	P2 ₁ /m	887.9
		magnetite	1	8.39	—	—	—	Fd3m	590.6
	3, 4	actinolite host	55					C2/m	
		C-cummingtonite on (100)	45					C2/m	
5, 6, 7	actinolite host	55					C2/m		
	P-cummingtonite on (100)	43					P2 ₁ /m		
	cummingtonite on (101)	2					P2 ₁ /m?		
2B	P-cummingtonite host	95	9.50	18.12	5.31	102°0'	P2 ₁ /m	894.1	
	actinolite on (101)	5	9.82	18.12	5.28	104°40'	C2/m	908.9	
HJ 163-60 ^a X-ray powder data from individual host grains (unheated)		actinolite		9.836(2)	18.086(6)	5.288(2)	104°39'(2)		910.0(4)
		cummingtonite		9.489(3)	18.137(3)	5.276(4)	101°39'(2)		889.4(6)
HJ 163-3F X-ray powder data of samples heated to 800°C, 2kbar 4 weeks		actinolite		9.794(6)	18.112(11)	5.276(5)	104°29'(4)		906.1(8)
		cummingtonite + magnetite + quartz		9.509(1)	18.090(2)	5.251(1)	102°1'(1)		883.5(2)
HJ 167-60, core 11, 385 feet	1	actinolite host	80	9.80	18.04	5.30	104°30'	C2/m	907.2
		P-cummingtonite on (101)	20	9.50	18.04	5.31	102°0'	P2 ₁ /m	890.1

^a Unit-cell parameters obtained by least-squares refinement of X-ray diffraction powder data.

^b C-cummingtonite refers to "C-centered" cummingtonite.

graph of the $h0l$ net of crystal 1 (Fig. 2C) shows, in addition to the patterns of actinolite and cummingtonite, a very weak "single-crystal" diffraction pattern of magnetite with $a = 8.39 \pm 0.02 \text{ \AA}$. It is assumed that the rod-like inclusions seen in the photomicrographs are magnetite and the apparent single-crystal pattern of this phase arises because the (211) planes and [111] zone axes of the opaque inclusions are all parallel to (010) and perpendicular to (203)* respectively of the actinolite. Similar spinel inclusions have been observed within pigeonite and orthopyroxene lamellae exsolved from augite (M. Ross, unpublished data). It is probable that unmixing in amphiboles and pyroxenes in some cases is accompanied by an oxidation reaction in which some ferrous iron of the chain silicate is oxidized to form the ferric iron component of magnetite.

Chemical composition. We noted that thick cummingtonite lamellae typically are free of actinolite lamellae. Actinolite lamellae, on the other hand, usually contain numerous cummingtonite intergrowths (Figs. 3A, 3B, 4A). In some of the actinolite grains, however, especially those from sample HJ-182-60, areas of homogeneous appearing actinolite were found. In order to obtain quantitative chemical information, Paul W. Weiblen (Dep. Geol., Univ. Minn.) kindly undertook an electron-microprobe study of sample HJ-182-60.

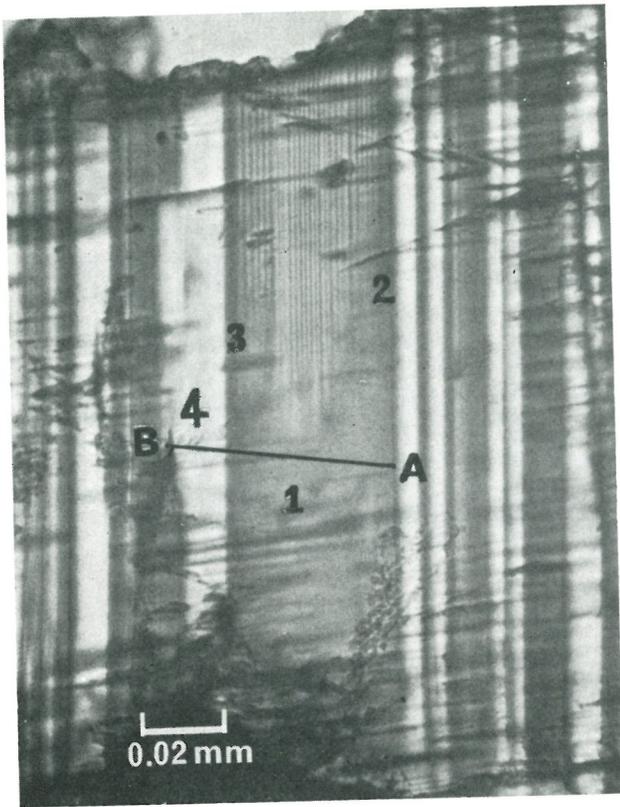
Uncovered thin sections polished on both sides were prepared for microprobe analysis. One of the actinolite grains analyzed is shown in Figure 4A, which is a photomicrograph taken in the electron probe. The thick cummingtonite lamellae appear to be optically homogeneous; the actinolite lamellae appear quite inhomogeneous, particularly the one approximately 40 microns thick in the center of the photomicrograph. In the upper part of this thick actinolite lamella can be seen numerous lamellae of cummingtonite approximately one micron thick. Electron-probe scans for calcium, iron, and magnesium along the trace *A-B* (Fig. 4A) are shown in Figure 5. The scan starts at *A*, within a homogeneous appearing lamella of cummingtonite, traverses the 40 μm -thick actinolite lamella, crosses another thick cummingtonite lamella, and ends within an area of actinolite. Quantitative analyses (Table 5) were made at positions 1 and 4 at the centers of the actinolite and cummingtonite lamellae, respectively, and at 2 and 3, on the right and left borders but within the thick actinolite lamella. The probe scans show that cummingtonite is much more nearly homogeneous than actinolite, particularly with respect to calcium. The probe scan for calcium across the thick actinolite lamella clearly reveals several lows in the calcium counting rates which correspond to thin cummingtonite lamellae (Fig. 5). As expected, the scan for iron shows that cummingtonite is enriched in this element with respect to actinolite. The scan for Mg indicates that there is nearly an equal concentration of this element in the two phases.

The analysis made in the center of the thick actinolite lamella (position 1, Fig. 4A; analysis 1, Table 5) shows an

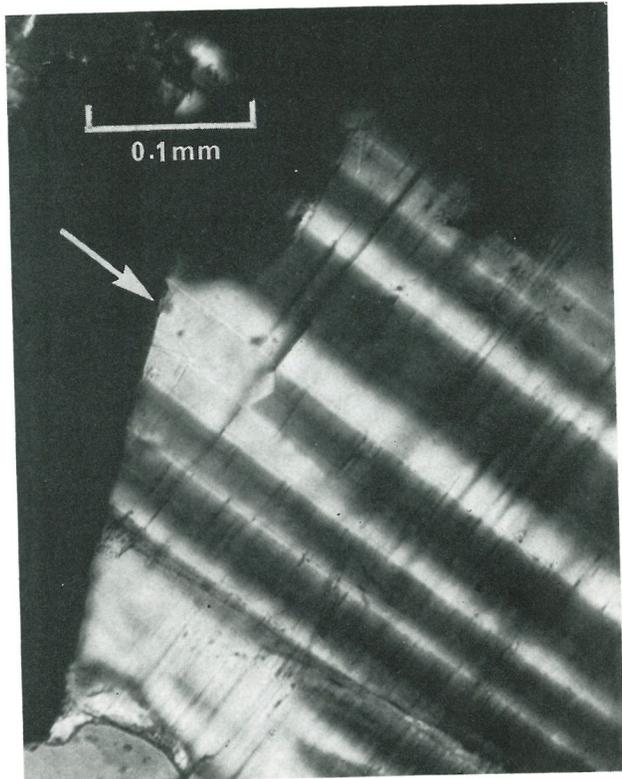
unusually low calcium content, *i.e.* $\text{Ca}_{1.19}(\text{Mg,Fe})_{6.06}\text{Si}_{7.8}\text{O}_{22}(\text{OH})_2$. This composition approaches the composition predicted from analysis of the single-crystal X-ray patterns of the bulk crystals in which the relative intensities indicate a ratio of actinolite $[\text{Ca}_2(\text{Mg,Fe})_5\text{Si}_8\text{O}_{22}(\text{OH})_2]$ to cummingtonite $[(\text{Mg,Fe})_7\text{Si}_8\text{O}_{22}(\text{OH})_2]$ of 45:55, suggesting the composition $\text{Ca}_{0.9}(\text{Mg,Fe})_{6.1}\text{Si}_8\text{O}_{22}(\text{OH})_2$ for the crystal before unmixing. The border zones (positions 2 and 3, Fig. 4A) are much higher in calcium, and analysis 3 gives an actinolite composition close to what we would expect for the completely unmixed actinolite, *i.e.*, $\text{Ca}_2(\text{Mg,Fe})_5\text{Si}_8\text{O}_{22}(\text{OH})_2$. As in the Gouverneur samples, the greatest partition of calcium is found at the border zones. Analysis 4, made in the center of a cummingtonite lamella, shows a very low calcium content, 0.095 atom per formula unit, which certainly indicates a nearly complete removal of the actinolite component. Two host cummingtonite grains found in the same polished section as the actinolite host grain shown in Figure 4A (analyses 1-4, Table 5) were also analyzed (analyses 5 and 6, Table 5). These two analyses agree well within the analytical error and do not differ greatly from the composition of the cummingtonite lamella included within the actinolite host (analysis 4).

The principal errors in the chemical analyses given in Table 5, other than those inherent to the electron probe, are: (1) inability to detect accurately small amounts of sodium and aluminum which probably occur in the actinolite phase, and (2) the assumption that all iron is ferrous. Silicon is low in analyses 1, 2, 3, and 4 suggesting that 0.1 to 0.2 atoms of Al substitute for Si. It is probable that some ferric iron is present in the actinolite. If Fe^{3+} is present, but assumed to be Fe^{2+} , the number of cations will be greater than 15 if the composition is calculated on the basis of $\text{O} = 22$ and $(\text{OH}) = 2$. None of the analyses deviates far from a cation sum of 15 which indicates that there is not a high concentration of ferric iron in these amphiboles. Probe analyses on the other three Ruby Mountain specimens have not yet been made, but a separate study in cooperation with P. W. Weiblen is planned.

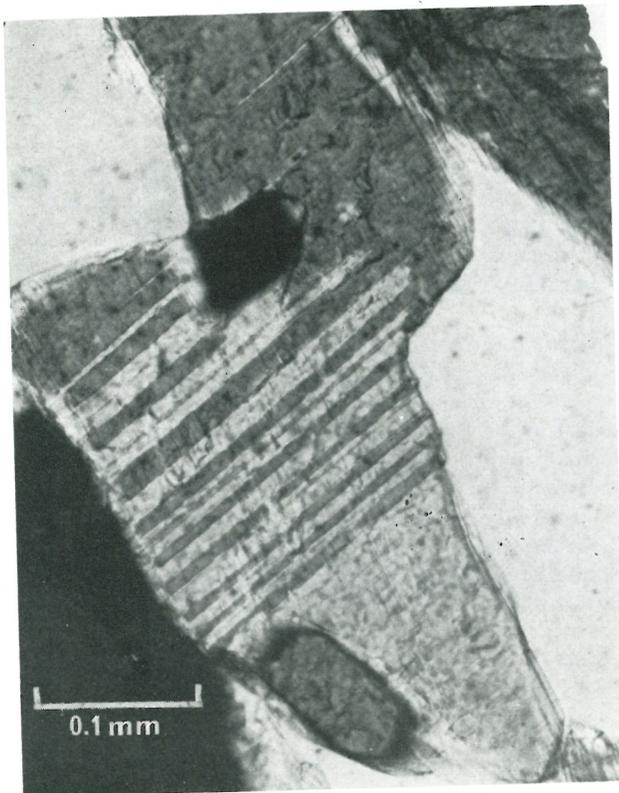
Exsolution and recrystallization textures in the Ruby Mountain amphiboles. The Ruby Mountain actinolites possess rather different intergrowth textures than those observed in the Gouverneur tremolites and cummingtonites. In the latter, the lamellae are always about 1 micron thick and oriented parallel to $(\bar{1}01)$; in the Ruby Mountain samples, the cummingtonite lamellae vary in thickness from about one micron to tens of microns and may be oriented parallel to both $(\bar{1}01)$ and (100). Furthermore, the solubility of the cummingtonite component in actinolite from samples HJ-163-60 and 182-60 is much greater than the solubility of cummingtonite in the Gouverneur tremolites. The host cummingtonite amphiboles from these two localities are more similar texturally in that they contain 5 to 10 percent calcic amphibole as thin lamellae, usually oriented parallel to $(\bar{1}01)$.



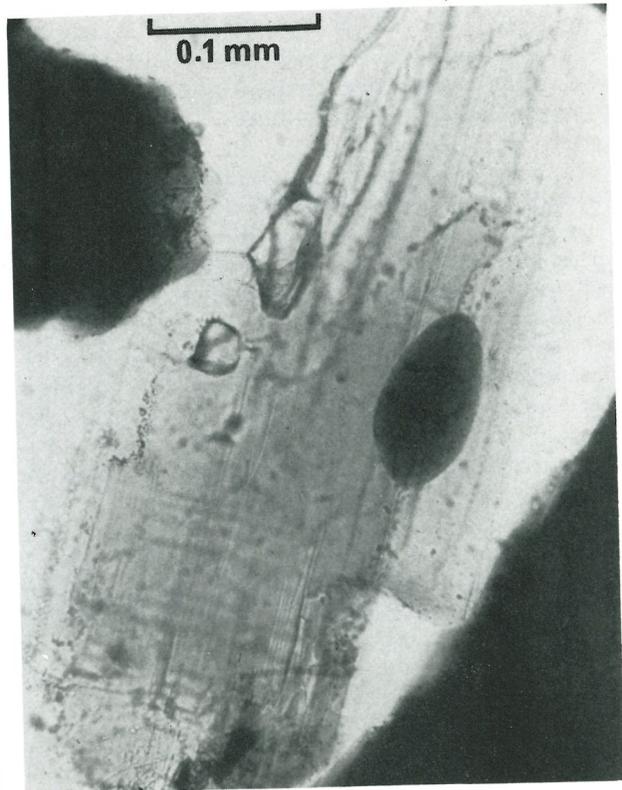
A



C



B



D

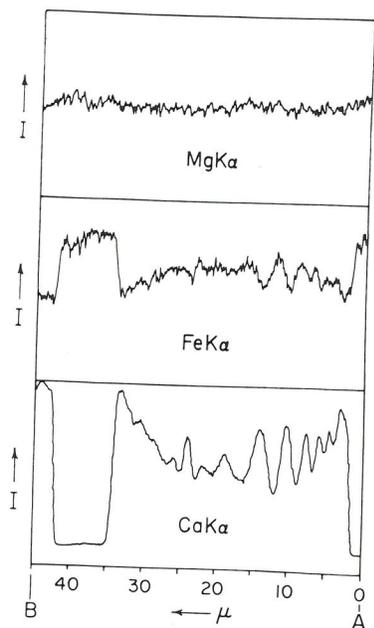


FIG. 5. Electron microprobe scans for Ca, Fe, and Mg content in the actinolite host grain (sample HJ-182-60) shown in Fig. 4A. The scans were made along the trace A-B. Intensity is in terms of CaK α , FeK α , and MgK α emission counting rates.

Many of the actinolite grains from samples HJ-163-60 and HJ-182-60 possess intergrowth textures which indicate that partial to nearly complete intragranular recrystallization has taken place. A sequence of recrystallization textures is pictured in four colored photographs (Fig. 7). The reddish-brown actinolite (Fig. 7A) contains fairly broad ($\bar{1}01$) lamellae of cummingtonite. Coalescence of two of these lamellae can be seen in the top-center of the photograph. Patches of homogeneous appearing cummingtonite also appear at the upper left and lower right. The actinolite grain shown in Figure 7B contains a complex grid of (100) and ($\bar{1}01$) cummingtonite lamellae (blue). The (100)

TABLE 5. ELECTRON MICROPROBE CHEMICAL ANALYSES OF ACTINOLITE AND CUMMINGTONITE HOST GRAINS FROM SAMPLE HJ 182-60, RUBY MOUNTAINS, MONTANA^a

Actinolite							
Analysis	Wt % ^b			No. of atoms ^c			
	1	2	3	1	2	3	
SiO ₂	52.84	53.06	52.91	7.877	7.783	7.772	Si
FeO	18.62	15.59	14.62	2.321	1.911	1.795	Fe ²⁺
MgO	16.84	17.56	16.96	3.740	3.838	3.711	Mg
CaO	7.43	10.73	12.40	1.186	1.685	1.951	Ca
Σ	95.73	96.94	96.89	15.124	15.217	15.229	Σ

Cummingtonite							
Analysis	Wt. % ^d			No. of atoms ^c			
	4	5	6	4	5	6	
SiO ₂	53.26	53.14	53.26	7.879	7.945	7.928	Si
FeO	24.28	24.99	25.40	3.003	3.124	3.161	Fe ²⁺
MgO	18.80	17.45	17.52	4.143	3.887	3.884	Mg
CaO	0.60	0.61	0.62	0.095	0.098	0.098	Ca
Σ	96.94	96.19	96.80	15.120	15.054	15.071	Σ

^a Analyst, Paul W. Weiblen.
^b Analyses at positions 1, 2, and 3 of Fig. 4A.
^c Number of atoms on the basis of O=22, OH=2.
^d Analysis at position 4 of Fig. 4A. Analyses of 5 and 6 are of areas appearing to be free of actinolite of two different host cummingtonite grains.

lamellae tend to pinch out near the thicker ($\bar{1}01$) lamellae. This is similar to some of the textures observed in many sulfides (Schwartz, 1931). The actinolite grain shown in Figure 7C contains very thick ($\bar{1}01$) cummingtonite lamellae (blue) that probably coalesced from two or more thin lamellae. The large lamellae in turn have coalesced into homogeneous appearing patches (lower left, upper left and right). Figure 7D illustrates the nearly complete coalescence of cummingtonite lamellae to form a large patch (blue) in the lower portion of the grain. Thin, relict ($\bar{1}01$) cummingtonite lamellae (trending 10-4 o'clock) appear as extensions from the large patch of cummingtonite into the adjacent area of actinolite (reddish-brown). Relict (100)



FIG. 4. Exsolution in amphiboles. A. Photomicrograph taken in the electron microprobe of an actinolite host grain. Dark lamellae are actinolite, colorless lamellae are cummingtonite oriented parallel to ($\bar{1}01$) of the host. Electron-probe scans (Fig. 5) for Ca, Fe, and Mg were made along the trace A-B. Electron-probe chemical analyses (Table 5) were made at points 1, 2, 3, and 4. Polished section, sample HJ-182-60, Ruby Mountains, Montana.

B. Photomicrograph of an actinolite host grain. The lower part of the grain (light areas) is nearly 100% cummingtonite; the upper part of the grain (dark areas) is nearly 100% actinolite. The center of the grain is composed of approximately equal amounts of cummingtonite and actinolite lamellae. Thin section, crossed polarizers, sample HJ-163-60, Ruby Mountains, Montana.

C. Photomicrograph of an actinolite host grain. The actinolite lamellae (dark bands) are retreating from the lower left part of the grain; note "ghosts" of old actinolite lamellae in this area. One actinolite lamella approximately 30 μ thick (marked by arrow) has retreated approximately 80 μ from the left edge of the grain. Thin section, crossed polarizers, sample HJ-182-60, Ruby Mountains, Montana.

D. Photomicrograph of a host C-centered cummingtonite grain. A dark bleb of hornblende appears within the homogeneous and colorless cummingtonite host. Oil mount, uncrossed polarizers, sample 7A8BX, Richmond area, New Hampshire.

cummingtonite lamellae (trending 2 o'clock-8 o'clock) appear in the reddish-brown host actinolite area at the upper right of this grain. It appears that the large patch of cummingtonite (lower part of grain) has formed by depletion of pre-existing lamellae in other parts of the grain.

The actinolite grain pictured in Figure 4B shows segregation of cummingtonite into the lower part (very light gray area) and actinolite into the upper part of the grain (dark gray area). The central part of the grain still contains thick (101) lamellae of both phases. Resorption of thick (101) actinolite lamellae can be seen in Figure 4C. The 30 micron thick actinolite lamella (marked by arrow), has retreated 80 microns from the left margin of the grain. "Ghosts" of actinolite lamellae are seen in the lower left portions of the grain, an area where coalescence of cummingtonite is taking place.

There is no evidence that extensive metasomatism in the Carter Creek iron deposit produced the textures described. Nor can they be explained as primary intergrowth textures, for it is inconceivable that the complex (101) and (100) orientations of the host and lamellar phases could arise during nucleation and crystal growth. Also, it has been demonstrated (Table 4) that the proportion of host to lamellar phase is constant within a given sample. If replacement of one phase by another or if primary intergrowth of two amphibole phases had occurred then a random variation of the relative proportion of the two phases in each grain would be expected. Toulmin (1964, p. A29) used this criterion to show that micropertthites (from the Peabody Granite, Salem, Massachusetts area) originated by exsolution rather than by "albitization" of a preexisting potassium feldspar. Toulmin states that the bulk compositions of different micropertthite crystals should vary erratically if metasomatism took place. The fact that the crystals do not show such variation is very strong evidence for exsolution as the origin of the micropertthite texture.

The following approximate ratios of host to lamellar amphibole phase were found in the Ruby Mountain samples (Table 4):

Sample	Host	Host
	actinolite:cummingtonite lamellae	cummingtonite:actinolite lamellae
HJ 182-60	45:55	93:7
HJ 163-60	55:45	95:5
HJ 162-60	70:30	—
HJ 167-60	80:20	—

Three of the four samples (182, 163, 162) came from the same drill core and were separated from each other by only a few feet. Thus they were probably subjected to almost identical *P-T* conditions. The bulk compositions of the actinolite grains of samples HJ-182-60 and HJ-163-60 are unusual and demonstrate a very large miscibility of cummingtonite in actinolite at the initial *T*, *P*, and composition of these rocks. The miscibility of the actinolite com-

ponent with the cummingtonite is much less in these samples. Optical inspection of cummingtonite grains from sample HJ 167-60 and HJ 162-60 also indicates a low solubility of the actinolite phase in the host cummingtonite.

Oxygen isotope determinations by Eugene C. Perry, Jr. (Univ. Minn., oral comm., January 30, 1969) on quartz-magnetite pairs from samples HJ 163-60, 182-60, and 109-60 give a Δ_{Q-M} of 9.5, 9.3, and 9.3 respectively. These values suggest minimum temperatures of metamorphism of about 620°C.

Compositions of the cummingtonite-actinolite associations observed in HJ-182-60 and HJ-163-60 suggest that the phase relations of these amphiboles are analogous to the phase relations along the join enstatite-diopside at 30 kbar (Davis and Boyd, 1966). In this system the enstatite limb of the solvus is very steep, with little of the calcic component soluble in the enstatite (Fig. 1, Davis and Boyd, 1966). The diopside limb of the solvus, on the other hand, is much less steep and shows a large solubility of the component $Mg_2Si_2O_6$ in diopside at higher temperatures.

Probable amphibole phase relations are given in Figure 6 which shows a hypothetical pseudo-binary join, cummingtonite-actinolite, with compositions and temperatures suggested by the Ruby Mountain samples. The actinolite field boundaries for samples HJ 182-60, 163-60, 162-60, and 167-60 are drawn increasing steepness in that order, as suggested by the apparent decrease in cummingtonite phase contained in the host actinolite grains. The inferred changes of slope may be due to change in iron-magnesium ratios, the steeper field boundaries representing more Mg-rich amphiboles, or may be due to differences in Na and Al content. Differences in chemical composition of the Ruby Mountain host cummingtonites are depicted as having much less effect on the cummingtonite solvi (Fig. 6). A very careful study of the Ruby Mountain amphibole compositions, both of the bulk grains and the unmixed phases, would be very useful in establishing the phase relations in the system $(MgFe)_7Si_8O_{22}(OH)_2-Ca_2(Mg,Fe)_5Si_8O_{22}(OH)_2$.

With the help of J. S. Huebner (U. S. Geol. Survey), several heating experiments were performed in an attempt to homogenize the two amphibole phases. A finely ground mix of actinolite and cummingtonite (sample HJ-163-60) was sealed in capsules with excess water. Two samples, one at 600°C and 1 kbar and another at 700°C and 1 kbar, both subjected to the graphite-methane buffer (Eugster and Skippen, 1967) for three weeks, showed no change in cell parameters, as determined by least-squares refinement of X-ray powder data. In a third experiment, the samples were heated in an unbuffered system and, after four weeks at 800°C and 2 kbar, recrystallization and decomposition occurred to yield magnetite, quartz, and two amphiboles whose cell parameters differed slightly from the unheated actinolite and cummingtonite (HJ-163-3F, Table 4). These reaction products were then finely ground and reheated with a methane buffer at 680°C and 1 kbar. By the end of

10 weeks, recrystallization had again occurred. Most of the quartz and magnetite were gone, and two amphiboles, whose d spacings differed only slightly from the previous ones, had formed. It is not clear from these experiments whether true homogenization could, in fact, be successfully accomplished.

It is clear that it is extremely difficult to homogenize the Gouverneur and Ruby Mountain amphiboles. Yund and McCallister (1968) state that exsolution lamellae in pyroxenes and amphiboles may be the result of "discontinuous precipitation" and if this is so homogenization of the phases will be much slower than their exsolution.

AMPHIBOLES FROM METAMORPHOSED VOLCANIC ROCKS OF MASSACHUSETTS AND NEW HAMPSHIRE

Robinson and Jaffe (1969) give a detailed report on the mineral associations and chemistry, including forty electron-probe and fourteen wet chemical analyses, of forty five amphiboles from the Orange and Quabbin Reservoir areas of Massachusetts and New Hampshire. In cooperation with Robinson and Jaffe we have examined eighteen of their specimens by X-ray single-crystal techniques in order to determine the structural state of the amphibole intergrowths and the relative proportion of host and exsolved phases. In this paper we will discuss only those observations that pertain to exsolution and recrystallization; the reader is referred to Robinson and Jaffe (1969) for further data on these specimens.

The amphiboles occur in amphibolites and related rocks of the Middle Ordovician Ammonoosuc Volcanics and Partridge Formation (Robinson, 1963, 1967). The rocks, initially basaltic lavas or tuffs, were metamorphosed to the kyanite and sillimanite zones under P - T conditions estimated by Robinson and Jaffe as 600–700°C and 5–7 kbar total pressure. Most of the samples studied here were collected in the "Amphibole Hill area" (Robinson and Jaffe, 1969, Fig. 1).

The X-ray unit-cell parameters and relative amounts of host and exsolved phase are given in Table 6 for four gedrites, two coexisting hornblende-anthophyllite pairs, two coexisting hornblende-cummingtonite pairs, and one coexisting three-amphibole assemblage (hornblende-cummingtonite-anthophyllite). In a second three-amphibole rock (I38A) intergrain contacts were observed between hornblende and cummingtonite and between cummingtonite and anthophyllite, but not between anthophyllite and hornblende. Commonly the host hornblende, cummingtonite, anthophyllite, and gedrite¹ grains contain, respectively, exsolution lamellae of cummingtonite, hornblende, gedrite, and anthophyllite. The amount of exsolved phase in these specimens ranges from zero to about fifty weight percent of the grain.

¹ Gedrite is a sodium-aluminum-rich orthoamphibole isostructural with anthophyllite. There is a miscibility gap between these two phases (Robinson, Jaffe, Klein, and Ross, 1969; Stout, 1969).

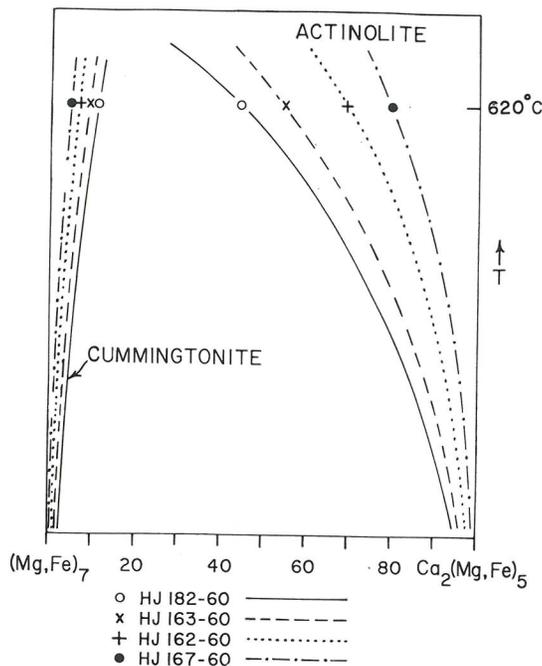


FIG. 6. Possible amphibole phase relations along the hypothetical pseudo-binary join cummingtonite-actinolite. The four Ruby Mountain coexisting cummingtonite-actinolite assemblages are plotted on the T - X diagram assuming the approximate proportions obtained from X-ray photographs and a temperature of 620°C.

The chemical formulas of the amphiboles listed in Table 6 are presented in Table 7, which shows the kinds and amounts of cations occupying the A , $M(4)$, octahedral, and tetrahedral crystallographic sites. The formulas were calculated on the basis of $O = 22$ and $OH + F = 2$ from the analyses given by Robinson and Jaffe (1969). Most of these analyses represent the bulk chemical compositions of the grains, and not the host phase alone.

The five hornblendes (Table 7) contain 0.24 to 0.44 atoms of R^{2+} (where $R^{2+} = Mg + Fe + Mn + Ni$) substituting for $Ca + Na$ in the $M(4)$ sites, so that unmixing of cummingtonite is expected. The four cummingtonites contain 0.06 to 0.29 atoms of $Ca + Na$ substituting for R^{2+} in the $M(4)$ site. The crystal-chemical reasons for unmixing in anthophyllite and gedrite are not completely clear but appear to have little to do with $M(4)$ occupancy since both types of orthoamphibole contain similar amounts of calcium and sodium in this position (Table 7). The A -site sodium and octahedral and tetrahedral aluminum content must be the main cause for unmixing. There appears to be a large miscibility of gedrite component in anthophyllite at high temperatures as witnessed by sample N30X in which gedrite makes up about 50 percent of the anthophyllite grain (Table 6). One gedrite (I34I) may be a possible end-member with the approximate composition $Na_{0.5}R_{2.5}^{2+}(R_{3.5}^{2+}Al_{1.5})(Si_{6.0}Al_{2.0})O_{22}(OH)_2$. The two aluminum atoms in tetrahedral coordination are charge-compensated by 1.5 oc-

TABLE 6. CRYSTALLOGRAPHIC DATA FOR AMPHIBOLES FROM METAMORPHOSED VOLCANICS FROM MASSACHUSETTS AND NEW HAMPSHIRE

No.	Crystal No.	Amphibole Intergrowths	% of grain	<i>a</i> (Å)	<i>b</i> (Å)	<i>c</i> (Å)	β	Space Group	<i>V</i> (Å ³)	
6A9X	1	hornblende host	90	9.80	18.00	5.32	105°0'	<i>C2/m</i>	906.5	
		<i>P</i> -cummingtonite on (100)	7	9.51	18.00	5.31	102°15'	<i>P2₁/m</i>	888.3	
		<i>P</i> -cummingtonite on (101)	3	9.50	18.00	5.31	102°0'	<i>P2₁/m</i>	888.2	
	2	anthophyllite host gedrite	70 30	18.57 18.57	18.05 17.88	5.29 5.29	— —	<i>Pnma</i> <i>Pnma</i>	1773.1 1756.4	
N30X	1	anthophyllite host gedrite	50 50	18.58 18.58	18.04 17.84	5.29 5.29	— —	<i>Pnma</i> <i>Pnma</i>	1753.5 1773.1	
		2	hornblende host <i>P</i> -cummingtonite on (101) <i>P</i> -cummingtonite on (100)	90 5 5	9.81 9.49 9.49	18.02 18.02 18.02	5.31 5.30 5.30	105°0' 101°55' 102°5'	<i>C2/m</i> <i>P2₁/m</i> <i>P2₁/m</i>	906.7 886.8 886.3
	7E8BX	1	<i>C</i> -cummingtonite	100	9.50	18.18	5.31	101°55'	<i>C2/m</i>	897.3
		2	hornblende host <i>C</i> -cummingtonite on (100)	99 1	9.77 9.51	18.02 18.02	5.32 5.30	104°40' 102°5'	<i>C2/m</i> <i>C2/m</i>	906.1 888.1
7A8BX	1	<i>C</i> -cummingtonite host	92					<i>C2/m</i>		
		hornblende on (100)	4					<i>C2/m</i>		
		hornblende on (101)	4					<i>C2/m</i>		
	2	<i>C</i> -cummingtonite host	88	9.51	18.15	5.31	102°5'	<i>C2/m</i>	896.2	
		hornblende on (101)	10	9.82	18.15	5.33	104°50'	<i>C2/m</i>	918.3	
		hornblende on (100)	2					<i>C2/m</i>		
	3	hornblende host	90					<i>C2/m</i>		
		<i>C</i> -cummingtonite on (101)	10					<i>C2/m</i>		
4	hornblende host	94	9.82	18.12	5.31	104°55'	<i>C2/m</i>	913.0		
	<i>C</i> -cummingtonite on (100)	3	9.51	18.12	5.33	102°10'	<i>C2/m</i>	897.8		
	<i>C</i> -cummingtonite on (101)	3					<i>C2/m</i>			
I34JX	1	gedrite host	>99	18.55	17.81	5.29	—	<i>Pnma</i>	1746.8	
		rutile	<1	4.58	—	—	—	—	—	
I34-I	1	gedrite	100	18.601(4) ^a	17.839(3)	5.284(2)	—	<i>Pnma</i>	1753.2(5)	
I38DX	1	gedrite host	80	18.56	17.87	5.29	—	<i>Pnma</i>	1754.5	
		anthophyllite	20	18.56	18.11	5.29	—	<i>Pnma</i>	1778.1	
W95JX	1	gedrite host	80	18.54	17.76	5.27	—	<i>Pnma</i>	1735.2	
		anthophyllite	20	18.54	17.98	5.27	—	<i>Pnma</i>	1756.8	
I38A	A	<i>C</i> -cummingtonite host	90	9.49	18.13	5.30	102°5'	<i>C2/m</i>	891.7	
		hornblende on (100)	6	9.80	18.13	5.30	104°45'	<i>C2/m</i>	910.6	
		hornblende on (101)	4							
	C-A	<i>C</i> -cummingtonite host	98					<i>C2/m</i>		
		hornblende on (100)	2					<i>C2/m</i>		
	1	hornblende host	85	9.80	18.08	5.31	104°45'	<i>C2/m</i>	909.8	
		<i>C</i> -cummingtonite on (100)	10	9.52	18.08	5.31	102°10'	<i>C2/m</i>	893.4	
		cummingtonite on (101)	5					<i>C2/m</i> ?		
2	anthophyllite host	55	18.58	18.11	5.28	—	<i>Pnma</i>	1776.6		
	gedrite	45	18.58	17.85	5.28	—	<i>Pnma</i>	1751.1		
QB27C	1-H	hornblende host	75	9.79	18.02	5.31	104°55'	<i>C2/m</i>	905.2	
		<i>P</i> -cummingtonite on (100)	25	9.48	18.02	5.31	102°5'	<i>P2₁/m</i>	887.0	
	1-P	<i>P</i> -cummingtonite host	85	9.45	18.00	5.30	102°5'	<i>P2₁/m</i>	881.6	
		hornblende on (100)	15	9.75	18.00	5.30	104°55'	<i>C2/m</i>	898.8	
	2-A	anthophyllite host	80	18.57	18.00	5.30	—	<i>Pnma</i>	1771.6	
		gedrite	20	18.57	17.81	5.30	—	<i>Pnma</i>	1752.9	

^a Unit-cell parameters obtained by least-squares refinement of X-ray diffraction powder data.

TABLE 7. CHEMICAL FORMULAS OF SEVENTEEN AMPHIBOLES FROM METAMORPHOSED VOLCANIC ROCKS FROM MASSACHUSETTS, AND NEW HAMPSHIRE (ROBINSON AND JAFFE, 1969; ROBINSON, JAFFE, KLEIN, AND ROSS, 1969)

Site	A	M(4) ^a			Octahedral, M(1), (2), (3)				Tetrahedral ^d		W (OH, F)
		(Na, K)	(Ca)	(Na, Li)	(R ²⁺) ^b	(R ²⁺)	(Al, Fe ³⁺ , Cr ³⁺)	(Ti ⁴⁺)	Σ	Si ^c	
Gedrite, W95JX	0.274	0.083	0.067	1.850	3.913	1.062	0.025	5.000	6.685	1.315	2.000
Gedrite, I34JX	0.320	0.071	0.109	1.820	3.680	1.270	0.050	5.000	6.405	1.595	2.053
Gedrite, I34I	0.528	0.042	0.040	1.918	3.473	1.500	0.027	5.000	5.954	2.046	2.616
Gedrite, I38DX	0.328	0.054	0.119	1.827	3.815	1.152	0.033	5.000	6.564	1.436	2.402
Anthophyllite, N30X	0.179	0.114	0.012	1.874	4.371	0.608	0.021	5.000	7.178	0.822	2.309
Anthophyllite, 6A9X	0.135	0.112	0.096	1.792	4.386	0.589	0.025	5.000	7.315	0.685	2.301
Anthophyllite, QB27C ^e	0.050	0.060	0.054	1.886	4.534	0.466	0	5.000	7.534	0.466	—
Anthophyllite, I38A ^e	0.059	0	0.027	1.973	4.674	0.326	0	5.000	7.642	0.358	—
Cummingtonite, 7E8BX	0	0.131	0.119	1.750	4.671	0.318	0.009	4.998	7.781	0.219	2.204
Cummingtonite, 7A8BX	0.007	0.210	0.075	1.715	4.577	0.366	0.020	4.963	7.740	0.260	2.387
Cummingtonite, QB27C ^e	0	0.208	0.017	1.775	4.677	0.277	0	4.954	7.827	0.173	—
Cummingtonite, I38A ^e	0.022	0.049	0.007	1.944	4.918	0.082	0	5.000	7.904	0.096	—
Hornblende, N30X	0.329	1.633	0.105	0.262	3.740	1.183	0.077	5.000	6.435	1.565	2.301
Hornblende, 6A9X	0.348	1.525	0.193	0.282	3.761	1.148	0.091	5.000	6.503	1.497	2.489
Hornblende, 7A8BX	0.281	1.521	0.228	0.251	3.862	1.028	0.110	5.000	6.708	1.292	2.306
Hornblende, 7E8BX	0.460	1.526	0.231	0.243	3.599	1.366	0.035	5.000	6.333	1.667	2.659
Hornblende, QB27C ^e	0.347	1.559	0	0.441	4.134	0.917	0	5.051	6.629	1.371	—

^a Σ = 2.000 atoms in M(4) position, no. of atoms on the basis of O = 22, OH + F = 2.

^b R²⁺ = Mg²⁺ + Fe²⁺ + Mn²⁺ + Ni²⁺.

^c Includes 0.002 to 0.009 atoms of P.

^d Σ = 8.000 atoms in tetrahedral positions.

^e Electron probe analysis, C. Klein, Jr., analyst.

tetrahedral aluminum atoms and 0.5 A-site sodium atoms. The work of Robinson and Jaffe clearly demonstrates that some gedrites contain essential A-site sodium.

Robinson and Jaffe (1969) point out that hornblendes associated with anthophyllite (N30X, 6A9X) contain less iron than do hornblendes associated with cummingtonite (7A8BX, 7E8BX). Also, they noted that hornblendes associated with anthophyllite have unmixed the primitive variety of cummingtonite, whereas the hornblendes associated with cummingtonite have unmixed C-centered cummingtonite. This observation is consistent with our hypothesis that a C-centered cummingtonite with an M(4) site rich in ferrous iron will more readily accommodate calcium without symmetry change than a cummingtonite with an M(4) site rich in magnesium. Since ferrous iron has a larger radius (0.74 Å) than magnesium (0.66 Å), calcium with a radius of 1.03 Å will more readily mix with Fe²⁺ than with Mg²⁺.

Jaffe, Robinson, and Klein (1968) and Robinson (1963) have reported on the exsolution textures seen in some of the amphiboles listed in Tables 6 and 7. Our optical studies support their observations. Most commonly we observed very thin (101) and/or (100) exsolution lamellae in the host hornblendes and cummingtonite such as depicted by Jaffe, Robinson, and Klein (1968, Fig. 1). In some grains a nearly complete intragranular recrystallization has taken place such as seen in Figure 4D which shows an oblate bleb of hornblende in the right center of an otherwise homogeneous appearing cummingtonite grain.

AMPHIBOLES FROM IRON FORMATIONS AND OTHER ROCK TYPES

X-ray data for amphiboles from several other localities are listed in Table 8. The riebeckite-tremolite from the metamorphosed Precambrian Wabush Iron Formation, Labrador (Klein, 1966, analysis No. 13), has the composition (on the basis of 46 charges) (Na,K)_{0.30}[(Ca,Na)_{1.41}R²⁺_{0.59}]_{2.00}(R²⁺,Fe³⁺)_{5.00}(Si,Al)_{8.00}O₂₂(OH,F)₂ where R²⁺ = Mg + Fe + Mn. X-ray single-crystal patterns of this amphibole show that 10 to 20 percent of the grain is composed of primitive cummingtonite. The large amount of R²⁺ in the M(4) site indicates the probability of unmixed cummingtonite from this phase. Riebeckite-tremolite No. 15A (Klein, 1966, analysis 15A) has the composition (Na, K)_{0.27}[(Ca, Na)_{1.84}R²⁺_{0.16}]_{2.00}(R²⁺, Fe³⁺)_{5.00}(Si, Al, Fe³⁺)_{8.00}O₂₂(OH,F)₂. X-ray and optical examination show no exsolved phase which is consistent with the very low R²⁺ (0.16 atoms) M(4) site occupancy. The manganoan cummingtonites (Nos. 2, 3, 4; Table 8) from the Wabush Iron Formation contain 0.18 to 0.22 atoms of Ca + Na in the M(4) site (Klein, 1964, p. 966). There is no evidence for exsolution in these amphiboles.

Some of the samples collected by J. J. Norton (U. S. Geol. Survey) from metamorphosed iron-formation in the Black Hills, South Dakota, contain amphiboles with textures similar to those seen in the Ruby Mountain rocks. X-ray photographs of one grain of pleochroic green actinolite (sample JJN 18-67) reveals a very complex unmixed

intergrowth. Four separate diffraction patterns are registered in a single photograph, and indicate that the grain now consists of two amphiboles, actinolite and *C*-centered cummingtonite, each in two different orientations (Table 8). In addition to the cummingtonite oriented parallel to $(\bar{1}01)$ and (100) of the actinolite host, there is actinolite oriented parallel to $(\bar{1}01)$ of the (100) cummingtonite.

A specimen of eclogite (No. 1725) from the Eiksundsdal Eclogite Complex of western Norway (Schmitt, 1964) consists of garnet and omphacite with lesser amounts of orthopyroxene, biotite, hornblende, plagioclase, and apatite. According to Schmitt, the rock formed by the recrystallization of pre-existing mafic rock under conditions corresponding to the eclogite facies. A later metamorphism, of sillimanite grade, resulted in the formation of coronas of hornblende on omphacite. The three hornblende grains examined with X rays showed 15 to 35 percent intergrown primitive cummingtonite. One crystal (No. 2-B) contained in addition to primitive cummingtonite a third phase—probably omphacite. The textures and X-ray studies indicate a partial replacement or reaction of omphacite with hornblende and subsequent unmixing of primitive cummingtonite.

The Moxie pluton of west-central Maine consists of varieties of troctolite, norite, and gabbro (Espenshade and Boudette, 1967). Most of the rocks contain hornblende (as much as 21 volume percent) and a few contain cummingtonite coexisting with hornblende. The exsolution textures appearing in these amphiboles of igneous origin do not appear to be significantly different from those seen in many of the metamorphic rocks.

Unit-cell data is also given in Table 8 for amphiboles from the Biwabik Iron Formation (Bonnichsen, 1969, this volume); the Gunflint Iron Formation; the Middletown Gneiss, eastern Connecticut; and an amphibolite, Cherry Creek area, Madison County, Montana. All of these amphiboles show typical exsolution textures.

AMPHIBOLE PHASE RELATIONS

The stability and compositional relationships among *C*-centered cummingtonite, primitive cummingtonite, and anthophyllite are not well understood. The chemical compositions of coexisting anthophyllite and cummingtonite may be quite similar (Klein, 1968; Robinson and Jaffe, 1969). Also, the compositions of these two phases appear to overlap, particularly with respect to iron content. The primitive cummingtonites identified in this study have compositions which appear to overlap those of both anthophyllite and cummingtonite, but since the space-group symmetries of most analyzed cummingtonites have not been determined it is at present impossible to define compositional fields.

A few general observations on chemical compositions can be made. First, cummingtonites usually have iron/magnesium ratios greater than 0.8; a few have been reported with Fe/Mg ratios much lower. Second, anthophyllites containing little aluminum generally have Fe/Mg ratios of 0.4

or less. Third, cummingtonites are generally more calcic and less aluminous and sodic than are anthophyllites. Fourth, anthophyllites (variety gedrite) can contain large amounts of aluminum and sodium. Finally, anthophyllite can contain very large amounts of iron when coupled with high aluminum content.

At higher temperatures, where atoms have large thermal energies, calcium, magnesium, iron, and manganese ions vibrate with large amplitudes and thus mix more readily in the large $M(4)$ site of the cummingtonite structure. On cooling the thermal energies decrease and the structure is stabilized by removal of the calcium from the $M(4)$ site through exsolution of a calcic amphibole by inversion to a new structure type capable of better adapting to the mixed $M(4)$ site occupancy, and/or by exchanging some of the $M(4)$ magnesium ions with $M(1)$, (2), and (3) iron or manganese ions. The $M(4)$ site of $P2_1/m$ cummingtonite can more readily accommodate large and small cations such as calcium and magnesium than can the equivalent site in *C*-centered cummingtonite (Papike, Ross, and Clark, 1969).

Since this paper went to press, Prewitt, Papike, and Ross (unpublished data) found from heating experiments with the Buerger precession camera that the primitive manganoan cummingtonite from Gouverneur, N. Y. (sample 115046) inverts to *C*-centered cummingtonite at temperatures of approximately 50°C or less. This observation coupled with our previous studies suggest the following possible cooling reactions for cummingtonites:

1. A $C2_1/m$ cummingtonite with a high concentration of Ca and Mg in the $M(4)$ site and Mg in $M(1)$, (2), and (3) sites may on cooling unmix sufficient calcic amphibole so that its composition lies in the stability field of anthophyllite. Low iron and manganese in $M(1)$, (2), and (3) preclude homogeneous exchange with a large amount of $M(4)$ magnesium. Recrystallization to the Ca-poor orthoamphibole anthophyllite should then occur but requires a complete reconstitution of the crystal structure, including the breaking and reforming of chemical bonds. This type of recrystallization may not be possible unless a fluid phase is present, or it may be prevented by "structural control" of the host clinoamphibole. If recrystallization is prevented, the $P2_1/m$ phase may form and persist in a metastable state. The cummingtonites from the Gouverneur, New York area, which are very magnesium-rich, may show part of the reaction sequence described above, but the reaction has stopped well before complete unmixing of the Ca-component and inversion to anthophyllite.

It is conceivable that the primitive cummingtonite has a field of stability at low temperatures and/or at high pressures. (2) the more iron-rich $C2/m$ cummingtonites may retain the $C2/m$ symmetry throughout their cooling history due to the greater amount of iron in the $M(4)$ site; inversion only taking place when a significant amount of Mg enters this position. The cummingtonites from the Ruby Mountain locality are a paradox in that some crystals from the same sample show $P2_1/m$ symmetry while others do not. Also the

TABLE 8. CRYSTALLOGRAPHIC DATA FOR AMPHIBOLES FROM IRON FORMATIONS AND OTHER ROCK TYPES

Sample No.	Crystal No.	Amphibole Intergrowths	% of grain	<i>a</i> (Å)	<i>b</i> (Å)	<i>c</i> (Å)	β	Space Group	<i>V</i> (Å ³)
13, Wabush Iron Formation, Labrador, Klein (1966)	2	riebeckite-tremolite host <i>P</i> -cummingtonite on (101)	80 10	9.77 9.48	18.04 18.04	5.29 5.30	104°20' 102°15'	<i>C2/m</i> <i>P2₁/m</i>	903.3 885.8
	1	riebeckite-tremolite host <i>P</i> -cummingtonite on (101) <i>P</i> -cummingtonite on (100)	80 10 10	9.779(4) ^a	18.021(8)	5.285(3)	104°18'(2)	<i>C2/m</i> <i>P2₁/m</i> <i>P2₁/m</i>	902.5(6)
15A, Wabush Iron Formation, Labrador, Klein (1966)	1	riebeckite-tremolite	100	9.789(2) ^a	18.003(3)	5.291(1)	104°8'(1)	<i>C2/m</i>	904.3(2)
2, Wabush Iron Formation, Labrador, Klein (1966)	1, 2	Mn <i>C</i> -cummingtonite	100	9.583(3) ^b	18.091(5)	5.315(4)	102°38'(2)	<i>C2/m</i>	899.1(6)
3, Klein (1964)	1, 2	Mn <i>C</i> -cummingtonite	100	9.560(2)	18.089(4)	5.309(4)	102°22'(2)	<i>C2/m</i>	896.8(5)
4, Klein (1964)	1	Mn <i>C</i> -cummingtonite	100	9.573(3)	18.115(5)	5.304(7)	102°21'(4)	<i>C2/m</i>	898.5(8)
JJN 18-67 Iron formation, Black Hills, S.D.	1	actinolite host	50	9.87	18.24	5.31	104°50'	<i>C2/m</i>	924.1
		<i>C</i> -cummingtonite on (100)	40	9.56	18.24	5.32	102°0'	<i>C2/m</i>	907.4
		<i>C</i> -cummingtonite on (101)	5	9.57	18.24	5.33	102°5'	<i>C2/m</i>	909.8
		actinolite on (101) of (100) cummingtonite	5	9.87	18.24	5.32	104°55'	<i>C2/m</i>	925.5
M12024 Biwabik Iron Formation, Dunka River, Minnesota, Bonnicksen (1969)	3	<i>C</i> -cummingtonite host	90	9.55	18.28	5.32	102°10'	<i>C2/m</i>	907.9
		hornblende on (100)	5	9.85	18.14	5.32	105°0'	<i>C2/m</i>	918.2
		hornblende on (101)	5						
GUN 306-B Gunflint Iron Formation, N. Minnesota	3	actinolite host	65	9.90	18.29	5.32	104°45'	<i>C2/m</i>	931.6
		<i>C</i> -cummingtonite on (101)	35	9.54	18.29	5.33	102°5'	<i>C2/m</i>	909.4
F-4-9 Middletown Gneiss, Connecticut (Dixon, Eaton and Lundgren, 1968)	2	<i>P</i> -cummingtonite host	90	9.48	18.08	5.29	102°0'	<i>P2₁/m</i>	886.9
		hornblende on (100)	7	9.77	18.08	5.29	104°45'	<i>C2/m</i>	903.6
		hornblende on (101)	3					<i>C2/m</i>	
1725, Eclogite, Norway, Schmitt (1964)	1	hornblende host	85	9.85	18.01	5.29	104°45'	<i>C2/m</i>	907.5
		<i>P</i> -cummingtonite on (100)	10	9.50	18.01	5.29	102°0'	<i>P2₁/m</i>	885.3
		<i>P</i> -cummingtonite on (101)	5	9.49	18.01	5.29	101°55'	<i>P2₁/m</i>	884.7
	2-A	hornblende host	65	9.85	18.02	5.30	104°50'	<i>C2/m</i>	909.4
		<i>P</i> -cummingtonite on (100)	35	9.50	18.02	5.29	102°0'	<i>P2₁/m</i>	885.8
	2-B	hornblende host	75					<i>C2/m</i>	
		<i>P</i> -cummingtonite on (100)	12				<i>P2₁/m</i>		
		omphacite (?) on (100)	13						
674, gabbro, Moxie pluton, Maine, Espenshade and Boudette (1967)	C	<i>C</i> -cummingtonite host	85					<i>C2/m</i>	
		hornblende on (101)	10					<i>C2/m</i>	
		hornblende on (100)	5					<i>C2/m</i>	
CC352C, amphibolite, Madison Co., Montana Rabbitt (1948)	1	<i>C</i> -cummingtonite host	95	9.53	18.28	5.33	101°55'	<i>C2/m</i>	908.5
		hornblende on (101)	5	9.83	18.28	5.34	104°55'	<i>C2/m</i>	927.2

^a Unit-cell parameters obtained by least-squares refinement of X-ray diffraction powder data (Klein, 1966).

^b Unit-cell parameters obtained by least-squares refinement of X-ray diffraction powder data (Klein, 1964).

reflections violating $C2/m$ symmetry are weak, often diffuse. It could be that the inversion temperature is close to room temperature for amphiboles of these compositions and what we are seeing in the X-ray photographs is a $C2/m$ cummingtonite that inverted from the $P2_1/m$ temperature soon after exposure to the X radiation.

The cummingtonite which is unmixed from the calcic host phase (tremolite, actinolite, or hornblende) usually has the same unit-cell parameters and space-group symmetry as the coexisting host cummingtonite so the reaction sequences suggested above could equally apply to exsolved cummingtonite.

Pigeonite is the pyroxene analog of primitive cummingtonite and unmixing of primitive cummingtonite from calcic amphibole such as actinolite is analogous to the unmixing of pigeonite from augite. On the other hand, inversion of pigeonite to orthopyroxene plus augite is common, whereas the analogous inversion of primitive cummingtonite to anthophyllite is as yet unknown. This may be because the pyroxenes have cooled from much higher temperatures. Also, inversion to the orthorhombic structure type may not be possible unless diffusion can occur between the mineral grains and the pore fluids. If little or no fluid phase is present inversion through reconstitution of the crystal structure may be very sluggish and there will be a tendency for metastable structures to persist.

In conclusion, we suggest that to make further progress the composition fields for anthophyllite, gedrite, (Al,Fe)-rich anthophyllite, and primitive and C-centered cummingtonite must be clearly defined. Phase equilibrium studies

will be of little assistance in solving some of these problems unless some method is found to synthesize crystals large enough for single-crystal X-ray study. Many of these amphiboles simply cannot be characterized structurally by X-ray powder methods alone. Our main effort must still be to exactly characterize coexisting phases obtained from rocks that are well understood geologically.

ACKNOWLEDGMENTS

We wish to thank Peter Robinson and Howard W. Jaffe (University of Massachusetts) for furnishing many samples and for advising us on petrological interpretations; to Harold L. James (U. S. Geological Survey) for providing thin sections and samples from the Ruby Mountain area; and to the many others, especially Cornelis Klein, Jr. (Harvard University), Bill Bonnichsen (Cornell University), Gilbert H. Espenshade and James J. Norton (U. S. Geological Survey), and H. H. Schmitt (NASA), who furnished amphibole samples.

We further wish to thank Paul W. Weiblen (University of Minnesota) for electron-probe analyses of sample HJ-182-60; J. J. Fahey (U. S. Geological Survey) and A. T. Anderson, Jr. (University of Chicago) for chemical and probe analyses of tremolite and primitive manganese cummingtonite; Eugene C. Perry, Jr. (University of Minnesota) for oxygen isotope studies on the Ruby Mountain samples; to J. Stephen Huebner (U. S. Geological Survey), and M. Charles Gilbert (V.P.I.) for assistance in the homogenization experiments; to Edward J. Dwornik (U. S. Geological Survey) for taking the electron micrographs; to Joan R. Clark, Daniel E. Appleman, and Judith A. Konnert (U. S. Geological Survey) for assistance in X-ray and computation problems; and to Priestley Toulmin, III, and Fred Barker (U. S. Geological Survey) for their helpful criticism of the manuscript.

REFERENCES

- ASKLUND, B. (1923) Petrological studies in the neighborhood of Stavsjö at Kolmårten. *Sveriges Geol. Unders. Årsb.* **17**, no. 6, 122 pp.
- , W. L. BROWN, AND J. V. SMITH (1962) Hornblende-cummingtonite intergrowths. *Amer. Mineral.* **47**, 160–163.
- BINNS, R. A. (1965) The mineralogy of metamorphosed basic rocks from the Willyama Complex, Broken Hill district, New South Wales, Part I. Hornblendes. *Mineral. Mag.* **35**, 306–326.

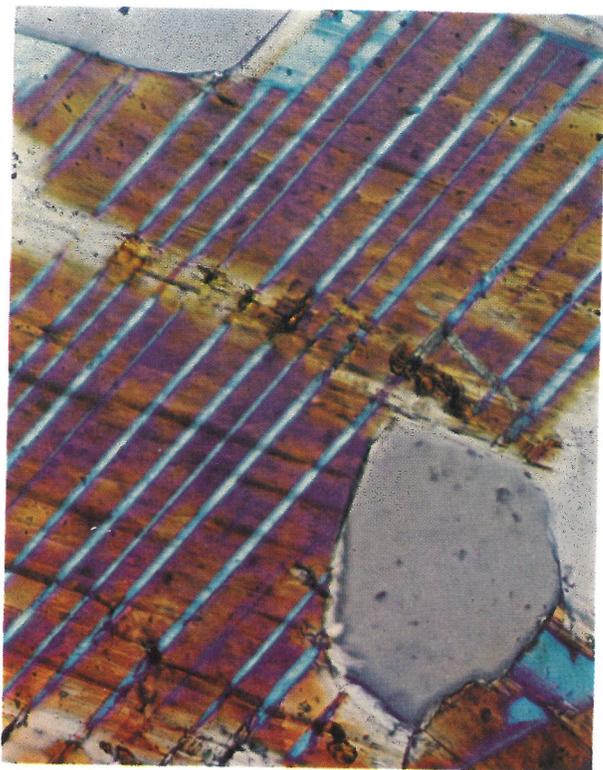
»»»→

FIG. 7. Exsolution in actinolite from Ruby Mountains, Montana. A. Grain consisting of about 80 wt% actinolite and 20% cummingtonite. Actinolite host areas appear reddish-brown; thin cummingtonite lamellae appear blue and are oriented parallel to $(\bar{1}01)$ of the actinolite. A pair of cummingtonite lamellae (blue) have begun to coalesce (top-center of photo). Thin section, crossed polarizers, blue filter, sample HJ-167-60, magnification 250 \times .

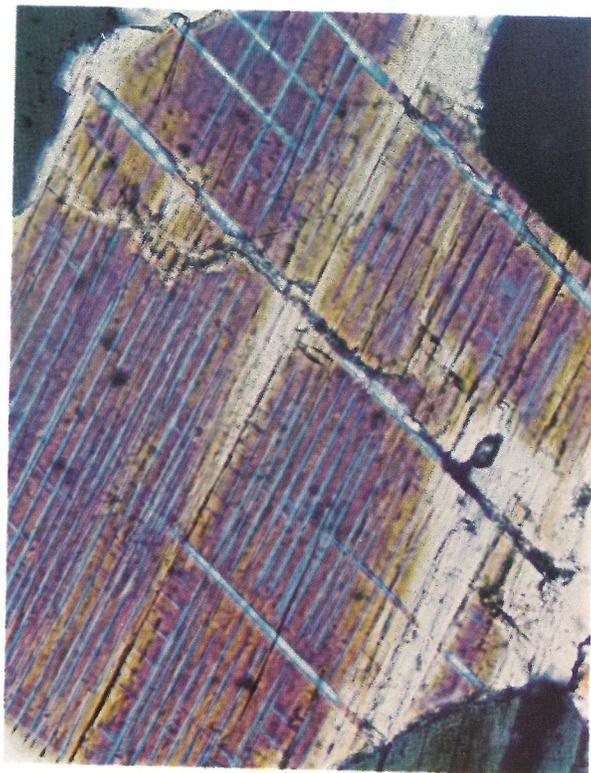
B. Same thin section as Fig. 7A. The primitive cummingtonite lamellae (blue) are oriented parallel to both $(\bar{1}01)$ (trending 11–5 o'clock) and (100) (trending 1–7 o'clock) of the actinolite (brown). Patchy area at the lower right side of the grain is mostly actinolite. Crossed polarizers, blue filter, magnification 250 \times .

C. Actinolite (gray-green) with cummingtonite lamellae (blue) oriented parallel to $(\bar{1}01)$. On the lower left and upper left and right, the cummingtonite lamellae have coalesced into homogeneous patches. The grain consists of approximately 55 wt% actinolite and 45 wt% cummingtonite. Thin section, crossed polarizers, blue filter, sample HJ-163-60, magnification 100 \times .

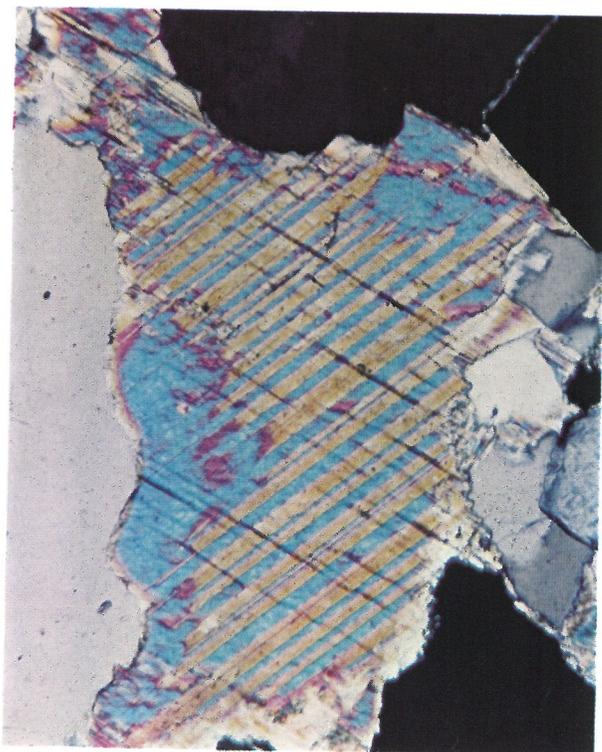
D. Same thin section as shown in Figs. 7A, 7B. The cummingtonite lamellae (blue) have coalesced mostly into one large patch in the lower right portion of the grain. The thin $(\bar{1}01)$ cummingtonite lamellae (trending 10–4 o'clock) are disappearing from the center and left of the grain. Relict cummingtonite lamellae (trending 2–8 o'clock) oriented parallel to (100) of the reddish-brown actinolite appear in the upper right part of the grain. Crossed polarizers, blue filter, magnification 250 \times .



A



B



C



D

EXSOLUTION TEXTURES IN AMPHIBOLES AS INDICATORS OF SUBSOLIDUS THERMAL HISTORIES¹

MALCOLM ROSS, J. J. PAPIKE, AND K. WIER SHAW
U. S. Geological Survey, Washington, D.C. 20242

ABSTRACT

New chemical, optical, and X-ray observations demonstrate that exsolution in calcic amphiboles, cummingtonites, and orthoamphiboles is common. Specimens studied include primitive manganoan cummingtonite and tremolite from talc schists, Gouverneur, New York; actinolite and primitive and C-centered cummingtonite from iron formation, Ruby Mountains, Montana; hornblende, anthophyllite, gedrite, and primitive and C-centered cummingtonite from metamorphosed volcanics, Massachusetts and New Hampshire; and amphiboles from four other iron formations, an amphibolite, a gneiss, an eclogite, and a gabbro. Most hornblendes, tremolites, and actinolites examined contain exsolution lamellae of either C-centered cummingtonite or primitive cummingtonite; the term "primitive" being given in allusion to the $P2_1/m$ space-group symmetry. Most primitive and C-centered cummingtonite studied contains exsolution lamellae of either hornblende, actinolite, or tremolite. Sodium- and aluminum-poor anthophyllite and sodium-aluminum-rich gedrite often contain exsolution lamellae of gedrite and anthophyllite, respectively. Some amphibole grains have undergone nearly complete intragranular recrystallization with two amphibole phases segregating into two distinct areas of the grain. Photomicrographs are given showing a variety of exsolution textures in amphiboles. These textures demonstrate the miscibility gap between calcium-rich and calcium-poor clinoamphiboles and between gedrite and anthophyllite. Primitive cummingtonite has a composition field that appears to partly overlap C-centered cummingtonite and anthophyllite.

Possible phase relations between primitive and C-centered cummingtonite and the calcic amphiboles are as follows: (1) a $C2/m$ cummingtonite highly enriched in Mg and containing some Ca, on cooling may exsolve a calcic clinoamphibole and either invert to Mg-anthophyllite or invert to a metastable $P2_1/m$ cummingtonite and (2) a more iron-rich cummingtonite will maintain space group symmetry $C2/m$ throughout its cooling history while unmixing the calcium component as calcic clinoamphibole.

INTRODUCTION

Complex amphibole intergrowths have been observed by many investigators and have been variously ascribed to: (1) primary intergrowth of two amphibole phases, (2) partial alteration or replacement of one amphibole by another, (3) overgrowth of one amphibole on another, and (4) exsolution or unmixing of two amphiboles from a homogeneous primary amphibole. Intergrowths of two amphiboles due to secondary effects such as overgrowth, alteration, and replacement can sometimes be recognized. It is more difficult to distinguish between primary intergrowths and exsolution textures.

Eskola (1914) was perhaps one of the first to observe what may be unmixing in amphiboles, although he interpreted cummingtonite-hornblende intergrowths as replacement of hornblende by cummingtonite. It appears that Asklund (1923) was the first to formally propose that a miscibility gap exists between hornblende and cummingtonite and that intergrowths of the two amphiboles are due to unmixing. This proposal was supported by Asklund, Brown, and Smith (1962) when they found, using X-ray and optical techniques, very fine lamellae of hornblende and cummingtonite within coexisting cummingtonite and hornblende respectively.

Seitsaari (1952) also described several specimens containing cummingtonite-hornblende intergrowths. He does not state that the textures are due to unmixing, but his Figure 4 clearly shows an exsolution-recrystallization tex-

ture of hornblende and a "colorless amphibole," which is probably cummingtonite. Recent reports of exsolution in amphiboles that have come to our attention are listed in Table 1.

The present study describes the amphibole intergrowth textures observed in specimens from thirteen localities. Three of these are discussed in detail: (1) tremolite and primitive manganoan cummingtonite from metasedimentary talc schists, northwest Adirondacks, (2) actinolite and cummingtonite from a metamorphosed iron formation, Ruby Mountains, Montana, and (3) hornblende, cummingtonite, anthophyllite, and gedrite from the metamorphosed volcanic rocks of the Orange and Richmond areas of Massachusetts and New Hampshire. By optical, single-crystal X-ray, and electron-microprobe techniques we will attempt to show that all the amphibole intergrowth textures observed may be explained as caused by unmixing, usually of a calcium-rich amphibole from a calcium-poor amphibole host or of a calcium-poor amphibole from a calcium-rich host. The unmixed amphibole phase usually appears as thin lamellae oriented parallel to $(\bar{1}01)$ and/or (100) of the host. In some specimens the lamellae are thicker and grade into patches of a nearly homogeneous amphibole. Such textures appear to be due to recrystallization within a single host grain resulting in the segregation of two amphiboles within the grain. No chemical exchange between grains, or between grains and an interstitial fluid, is indicated with the possible exception of oxidation of ferrous iron. Exsolution in amphiboles appears to be especially common in rocks containing two or more coexisting amphiboles.

Crystal structure determinations were obtained for three

¹ Studies of silicate minerals (15). Publication authorized by the Director, U. S. Geological Survey.

- BONNICHSEN, BILL (1969) Coexisting orthopyroxenes, inverted iron-rich pigeonites, Ca pyroxenes, and amphiboles from thermally metamorphosed iron formation, Dunka River area, Minnesota. *Mineral. Soc. Amer. Spec. Paper* 2, 217-239.
- BORIANI, A., AND L. MINUTTI (1965) La cummingtonite di Ornavasso. *Rend. Inst. Lombardo Sci. Lett.*, 99, 412-424.
- BOWN, M. G. (1965) A new amphibole polymorph in intergrowth with tremolite: clino-anthophyllite? (abstr.). *Amer. Mineral.* 51, 259-260 (1966).
- CALLEGARI, E. (1965-66) Osservazioni su alcune cummingtoniti del massiccio dell' Adamello. *Mem. Accad. Patavina, Cl. Sci. Mat.-Nat.* 78, 273-310.
- DAVIS, B. T. C., AND F. R. BOYD (1966) The join $Mg_3Si_2O_6$ - $CaMgSi_2O_6$ at 30 kilobars pressure and its application to pyroxenes from kimberlites. *J. Geophys. Res.* 71, 3567-3576.
- DIXON, H. ROBERTA, G. P. EATON, AND L. W. LUNDGREN, JR. (1968) A structural and stratigraphic cross-section traverse across eastern Connecticut. In P. M. Orville (Ed.) *Guidebook for Fieldtrips in Connecticut*. New England Intercollegiate Geol. Conf., 60th Ann. Meet., Yale Univ., New Haven, Conn., Trip F-4, p. 1-23.
- ENGEL, A. E. J. (1962) The Precambrian geology and talc deposits of the Balmat-Edwards district, northwest Adirondack Mountains, New York. *U. S. Geol. Surv. Open File Rep.*, 357 pp.
- , AND C. G. ENGEL (1953) Grenville series in the northwest Adirondack Mountains, New York. *Geol. Soc. Amer. Bull.* 64, 1013-1097.
- , AND — (1958) Progressive metamorphism and granulization of the major paragneiss, northwest Adirondack Mountains, New York. *Geol. Soc. Amer. Bull.*, 69, 1368-1414.
- , AND — (1960) Progressive metamorphism and granulization of the major paragneiss, northwest Adirondack Mountains, New York. Part 2. Mineralogy. *Geol. Soc. Amer. Bull.* 71, 1-58.
- , AND — (1962) Progressive metamorphism of amphibolite, northwest Adirondack Mountains, New York. In A. E. J. Engel, H. L. James, and B. F. Leonard, (Eds.) *Petrologic Studies: A Volume to Honor A. F. Buddington*. Geological Society of America, New York, p. 37-82.
- ESKOLA, P. (1914) On the petrology of the Orijärvi Region in south western Finland. *Bull. Comm. Geol. Finlande* 40, 167-224.
- ESPENSHADE, G. H., AND E. L. BOUDETTE (1967) Geology and petrology of the Greenville quadrangle, Piscataquis and Somerset Counties, Maine. *U. S. Geol. Surv. Bull.* 1241-F, 60 pp.
- EUGSTER, H. P., AND GEORGE B. SKIPPER (1967) Igneous and metamorphic reactions involving gas equilibria. In Philip H. Abelson, (Ed.) *Researches in Geochemistry*. Vol. 2, John Wiley and Sons, Inc., New York, p. 492-520.
- FINGER, LARRY W. (1967) *The Crystal Chemistry of Ferrromagnesian Amphiboles*. Ph.D. thesis, Univ. Minnesota, 73 pp.
- GILETTI, B. J. (1966) Isotopic ages from southwestern Montana. *J. Geophys. Res.* 71, 4029-4036.
- HEINRICH, E. WM. (1960) Pre-Beltian geology of the Cherry Creek and Ruby Mountain areas, southwestern Montana. *Mont. Bur. Mines Geol. Mem.* 38, pt. II., p. 15-40.
- JAMES, H. L., AND K. L. WIER (1961) Carter Creek iron deposit, Madison and Beaverhead Counties, Montana. *U. S. Geol. Surv. Open File Rep.*, 4 sheets.
- JAFFE, H. W., P. ROBINSON, AND C. KLEIN, JR., (1968) Exsolution lamellae and optic orientation of clinoamphiboles. *Science* 160, 776-778.
- KLEIN, CORNELIS, JR. (1964) Cummingtonite-grunerite series: a chemical, optical, and x-ray study. *Amer. Mineral.* 49, 963-982.
- (1966) Mineralogy and petrology of the metamorphosed Wabush Iron Formation, southwestern Labrador. *J. Petrology* 7, 246-305.
- (1968) Coexisting amphiboles. *J. Petrology* 9, 281-330.
- PAPIKE, J. J., M. ROSS, AND JOAN R. CLARK (1969) Crystal-chemical, optical, and X-ray study. *Amer. Mineral.* 49, 963-structure refinements. *Mineral. Soc. Amer. Spec. Pap.* 2, 117-136.
- RABBITT, J. C. (1948) A new study of the anthophyllite series. *Amer. Mineral.* 33, 263-323.
- ROBINSON, P. (1963) *Gneiss Domes of the Orange Area, Massachusetts and New Hampshire*. Ph.D. thesis, Harvard Univ., 253 pp.
- (1967) Gneiss domes and recumbent folds of the Orange area, west central Massachusetts. In P. Robinson, (Ed.) *Guidebook for Field Trips in the Connecticut Valley of Massachusetts*, Intercollegiate Geol. Conf., Amherst, Mass., p. 17-47.
- , AND HOWARD JAFFE (1969) Chemographic exploration of amphibole assemblages from central Massachusetts and southwestern New Hampshire. *Mineral. Soc. Amer. Spec. Pap.* 2, 251-274.
- , —, C. KLEIN, JR., AND M. ROSS (1969) Equilibrium coexistence of three amphiboles. *Contrib. Mineral. Petrology*, (in press).
- ROSS, M., J. J. PAPIKE, AND P. W. WEIBLEN (1968) Exsolution in clinoamphiboles. *Science* 159, 1099-1104.
- , W. L. SMITH, AND W. H. ASHTON (1968) Triclinic talc and associated amphiboles from Gouverneur Mining District, New York. *Amer. Mineral.* 53, 751-769.
- SCHMITT, H. H. (1964) *Petrology and Structure of the Eiksundal Eclogite Complex, Hareidland, Sunnmøre, Norway*. Ph.D. thesis, Harvard Univ.
- SCHWARTZ, G. M. (1931) Textures due to unmixing of solid solutions. *Econ. Geol.* 26, 739-763.
- SEGELER, C. G. (1961) First U. S. occurrence of manganoan cummingtonite, tirodite. *Amer. Mineral.* 46, 637-641.
- SEITSAART, J. (1952) On association of cummingtonite and hornblende. *Ann. Acad. Scient. Fennicae, Ser. A, III, Geologica-Geographica*, 30, 5-20.
- STOUT, J. H. (1969) An electron microprobe study of coexisting orthorhombic amphiboles (abstr.). *Trans. Amer. Geophys. Union* 50, 359.
- TOULMIN, PRIESTLEY, III (1964) Bedrock geology of the Salem quadrangle and vicinity, Massachusetts. *U. S. Geol. Surv. Bull.* 1163-A, 79 pp.
- VERNON, R. H. (1962) Co-existing cummingtonite and hornblende in an amphibolite from Duchess, Queensland, Australia. *Amer. Mineral.* 47, 360-370.
- YUND, R. A., AND R. H. MCCALLISTER (1968) The kinetics and mechanisms of exsolution. *Geol. Soc. Amer., Prog. 1968 Ann. Meet., Mexico City*.