An Experimental Study of Actinolite-Cummingtonite Phase Relations with Notes on the Synthesis of Fe-rich Anthophyllite

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Abstract

An amphibole quadrilateral, analogous to the well-known pyroxene quadrilateral, is defined by the end member compositions CaMg₅Si₂₄O₇(OH)₂ (tremolite), CaFe₅Si₂₄O₇(OH)₂ (ferrotremolite), Mg₅Si₂₄O₇(OH)₂ (anthophyllite), and Fe₅Si₂₄O₇(OH)₂ (grunerite). Phase relations along the join Mg₅Fe₅Si₂₄O₇(OH)₂-Ca₅Mg₅Fe₅Si₂₄O₇(OH)₂ (grunerite) through the center of the quadrilateral, were studied at T = 2 kbar and at oxygen fugacities defined by the FMQ buffer. Compositions along this join are expressed in mole percent of the calcic end member, actinolite (e.g., Act). It has not been possible to bracket phase boundaries with reversals because of the reluctance of new phases to nucleate in crystalline starting materials.

Three amphiboles with intermediate Fe : Mg ratios—anthophyllite, cummingtonite, and actinolite—were synthesized. Cummingtonite and actinolite are separated by a solvus that is asymmetric with a very steep cummingtonite limb near Act₈ and an actinolite limb that is measurably temperature dependent (Act₈ at 500°C, Act₆ at 600°C). The maximum solubility of the Ca-free component in the actinolite is reached at ~600°C. Actinolites with Fe : Mg ≈ 1, if coexisting with cummingtonites, are useful geothermometers and may be used to estimate, or put lower limits on, temperatures of metamorphism with a minimum uncertainty of ±35°C.

Actinolite decomposes by the reaction, actinolite → cummingtonite + clinopyroxene + quartz + H₂O. The products of this reaction differ from those reported for the decomposition of tremolite (Boyd, 1959) and ferrotremolite (Ernst, 1966).

Anthophyllite is the only amphibole which has been synthesized in Ca-free runs (i.e., Act₀). The presence of Ca facilitates nucleation of cummingtonite, and the assemblage anthophyllite + cummingtonite grows at the expense of less stable assemblages between about Act₁ and Act₅. Cummingtonite alone is present at ~Act₁₀. Some experiments suggest that the absence of cummingtonite in Ca-free runs is only a nucleation problem, but the phase relations between the two Ca-poor amphiboles have not been resolved satisfactorily.

Introduction

Coexisting amphiboles have been reported from many metamorphic terranes, and much interest has developed concerning their compositions and phase relations (e.g., Klein, 1968; Robinson and Jaffe, 1969; Ross, Papike, and Shaw, 1969; Robinson, Ross, and Jaffe, 1971; Stout, 1971). The compositions of many of these coexisting amphiboles lie in or near the system CaO-MgO-FeO-SiO₂-H₂O and can be depicted in an amphibole quadrilateral (Fig. 1) analogous to the familiar pyroxene quadrilateral. The three series of amphiboles that can be represented in this quadrilateral are (1) the Ca-rich monoclinic amphiboles tremolite-ferrotremolite, (2) the Ca-poor monoclinic amphiboles cummingtonite-grunerite, and (3) the Ca-poor orthorhombic amphiboles anthophyllite-ferroanthophyllite. The complete series tremolite-ferrotremolite is known from nature, and the two end members have been synthesized (Boyd, 1959; Ernst, 1966). Both series of Ca-poor amphiboles are stable at intermediate Fe: Mg, but neither pure-Fe ferroanthophyllite nor pure-Mg cummingtonite has been synthesized or found in nature. The approximate compositional limits of the two Ca-poor series are shown in Figure 1. In the following discussion Ca-rich and Ca-poor monoclinic amphiboles with intermediate Fe: Mg are referred to as actinolite and cummingtonite, respectively. Anthophyllite is used as a general term for all orthorhombic amphiboles.

Coexisting Ca-rich and Ca-poor amphiboles are
Amphiboles in the system CaO-MgO-FeO-SiO$_2$-H$_2$O were chosen for this experimental study because they are relatively simple chemically and are well represented in natural assemblages. The objectives of this study were twofold: (1) to determine the amount of solution between coexisting Ca-rich and Ca-poor monoclinic amphiboles as a function of temperature, and (2) to elucidate the phase relations between cummingtonite and anthophyllite. A join with intermediate Fe:Mg was required in order to satisfy the latter objective, and the join Mg$_{5.4}$Fe$_{0.5}$Si$_6$O$_{22}$(OH)$_2$-Ca$_2$Mg$_{2.5}$Fe$_{2.5}$Si$_6$O$_{22}$(OH)$_2$ was chosen for study (Fig. 1). Compositions along this join are abbreviated henceforth as mole percentages of the calcic end member actinolite (e.g., Act$_{50}$). The actual compositions of all amphiboles in multiphase assemblages will lie slightly off this join because Fe and Mg are not distributed equally between coexisting phases.

There have been several other experimental studies of phases or joins in this amphibole quadrilateral. Greenwood (1963), Boyd (1959), and Ernst (1966) determined the pressure-temperature stabilities of pure-Mg anthophyllite, tremolite, and ferrotremolite, respectively. Hinrichsen (1967), Schürrmann (1967, 1968), and Hellner and Schürrmann (1966) investigated the lower thermal stabilities, at $P_{\text{total}} = 1000$ bars, of the series anthophyllite-ferroanthophyllite, cummingtonite-grunerite, and tremolite-ferrotremolite, respectively. The bulk compositions of Hinrichsen’s study were Ca-free, whereas those of Schürrmann’s (1967, 1968) contained 2.7 wt percent CaO. Popp, Gilbert, and Craig (1974) report cell parameters of synthetic amphiboles on the join Mg$_{5.4}$Si$_6$O$_{22}$(OH)$_2$-Fe$_{5.4}$Si$_6$O$_{22}$(OH)$_2$.

**Experimental Techniques**

**Apparatus**

Hydrothermal experiments were conducted with conventional “cold seal” equipment (Tuttle, 1949) at Virginia Polytechnic Institute and State University. Experiments employed both vertically and horizontally mounted furnaces. Temperatures were measured with bare-wire chromel-alumel thermocouples inserted in the end wall of the pressure vessels, read with a Leeds and Northrup Type K-4 potentiometer, and monitored with an Esterline Angus strip chart recorder. New thermocouple junctions were made before each run. Solid-state temperature controllers (Hadjidakios, 1969) were used with the horizontal furnaces and temperature fluctuations were generally no
more than \( \pm 1^\circ \text{C} \). West Instrument Corporation Model JPLT3 controllers were used with the vertically mounted Tem-Pres furnaces, and fluctuations were on the order of \( \pm 2^\circ \) to \( 3^\circ \text{C} \). Several thermocouple-pressure vessel assemblies were calibrated at 1 atmosphere against the melting points of dried \( \text{NaCl} \) (800.4°C) and \( \text{CsCl} \) (646°C).

Pressure vessels were machined from the nickel-based alloy René 41, and a common pressure line connected them to a 5000 bar Heise pressure gauge. All runs were made at \( P_{\text{total}} = 2000 \text{ bars} \) (\( \pm 30 \text{ bars} \)), and were brought to temperature and quenched isobarically. Runs were quenched by removing the pressure vessels from the furnaces and passing a stream of compressed air over them for about a minute. The temperatures of the pressure vessels were usually 200°C or less, 15-20 minutes after they were removed from the furnaces. There was no optical or other evidence to suggest that any reaction took place during the quench.

Oxygen fugacities were controlled by the fayalite-magnetite-quartz (FMQ) buffer utilizing the double capsule technique described in Eugster and Wones (1962). Inner capsules were \( \text{Ag}_{70}\text{Pd}_{30} \); outer capsules were \( \text{Au} \). At the completion of each run the buffers were checked optically for the assemblage fayalite-magnetite-quartz-\( \text{H}_2\text{O} \).

**Starting Materials**

All charges were crystallized initially from either glass or an oxygen-balanced mix. These starting materials were prepared at compositional intervals of 10 mole percent actinolite along the join. Additional mixes were prepared on the compositions \( \text{Act}_{1.26}, \text{Act}_{1.5}, \text{Act}_{6}, \text{Act}_{16}, \text{Act}_{16}, \text{and } \text{Act}_{16} \). The mixes were composed of Materials Research Corporation \( \text{MgO} \); Johnson, Matthey and Co. grade I iron sponge; Fisher’s “Certified” reagent \( \text{CaCO}_3 \) and \( \text{Fe}_2\text{O}_3 \); and Corning 7940 silica glass. The components of the mix were ground together for one hour under acetone with an agate mortar and pestle. Glasses were prepared at the Geophysical Laboratory under the supervision of the late Dr. J. F. Schairer.

**Phase Identification and Characterization**

Run products were examined optically using a polarizing microscope and white light. Refractive indices were determined in oils calibrated at intervals of 0.002 units below 1.70 and 0.005 above 1.70.

Charges were scanned routinely at \( 2^\circ/\text{min} \) from 8° to 65° 2θ on a Norelco X-ray diffractometer equipped with a high intensity copper tube and a graphite monochromator. In addition, many charges were scanned at 1/8° per minute over critical 2θ ranges. Unit cell parameters were determined using data from 3 oscillations (6 scans) run at 1/4 degree per minute with a time constant of 4, and with a spinel (\( \text{MgAl}_2\text{O}_4 \)) internal standard \( (a = 8.083 \, \text{Å}) \) supplied by Professor G. V. Gibbs. The diffractometer tracings were at a scale of \( 1^\circ 2\theta = 1 \) inch, and the positions of the peak tips were measured to 0.005°.

The standard deviation of most peak positions was between 0.015° and 0.025°. Narrow, sharp peaks commonly had standard deviations of less than 0.010°, whereas the standard deviations of weak and/or irregular peaks were usually no larger than 0.035°. Unit cell parameters were refined using the computer program LSUCR (Evans, Appleman, and Handwerker, 1963). Copper \( K\alpha \) wavelength (1.54180) was used below 40° 2θ and \( K\alpha \) (1.54051) was used at higher angles. Reflections were weighted in the refinement by the factor \( 1/10000\sigma^2 \) which gave a weight of 1.0 to peaks with standard deviations of 0.010. Unit cell parameters calculated using weighted reflections were usually within the error of the parameters calculated giving all reflections unit weights. Weighting was used in all cell refinements, however, because it gave slightly smaller standard errors and fewer reflections were rejected in the refinement.

**Properties of Phases**

**Actinolite**

Actinolite forms small crystals which are generally less than 5 microns long and 0.5 microns wide. Neither the precise refractive indices nor the extinction angle can be determined because of the small size of the crystals. However, the oil of refractive index 1.660 is intermediate between the \( \alpha \) and \( \gamma \) indices of clumps of crystals in run 73B-1001.

The presence of actinolite is established only by examination of X-ray diffractometer powder patterns. The powder pattern of a charge containing \( \sim 90 \) percent actinolite—the remainder being cummingtonite, clinopyroxene, and quartz—is shown in Figure 2A. The actinolite peaks were indexed by comparison with the calculated powder pattern of tremolite (Borg and Smith, 1969, p. 264). Actinolite is distinguished from the Ca-poor amphiboles by the position of the 310 peak at 28.4° (cf, cummingtonite 310 and anthophyllite 610 both at about 29.1°). Reflections used to refine the cell parameters of actinolites are

\[^1\]The number following the hyphen in the run number is the bulk composition of the charge expressed as mole percent actinolite.
Reflections Used to Refine Unit Cell Parameters of Actinolite and Cummingtonite

<table>
<thead>
<tr>
<th>h &amp; k</th>
<th>Actinolite Run 73B-100</th>
<th>Cummingtonite Run 55D-10</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>( \theta_{\text{obs}} )</td>
<td>( \theta_{\text{cal}} )</td>
</tr>
<tr>
<td>0 4 0</td>
<td>19. 54 (5)</td>
<td>19. 52 (5)</td>
</tr>
<tr>
<td>1 3 1</td>
<td>22. 84 (5)</td>
<td>22. 83 (3)</td>
</tr>
<tr>
<td>1 3 1</td>
<td>26. 21 (5)</td>
<td>26. 21 (3)</td>
</tr>
<tr>
<td>2 0 0</td>
<td>27. 04 (5)</td>
<td>27. 05 (3)</td>
</tr>
<tr>
<td>3 1 0</td>
<td>28. 38 (5)</td>
<td>28. 40 (3)</td>
</tr>
<tr>
<td>0 6 1</td>
<td>34. 37 (5)</td>
<td>34. 37 (3)</td>
</tr>
<tr>
<td>2 0 2</td>
<td>35. 30 (5)</td>
<td>35. 31 (3)</td>
</tr>
<tr>
<td>2 6 1</td>
<td>36. 69 (5)</td>
<td>36. 66 (3)</td>
</tr>
<tr>
<td>2 0 2</td>
<td>44. 11 (5)</td>
<td>44. 09 (3)</td>
</tr>
<tr>
<td>0 6 1</td>
<td>54. 39 (5)</td>
<td>54. 36 (3)</td>
</tr>
</tbody>
</table>

CuK\( \alpha \) radiation below 40°\( \theta \); CuK\( \alpha \), above 40°

parameters for such an amphibole derived from those of the synthetic end-members tremolite-ferrotremolite\(^2\) (Ernst, 1968, p. 13) assuming a linear relationship between the cell parameters and Fe content.

### Cummingtonite

Cummingtonite crystals are generally somewhat larger than those of actinolite, and they reach a maximum size of \( \sim 15 \times 3 \) microns. Nevertheless, when both amphiboles are present, they cannot be distinguished optically with confidence. The extinction angle, \( Z \perp c \), of cummingtonite (1631-10) is 15°, and the refractive indices are \( \alpha : 1.650(3) \) and \( \beta : 1.668(3) \).

The X-ray powder pattern of cummingtonite

\(^2\)The values given in Ernst (1968, p. 13) for the \( a \) axis, \( d_{100} \) and \( V \) of ferrotremolite should be 9.974, 9.65, and 939 \( \text{Å} \) respectively (Ernst, 1966, p. 43).

\(^3\)The uncertainty in the refractive index is given in parentheses and refers to the last digit.

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**Table 2. Unit Cell Parameters**

<table>
<thead>
<tr>
<th></th>
<th>( a(\lambda) )</th>
<th>( b(\lambda) )</th>
<th>( c(\lambda) )</th>
<th>( \beta (\degree) )</th>
<th>( V(\lambda^3) )</th>
<th>( d_{100}(\lambda) )</th>
</tr>
</thead>
<tbody>
<tr>
<td>Actinolite(^b)</td>
<td>9.892 (5) ( * )</td>
<td>18.191 (12)</td>
<td>5.295 (3)</td>
<td>104.65 (5)</td>
<td>921.9 (7)</td>
<td>9.572 (5)</td>
</tr>
<tr>
<td>Actinolite (^d)</td>
<td>9.903</td>
<td>18.200</td>
<td>5.285</td>
<td>106.51</td>
<td>922.0</td>
<td>9.582</td>
</tr>
<tr>
<td>Cummingtonite (^e)</td>
<td>9.544 (5)</td>
<td>18.171 (8)</td>
<td>5.316 (3)</td>
<td>102.33 (5)</td>
<td>900.8 (6)</td>
<td>9.326 (5)</td>
</tr>
<tr>
<td>Cummingtonite (^d)</td>
<td>9.526 (9)</td>
<td>18.190 (13)</td>
<td>5.320 (5)</td>
<td>102.04 (5)</td>
<td>901.5 (1)</td>
<td>9.316 (0)</td>
</tr>
<tr>
<td>Anthophyllite (^f)</td>
<td>18.622 (12)</td>
<td>18.185 (21)</td>
<td>5.309 (5)</td>
<td>90.0</td>
<td>1797.7 (21)</td>
<td>9.311 **</td>
</tr>
<tr>
<td>Pure-Mg Anthophyllite (^f)</td>
<td>18.608 (35)</td>
<td>17.964 (59)</td>
<td>5.303 (12)</td>
<td>90.0</td>
<td>1772.5 (62)</td>
<td>9.304 **</td>
</tr>
<tr>
<td>Pure-Mg Anthophyllite (^g)</td>
<td>18.612 (6)</td>
<td>18.01 (6)</td>
<td>5.241 (4)</td>
<td>90.0</td>
<td>1756.2</td>
<td>9.305 **</td>
</tr>
</tbody>
</table>

\(^a\) Actinolite, 73B-100, 558 °C, 39 days
\(^b\) Actinolite, predicted - see text
\(^c\) Cummingtonite, 55D-10, 620 °C, 102 days
\(^d\) Cummingtonite, predicted (Klein & Waldbaum, 1967)
\(^e\) Anthophyllite, 30C-0, 675 °C, 44 days
\(^f\) Pure-Mg anthophyllite, new parameters, see Table 4.
\(^g\) Pure-Mg anthophyllite (Greenwood, 1963)

\(*\)Calculated standard errors are given in parentheses and refer to the last decimal place(s).

**\( d_{100} \)**

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**Fig. 2. X-ray diffractometer tracings of clinoamphiboles.** (A) 80 percent actinolite + cummingtonite + clinopyroxene + quartz; run 73B-100, 558 °C, 39 days. (B) actinolite + cummingtonite; run 46A-50, charge run at 3 different temperatures: 655 °C-46 days, 700 °C-14 days, 500 °C-13 days; total time, 73 days. (C) cummingtonite, run 55D-10, 620 °C, 102 days. Abbreviations: A-actinolite, C-cummingtonite, Cp-clinopyroxene, Q-quartz, M-multiple reflection. Instrument settings: CuK\( \alpha \) radiation; 40 kV; 20 mA; 200 cps; time constant, 16; scan speed, 1/8 degree \( \theta \) per minute; chart speed, 1/16 an inch per minute.

Given in Table 1. Where clinopyroxene plus other products exceeded 25 percent, the actinolite 202 and 202 reflections were not used because of the interference of strong clinopyroxene peaks. Cell parameters of an actinolite believed to be on or very near the composition \( \text{Ca}_2 \text{Mg}_{8.5} \text{Fe}_{2.5} \text{Si}_{10.5} \text{OH}_2 \) are given in Table 2. Also included are the cell parameters for such an amphibole derived from those of the synthetic end-members tremolite-ferrotremolite\(^2\) (Ernst, 1968, p. 13) assuming a linear relationship between the cell parameters and Fe content.
(Figures 2C and 3A) was indexed by comparison with the calculated powder pattern of cummingtonite in Borg and Smith (1969, p. 372). Cummingtonite is distinguished from actinolite by its 310 peak at 29.1°2θ (Fig. 2B), and from anthophyllite by its 131 peak at 26.0°2θ (cf. Figs. 3A, 3C). Cell parameters of cummingtonite, \( \text{Ca}_9\text{Mg}_3\text{Fe}_3\text{Si}_6\text{O}_{22}(\text{OH})_2 \), are given in Table 2 along with those calculated using the equations of Klein and Waldbaum (1967). The reflections used to determine the cell parameters are listed in Table 1.

**Estimation of the Compositions of Clinoamphiboles in Multiphase Assemblages**

Figure 2B shows the powder pattern of a charge that contains nearly 100 percent amphibole and is composed of subequal amounts of actinolite and cummingtonite. Six relatively strong clinoamphibole peaks—131, 240, 310, 202, 202, and 661—show measurable separation (see Table 1 for approximate separations). Where cummingtonite and actinolite are present in subequal amounts, a unique 2θ can be measured for each of the above peaks; thus it is possible to determine unit cell parameters of both amphiboles. No correction is made for overlap of peaks. The 040 peak, which shows no separation (Fig. 2B), is also measured and included in the refinement of both amphiboles. The inclusion of the 040 peak has little effect on the value of \( b \), but it does significantly reduce the standard error of \( b \).

The important chemical variables of the coexisting actinolite and cummingtonite are Ca content and Fe : Mg ratio. Both X-ray powder and electron microprobe methods have been used to estimate the compositions of the amphiboles in this study. Colville, Ernst, and Gilbert (1966, p. 1741) have shown that, within any given amphibole series, the \( b \) axis increases with replacement of Fe for Mg in the \( M(2) \) site. The \( b \) axis was chosen to estimate the Fe content of amphiboles in this study because relative to other cell parameters it shows a large change in length with varying Fe : Mg. Moreover, the first four entries in Table 2 show that, at the Fe : Mg of this study, variation in Ca has little effect on the \( b \) axes of the clinoamphiboles. Klein and Waldbaum (1967) have shown that in the series cummingtonite–grunerite there is a linear relationship between Fe content and the \( b \) cell dimension. Their determinative curve, which has a standard error of about ±2.5 mole percent of the Fe end-member, is used to estimate the Fe content of cummingtonites in this study.

A comparison of the \( b \) cell dimensions of tremolite (18.054 Å, Ernst, 1968, p. 13), actinolite with Fe : Mg = 1 (18.191 Å, Table 2), and the average value of \( b \) for ferrotremolite (18.34 Å, Ernst, 1966) suggests that in the series tremolite–ferrotremolite there is also a linear relationship between Fe content and \( b \), and such a determinative curve was used in this study. Ernst reports a range in \( b \) from 18.31 to 18.39 Å for ferrotremolite; these values result in an uncertainty of ±5 mole percent of the Fe end-member at Fe : Mg = 1.

Calcium is confined to the \( M(4) \) site in amphiboles, and Whittaker (1960) and Gibbs (in Appleman et al., 1966) have shown that among the different
amphibole series the $\beta$ angle is related to the mean ionic radius of the cations in $M(4)$. In this study linear relations were tested between the cell parameters and Ca contents of three charges that had been converted largely to a single amphibole (cummingtonite, 55D-10, 99 percent; actinolite, 53D-80, 85 percent; and actinolite, 73B-100, 90 percent). The calculated $d_{100}$ ($a \sin \beta$) proved to be the best estimator of the Ca content of the charge and presumably of the amphibole; therefore, $d_{100}$ rather than the $\beta$ angle was used to estimate the Ca-content of the amphiboles.

A linear determinative curve for Ca vs $d_{100}$ was constructed using the data from 55D-10 and 73B-100. The calcium content of the amphiboles is expressed as mole percent actinolite Car($Mg,Fe$)$_{6}$Si$_{6}$O$_{22}$($OH$)$_{8}$ with an unspecified $Fe : Mg$. The $d_{100}$ standard error of most amphiboles in this study is $<0.01$ which introduces an uncertainty of $\pm 4$ mole percent actinolite assuming that there is no error in the determinative curve.

The amphiboles synthesized in this study are too small for conventional electron microprobe analysis. Nevertheless it is possible to make partial analyses of such material using an intensity ratio method recently discussed in detail by Eugster et al (1972). Analyses in this study were made at the State University of New York at Stony Brook using an ARL-EMX-5M electron microprobe. Intensity data were collected simultaneously for Ca, Fe, Mg, and Si. Calibration curves, calculated using the correction procedure of Bence and Albee (1968), were used to relate the observed intensity ratios—Ca/Si, Fe/Si, and Mg/Si—to the Ca contents and Fe-Mg ratios of the synthetic amphiboles.

**Anthophyllite**

Anthophyllite crystals average 5-10 microns long and 1-2 microns wide, but crystals up to 20 microns in length have been observed. Refractive indices of anthophyllite—$\alpha = 1.650(3), \gamma = 1.666(3)$—are identical, within error, to those of cummingtonite. Anthophyllite crystals, being orthorhombic, have parallel extinction in many orientations, but they can have inclined extinction when planes of the type $(hkl)$ are parallel to the microscope stage (Johannsen, 1937, p. 212).

The powder pattern of anthophyllite (Fig. 3C) was indexed by comparison with that calculated for anthophyllite by Borg and Smith (1969). The calculated patterns (Borg and Smith, 1969) suggest that some of Greenwood's anthophyllite peaks are incorrectly indexed. These were reindexed (Table 3) and new cell parameters were calculated. Except for $c$, the parameters are identical, within error, to those reported by Greenwood (Table 2).

**Other Phases**

Orthopyroxene forms bladed crystals averaging 30$\mu$ long and is readily detected in X-ray scans by the presence of the 420 peak at 27.9° 2$\theta$. Clinopyroxene, present as small equant crystals about 5-10$\mu$ in size, is identified in X-ray scans by its 211 peak at 29.7° 2$\theta$. The precise composition of the clinopyroxene is not known, but it always coexists with a less calcic phase such as actinolite, cummingtonite, and/or orthopyroxene. Natural metamorphic clinopyroxenes that coexist with these phases contain 10-15 percent hypersthene in solid solution.

Talc, cristobalite, magnetite, olivine, and a carbonate mineral were crystallized from mixes at low temperatures. Cristobalite was found only in runs of
ACTINOLITE-CUMMINGTONITE PHASE RELATIONS

Table 3. Reflections Used to Refine Unit Cell Parameters of Anthophyllite

<table>
<thead>
<tr>
<th></th>
<th>Anthophyllite (this study)</th>
<th>Anthophyllite, Greenwood (1963)</th>
</tr>
</thead>
<tbody>
<tr>
<td>h k l</td>
<td>d(calc) Å</td>
<td>d(obs) Å</td>
</tr>
<tr>
<td>0 2 0</td>
<td>9.0922</td>
<td>9.0705</td>
</tr>
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</tr>
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</tr>
<tr>
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<td>2 3 1</td>
<td>3.6704</td>
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</tr>
<tr>
<td>0 5 3</td>
<td>1.5913</td>
<td>1.5918</td>
</tr>
</tbody>
</table>

*Calculated using indices shown for anthophyllite, this study

R Rejected in refinement

CuKα radiation below 40°28; CuKα, above 40°

less than two weeks duration, whereas longer runs produced quartz.

Phase Relations

Introduction

Phase relations along the join MgFe,SiO(OH) - CaMgFe,SiO(OH) are shown in Figure 4. Explanation of abbreviations used in the following figures, tables, and text are given in Table 4.

It has not been possible to bracket any field boundaries in Figure 4 with reversal runs because of the reluctance of new phases to nucleate in crystalline starting material. However, in many runs (triangles, Fig. 4) the starting assemblage was changed during the run by the loss of one or more phases, and in some cases these set either minimum or maximum limits for boundary curves.

The open symbols in Figure 4 represent synthesis data, because the final assemblage was not produced from an assemblage stable in an adjacent field. Most of these open symbols, however, indicate run series, that is, charges that have been rerun several times at the same temperature. It was necessary to rerun charges because mix and glass starting materials usually produce mixtures of amphiboles plus carbonate-free anhydrous phases when run between ~550-750°C. When the charges were rerun, the amphiboles usually grew at the expense of the anhydrous phases; therefore, most charges were rerun until the assemblage was changed by the loss of one or more anhydrous phases and/or until the proportion of phases stopped changing.

The letter in the run number (Table 5) indicates the number of times that the charge was rerun as close as possible to the same temperature; thus, A indicates that the charge was run only once, whereas D indicates that the charge was run four times. The assemblage listed under "Solid starting material" (Table 5) for run series (i.e., runs with a B or higher letter in the run number) is the product of the first run of the series. This is the starting assemblage for the following runs of the series. The temperatures shown in Table 5 are for the final run of the series. Unless otherwise indicated the uncertainty due to temperature fluctuations during runs, and temperature variation from run to run, are less than ±3°C.

The total percentages of amphibole(s) in both the starting and final assemblages (Table 5) are "eyeball" estimates made principally from X-ray scans; however, they were found to be reproducible to about ±10 percent at mid range, and to ±5 percent at the lower (10 percent) and upper (90 percent) extremes. Five percent of a phase generally indicates that it is barely, but definitely, detectable on the X-ray scan; one percent indicates that it is detectable only optically and then as scattered grains. No claim is made that these estimations are correct in an absolute sense, but they are useful in demonstrating the amount of reaction that took place.

Actinolite-Cummingtonite Phase Relations

The phase relations involving actinolite, cummingtonite, and their dehydration assemblages are shown in Figure 4. Reaction rates were more sluggish, and amphibole yields were generally lower, in Ca-rich charges than Ca-poor ones. Several charges less
calcic than Act$_{90}$ were converted to about 99 percent amphibole(s) in runs of 40 to 60 day durations (Table 5).

Cummingtonite appears to be the only amphibole present in charges with the bulk composition of Act$_{10}$ (Figs. 2C, 3A). These charges contain 1.27 wt percent CaO. A field of Act + Cum exists between about Act$_{10}$ and Act$_{90}$, with the actinolite limb of the solvus being more temperature-sensitive than the cummingtonite limb. The determination of the solvus is discussed in a later section. Because the join studied is not parallel to the slope of the tielines that connect coexisting actinolite and cummingtonite (Fig. 1), the amphiboles at any given temperature become more magnesium rich as the bulk compositions become less calcic. Consequently, it is possible for actinolites on the Ca-poor side of the diagram to have a higher thermal stability than those on the Ca-rich side; and as a result the upper boundary for the field Act+Cum rises to the Ca-poor side.

Most charges run in the field Act (Fig. 4) have produced relatively low yields of amphibole (Table 5). The maximum yield of actinolitic amphibole, estimated as ~90 percent (Fig. 2A; run 73B-100) was produced at 558°C using as starting material Tc+Cc+Crist+Mag, seeded with about 10 percent

---

**Table 4. Abbreviations Used in Tables, Text, and Following Figures**

<table>
<thead>
<tr>
<th>Abbreviation</th>
<th>Meaning</th>
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<tbody>
<tr>
<td>Act</td>
<td>actinolite</td>
</tr>
<tr>
<td>Anth</td>
<td>anthophyllite</td>
</tr>
<tr>
<td>Carb</td>
<td>carbonate (undifferentiated)</td>
</tr>
<tr>
<td>Cc</td>
<td>calcite</td>
</tr>
<tr>
<td>Cpx</td>
<td>clinopyroxene</td>
</tr>
<tr>
<td>Crist</td>
<td>cristobalite</td>
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<td>Cum</td>
<td>cummingtonite</td>
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<td>Ferrotre</td>
<td>ferro tremolite</td>
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<td>Grun</td>
<td>grunerite</td>
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<td>Hem</td>
<td>hematite</td>
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<tr>
<td>Hd</td>
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<td>Mag</td>
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<td>Ol</td>
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<tr>
<td>Opx</td>
<td>orthopyroxene</td>
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<td>Qtz</td>
<td>quartz</td>
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<td>Tc</td>
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<tr>
<td>Tre</td>
<td>tremolite</td>
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**Fig. 4.** Phase diagram for the pseudobinary join, Mg$_2$Fe$_2$Si$_2$O$_7$(OH)$_2$-Ca$_2$Mg$_2$Fe$_2$Si$_2$O$_7$(OH)$_2$ + excess H$_2$O, at $P_{\text{fluid}} = 2000$ bars and at oxygen fugacities defined by the FMQ buffer. All assemblages coexist with a vapor phase. Symbols: solid triangles, final assemblage produced from a starting assemblage stable in an adjacent field—the apex of the triangle pointing to the boundary separating the fields; open triangles that point up, Act+Cum+Cpx+Qtz produced by loss of Opx from the assemblage Act+Cum+Cpx; open triangles that point down, Act+Cum produced from an assemblage containing tale; open rectangles, all other synthesis runs. Approximate temperature uncertainty shown by height of symbol. Abbreviations are given in Table 4.
Act+Cpx+Qtz from a previous run. When the charge was rerun, however, the actinolite showed no signs of growth at the expense of the remaining 10 percent Cum+Cpx+Qtz. Similarly seeded runs using "oxide" mix and glass as starting material produced 70 and 30 percent actinolite respectively at ~$550^\circ\text{C}$.

It has not been possible to locate with accuracy the upper boundary of the field Act because of the metastable persistence of cummingtonite, clinopyroxene, and quartz at temperatures ($500^\circ-550^\circ\text{C}$) where actinolite alone is presumably stable and because actinolite, once crystallized, will not react to form its breakdown products except at relatively high temperatures ($700^\circ\text{C}$). A field of Act+Cpx+Qtz is believed to separate the fields of Act and Act+Cum+Cpx+Qtz, but runs within the boundaries of this field (Fig. 4) have all produced the three appropriate phases plus cummingtonite.

Using Figure 5 it can be easily shown that the field Act+Cpx+Qtz should separate the fields of Act and Act+Cum+Cpx+Qtz. In this schematic isothermal-isobaric section the assemblage Act+Cpx+Qtz is believed to separate the fields of Act and Act+Cum+Cpx+Qtz. In this schematic isothermal-isobaric section the assemblage Act+Cpx+Qtz is believed to separate the fields of Act and Act+Cum+Cpx+Qtz. In this schematic isothermal-isobaric section the assemblage Act+Cpx+Qtz is believed to separate the fields of Act and Act+Cum+Cpx+Qtz. In this schematic isothermal-isobaric section the assemblage Act+Cpx+Qtz is believed to separate the fields of Act and Act+Cum+Cpx+Qtz. In this schematic isothermal-isobaric section the assemblage Act+Cpx+Qtz is believed to separate the fields of Act and Act+Cum+Cpx+Qtz.

### Table 5: Run Data for the Join, $\text{Mg}_8\text{Fe}_2\text{Si}_2\text{O}_8(\text{OH})_2-\text{Ca}_3\text{Mg}_8\text{Fe}_{5+}\text{Si}_2\text{O}_8(\text{OH})_2$

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<th>Run no. *</th>
<th>Solid starting material</th>
<th>Temp. (°C)</th>
<th>Total Duration (days)</th>
<th>Percentage amphiboles start final</th>
<th>Final crystalline assemblage **</th>
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<td>19</td>
<td>80 98</td>
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Field: Act+Cum+Cpx+Qtz

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<th>Temp. (°C)</th>
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<th>Percentage amphiboles start final</th>
<th>Final crystalline assemblage **</th>
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<td>40</td>
<td>95 99</td>
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<td>65</td>
<td>85 95</td>
<td>Act+Cum+(Cpx+Qtz)</td>
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Field: Act+Cpx+Qtz

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<td>60 85</td>
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Field: Act+Cum+Cpx+Qtz

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<td>Act+Cpx+Qtz+5% (Cum+10pox)</td>
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Field: Act+Cum+Cpx+Qtz

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<td>47</td>
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* See Table 4 for abbreviations.
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</tr>
<tr>
<td>95B-5</td>
<td>?Anth+Cum+O1Qtz</td>
<td>698</td>
<td>30</td>
<td>95 99</td>
<td>Anth+?Cum+O1Qtz</td>
</tr>
</tbody>
</table>

**Anthophyllite data**

<table>
<thead>
<tr>
<th>Anthophyllite data</th>
</tr>
</thead>
<tbody>
<tr>
<td>183A-0</td>
</tr>
<tr>
<td>174A-0</td>
</tr>
<tr>
<td>175B-0</td>
</tr>
<tr>
<td>170A-0</td>
</tr>
<tr>
<td>118C-1.25</td>
</tr>
<tr>
<td>102G-2.5</td>
</tr>
<tr>
<td>109B-2.5</td>
</tr>
</tbody>
</table>

*The number following the dash is the Ca content of the charge expressed as mole percent actinolite.
**Phase(s) in parentheses interpreted as metastable.
*Phase may be present.
Amphibole which showed significantly more growth.
#Temperature variation ± 3°C to ± 5°C; otherwise less than ± 3°C.

+Qtz(+H₂O) is invariant, and is projected as a triangle. This triangle can only touch the field Act at one point; hence the composition of the actinolite is fixed, and, as discussed in a following section, this composition has been determined to be about Act₈₆. Therefore, some third field must separate the fields Act and Act+Cpx+Qtz along the join Ca₄Mg₆Si₆O₂₂(OH)₂-Ca₂Fe₂Si₂O₅ in Figure 5, and the field Act+Cpx+Qtz appears to be the most reasonable choice. At Pfluid = 2000 bars and fO₂ defined by the FMQ buffer, actinolite of the composition Ca₄Fe₂Mg₆Si₆O₂₂(OH)₂ begins to decompose by reaction to clinopyroxene, quartz, and a less calcic actinolite (i.e., enriched in the cummingtonite component). The least calcic composition that amphibole can maintain under these conditions is about 85 mole percent actinolite, which is reached at ~600°C. Above this temperature the actinolitic amphibole begins reacting to Cum+Cpx+Qtz, the ultimate breakdown assemblage of the actinolite. The decom-
position products of actinolite differ from those found experimentally by Boyd (1959, p. 383) for tremolite (Cpx+Opx+Qtz+H₂O), and by Ernst (1966, p. 57) for ferrotremolite (Cpx+Fa+Mag+Qtz+H₂O) using the FMQ buffer.

The upper boundary of the field Act+Cum+Cpx+Qtz is drawn where the presence of actinolite can no longer be definitely established on X-ray scans. Attempts to produce this assemblage from the assemblages Act+Cum, Act+Cpx+Qtz, and Cpx+Opx+Qtz were unsuccessful. On the bulk composition Act₁₀₀, cummingtonite—but not actinolite—nucleated and grew from Cpx+Opx+Qtz at 671°C.

The lower boundary of the field Cum+Cpx+Opx+Qtz is placed just above those runs in which orthopyroxene disappeared. All runs in this field are synthesis data; however, most have been rerun and showed either some growth of cummingtonite or no change in phase proportions.

The lower boundary of the field Cpx+Opx+Qtz is placed immediately below runs which produced no amphiboles on synthesis from mixes.

The lower thermal stabilities of the amphiboles were not studied in detail, but amphiboles were observed to grow at the expense of low temperature assemblages. A glass starting material of composition Act₁₀₀ produced Act+Cpx+Qtz at 500°C (51B-100). In contrast, mixes from Act₁₀₀ to Act₁₀ initially produced Tc+Cc+Mag+Crist+Ol+traces of amphibole(s). When the charges were rerun, the amphiboles grew and some long runs (≥80 days) produced high yields of amphiboles (e.g., 16E-50, 92D-90, Table 5).

Cummingtonite, in a charge with the bulk composition of Act₁₀, nucleated and grew from Tc+Ol+Cc+Mag+Crist at about 500°C. After 111 days the amphibole comprised 50 percent of the charge and was still growing. Because equilibrium was not reached, the final stable assemblage is not known.

The Actinolite-Cummingtonite Solvus

X-ray Data. It was argued in a preceding section that the Ca contents and Fe:Mg ratios of the clino-
amphiboles can be estimated from their $d_{hkl}$ spacings and $b$ cell dimensions, respectively. Cell parameters and estimated compositions are given in Table 6, and the estimated Ca contents of clinoamphiboles in multiphase assemblages are shown in Figure 6.

The Ca content of actinolite in the field Act+Cum decreases regularly from about 96 mole percent actinolite at 500°C to about 85 mole percent at 600°C. A straight line appears to give the best fit to the present data on the actinolite limb of the solvus, but the solvus should be somewhat concave toward the Ca-poor side. In the field, Act+Cum+Cpx+Qtz, the Ca content of the actinolite appears to remain constant at about Act$\text{Ca}_{10}$ (Fig. 6).

The Ca-poor limb of the solvus is much steeper than the Ca-rich limb. Within the calculated error on $d_{hkl}$, the Ca-poor limb could be vertical at about 10 mole percent actinolite.

The Fe-Mg contents of the amphiboles shown in Table 6 are in the general range expected, because all charges have equal mole proportions of FeO and MgO. The uncertainties are relatively large on these estimates; nevertheless cummingtonite is invariably enriched in Fe relative to coexisting actinolite which is consistent with natural assemblages (Fig. 1).

**Electron Microprobe Data.** To test the technique for analyzing fine powders described by Eugster et al. (1972), amphiboles of known compositions were analyzed first. Actinolite grains from 73B-100 and cummingtonite grains from 148C-10 are from charges that had been converted largely to a single phase (Table 5); their compositions, therefore, should be that of their charges. The average compositions of the analyzed grains, shown in Table 7, agree quite well with the known compositions of the charges.

The success of analyzing individual phases from multiphase assemblages depends upon being able to disaggregate and disperse the sample. The problem of sample aggregation plagued attempts to analyze individual amphiboles from two-amphibole assemblages. The synthetic amphiboles have an almost acicular habit, and they form mats which usually are impossible to disaggregate. Samples which clearly contained two amphiboles (e.g., 17D-50) yielded only analyses that were near the composition of the charge. Only in sample 13B-50 was it possible to obtain analyses of two distinct populations of amphiboles (Table 7). The Ca contents, especially that of the actinolite, are in good agreement with those expected from the X-ray powder study (Fig. 6); thus the electron microprobe analyses support the credibility of the compositions determined from X-ray data.

**Table 6. Unit Cell Parameters and Estimated Compositions**

<table>
<thead>
<tr>
<th>Run no</th>
<th>Temp. ('C)</th>
<th>Assemblage</th>
<th>$a$(Å)</th>
<th>$b$(Å)</th>
<th>$c$(Å)</th>
<th>$\beta(\degree)$</th>
<th>$V$(Å$^3$)</th>
<th>$d_{100}$ (Å)</th>
<th>mole* percent actinolite</th>
<th>100xFeO</th>
<th>FeO+MgO</th>
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</thead>
<tbody>
<tr>
<td>95B-5</td>
<td>698</td>
<td>Anth+Cum</td>
<td>9.528(3)</td>
<td>18.180(10)</td>
<td>5.316(2)</td>
<td>102.08(4)</td>
<td>900.4(5)</td>
<td>9.316</td>
<td>7</td>
<td>48(2)</td>
<td></td>
</tr>
<tr>
<td>878-25</td>
<td>652</td>
<td>Act+Cum</td>
<td>9.545(6)</td>
<td>18.201(12)</td>
<td>5.314(5)</td>
<td>102.28(8)</td>
<td>902.1(8)</td>
<td>9.326</td>
<td>10</td>
<td>52(2)</td>
<td></td>
</tr>
<tr>
<td>40C-60</td>
<td>699</td>
<td>Act+Cum+Cpx+Qtz</td>
<td>9.533(7)</td>
<td>18.211(15)</td>
<td>5.315(5)</td>
<td>102.44(8)</td>
<td>903.0(9)</td>
<td>9.329</td>
<td>12</td>
<td>54(3)</td>
<td></td>
</tr>
<tr>
<td>168-50</td>
<td>505</td>
<td>Act+Cum</td>
<td>9.533(7)</td>
<td>18.189(25)</td>
<td>5.317(5)</td>
<td>102.25(9)</td>
<td>901.0(13)</td>
<td>9.316</td>
<td>7.5</td>
<td>50(5)</td>
<td></td>
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<tr>
<td>170-50</td>
<td>550</td>
<td>Act+Cum</td>
<td>9.536(7)</td>
<td>18.191(13)</td>
<td>5.312(4)</td>
<td>102.26(6)</td>
<td>900.4(8)</td>
<td>9.318</td>
<td>7.5</td>
<td>50(2)</td>
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</tr>
<tr>
<td>18C-50</td>
<td>582</td>
<td>Act+Cum</td>
<td>9.546(5)</td>
<td>18.165(22)</td>
<td>5.313(3)</td>
<td>102.30(4)</td>
<td>899.9(11)</td>
<td>9.325</td>
<td>10</td>
<td>45(4)</td>
<td></td>
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<tr>
<td>15B-50</td>
<td>603</td>
<td>Act+Cum</td>
<td>9.551(8)</td>
<td>18.195(29)</td>
<td>5.317(4)</td>
<td>102.35(7)</td>
<td>902.7(13)</td>
<td>9.330</td>
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<td>51(6)</td>
<td></td>
</tr>
<tr>
<td>43C-50</td>
<td>665</td>
<td>Act+Cum</td>
<td>9.553(7)</td>
<td>18.198(21)</td>
<td>5.315(3)</td>
<td>102.45(7)</td>
<td>902.5(10)</td>
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<td>51(4)</td>
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</tr>
<tr>
<td>55D-10</td>
<td>620</td>
<td>Cum</td>
<td>9.540(5)</td>
<td>18.171(8)</td>
<td>5.316(3)</td>
<td>102.33(5)</td>
<td>900.8(6)</td>
<td>9.324</td>
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<td>47(2)</td>
<td></td>
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<table>
<thead>
<tr>
<th>Actinolite</th>
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<tr>
<td>16E-50</td>
</tr>
<tr>
<td>17D-50</td>
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<tr>
<td>18E-50</td>
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<tr>
<td>15B-50</td>
</tr>
<tr>
<td>43C-50</td>
</tr>
<tr>
<td>38C-80</td>
</tr>
<tr>
<td>53D-80</td>
</tr>
<tr>
<td>92D-90</td>
</tr>
<tr>
<td>42E-90</td>
</tr>
<tr>
<td>61B-100</td>
</tr>
<tr>
<td>73B-100</td>
</tr>
</tbody>
</table>

*Uncertainty ± 4 mole percent
Cummingtonite analyses from 128B-20 are similar to those from 13B-50 (Table 7). However, no analyses of actinolite were obtained from 128B-20, and the phases may not have been completely separated.

**Anthophyllite Stability and Anthophyllite-Cummingtonite Relations**

Anthophyllite is the only amphibole which has been synthesized on the bulk composition Act₁. There is no optical or X-ray evidence that a monoclinic amphibole is present in any Ca-free runs.

Greenwood (1963) determined that pure-Mg anthophyllite decomposes to enstatite, quartz, and H₂O at ~765°C at P₃H₀ = 2 kbar. In this study the maximum thermal stability of anthophyllite, Mg₃.₅Fe₃.₅Si₂O₉(OH)₃, has been bracketed between 715°C and 742°C (Table 5). As temperature increases, the amphibole reacts to form a more Mg-rich anthophyllite, orthopyroxene, quartz, and H₂O. The boundary between the assemblages Anth+Opx+Qtz and Opx+Qtz has been bracketed between 750°C and 782°C (Table 5). Even the minimum temperature of this bracket seems surprisingly high when compared to Greenwood's data.

Cummingtonite together with anthophyllite crystallizes from unseeded mixes at Act₁₂₅ (about 0.2 wt percent CaO). Charges at Act₁₂₅, Act₁₅, and Act₅

TABLE 7. Electron Microprobe Analyses of Synthetic Amphiboles

<table>
<thead>
<tr>
<th>Run no.*</th>
<th>Assemblage Phase</th>
<th>Number of Analyses</th>
<th>Mean Ca content**</th>
<th>Mean 100FeO/(Fe⁺MgO)</th>
</tr>
</thead>
<tbody>
<tr>
<td>738-100</td>
<td>Act Act</td>
<td>10</td>
<td>99(6)***</td>
<td>53(7)</td>
</tr>
<tr>
<td>148C-10</td>
<td>Cm Cm</td>
<td>6</td>
<td>11(2)</td>
<td>54(3)</td>
</tr>
<tr>
<td>138-50</td>
<td>Act+Cm Act Act</td>
<td>7</td>
<td>82(4)</td>
<td>50(6)</td>
</tr>
<tr>
<td>138-50</td>
<td>Act+Cm Act+Cm Cm</td>
<td>4</td>
<td>16(4)</td>
<td>55(3)</td>
</tr>
<tr>
<td>128B-20</td>
<td>Act+Cm Act+Cm Cm</td>
<td>12</td>
<td>16(8)</td>
<td>54(7)</td>
</tr>
</tbody>
</table>

*The number following the dash is the Ca content of the charge expressed as mole percent actinolite.
**Ca content expressed as mole percent actinolite.
***One standard deviation given in parentheses.
These experiments were made at the State University of New York at Stony Brook.

Discussion of Natural Assemblages

The compositions of natural actinolites (with Fe : Mg near 1) that coexist with cummingtonites can be used to estimate or to put lower limits on metamorphic temperatures, if it is assumed that the actinolite limb of the solvus (Fig. 6) is not signifi-

cantly affected by the following: (1) changes in \( P_{\text{total}} \), (2) Fe : Mg ratios slightly different from those of this study, and (3) small amounts of Na, Mn, Al, Fe\(^{3+}\), etc, which are present in most natural actinolites. The estimated uncertainty of ±4 mole percent actinolite in the compositions used to determine the solvus results in a minimum uncertainty of ±35°C in the temperature of equilibration. The temperature uncertainty increases as the natural amphiboles depart from the compositions of this study and as the physical conditions of metamorphism depart from the experimental conditions.

Wabush Iron Formation, Canada

The petrology of the metamorphosed Precambrian Wabush Iron Formation in Labrador and Quebec has been investigated in unusual detail by Mueller (1960), Kranck (1961), Klein (1966), and Butler (1969). The mineral assemblages from the Wabush Iron Formation that are relevant to this study are summarized in Table 8.

Klein found coexisting Act<sup>+</sup>Cum(*Cpxf Mag* Carb) at one locality near Wabush Lake (analyses in Klein, 1968, no. 4-6). The calcic amphibole has a composition of 91 mole percent actinolite, which suggests a temperature of equilibration of -550°C. Based on the melting curve of granite, Klein (1966, p. 29) estimated the maximum temperature of metamorphism to be “below about 600°C.” The metamorphism is of the kyanite-staurolite zone, and the presence of kyanite at 550-600°C suggest \( P_{\text{total}} \) ≥ 5 kbar (Richardson, Gilbert, and Bell, 1969).

About 20 miles to the southwest at Bloom Lake (Mueller, 1960), the calcic amphiboles coexisting with cummingtonite range in composition from 82-92 mole percent actinolite, thus suggesting metamorphic temperatures between ~550°C and 600°C. Neither Kranck (1961) nor Butler (1969) found coexisting actinolite-cummingtonite in the Mount Reed area, approximately 70 miles southwest of Bloom Lake. Because of the abundance of pyroxenes at Mount Reed, Butler concludes that the grade of metamorphism increases from the Wabush and Bloom Lake areas southwest to the Mount Reed area. A comparison of the mineral assemblages in Table 8 and those in Figure 4 indicates that the mineral assemblages reported by Butler are those expected if the metamorphic grade is indeed higher to the southwest.

The assemblage Mg-rich Act<sup>+</sup>Cpx<sup>+</sup>Opox<sup>+</sup>Qtz (+Carb+Mag+Hem) is reported by Butler (1969, p. 75, assemblage 274). This assemblage is consistent
with the decomposition reaction $\text{Tre} \rightarrow \text{Cpx} + \text{Opx} + \text{Qtz} + \text{H}_2\text{O}$ studied experimentally by Boyd (1959). Kranck (1961) recognized, however, that intermediate Fe-Mg actinolite decomposes to $\text{Cum} + \text{Cpx} + \text{Qtz} + \text{H}_2\text{O}$. He did not find the assemblage $\text{Act} + \text{Cum} + \text{Cpx} + \text{Qtz}$, but the assemblage $\text{Cum} + \text{Cpx} + \text{Qtz}$ is common. The stable tie line between cummingtonite and clinopyroxene plus quartz precludes a tie line between actinolite and orthopyroxene plus quartz (Fig. 5). The conclusion that the decomposition assemblage of the intermediate Fe-Mg actinolites is $\text{Cum} \text{Cpx} \text{Qtz}$ was reached independently in this study.

Iron Formation, Ruby Mountains, Montana

Coexisting actinolites and cummingtonites from the metamorphosed iron formation of the Ruby Mountains are of particular interest because of the conspicuous abundance of exsolution lamellae in some of the amphiboles (Ross et al., 1969). The rocks are composed principally of amphiboles, magnetite, and quartz with minor amounts of apatite, chlorite, and biotite. Ross et al. report that cummingtonite host crystals contain few exsolution lamellae of actinolite, whereas cummingtonite lamellae are numerous in actinolite host crystals. From these observations they concluded that the actinolite-cummingtonite solvus is asymmetric with a very steep cummingtonite limb. This shape has been confirmed in this experimental study.

The actinolite in some of the Ruby Mountain samples appears to have contained considerably more cummingtonite in solid solution than would be expected from this study. In some samples the solubility of cummingtonite in actinolite is as much as 50 percent, which compares with a maximum of about 15 percent shown in Figure 6. The greater solid solution is not an effect of chemical components not present in the synthetic system; Papile, Cameron, and Shaw (1973) found that in the Ruby Mountain samples the solubility of cummingtonite in actinolite decreases with increasing Na and Al content. Although the solvus in Figure 6 has not been reversed experimentally, it is unlikely that the synthetic actinolite coexisting with cummingtonite is less calcic than the stable compositions. The greater solid solution suggests that the Ruby Mountain samples were metamorphosed at higher temperatures and higher $P\text{H}_2\text{O}$ than the experimental conditions of this study. Increased $P\text{H}_2\text{O}$ should raise the upper boundary of the field $\text{Act} + \text{Cum}$ (Fig. 4), thereby exposing more of the solvus and allowing increased solubility of cummingtonite in actinolite.

Decomposition of Ferrotremolite and Mineral Assemblages from an Iron Formation, Gulf of Bothnia, Finland

Ernst (1966) concluded from his experimental studies that pure-Fe ferrotremolite decomposes to $\text{Cpx} + \text{Fa} + \text{Mag} + \text{Qtz} + \text{H}_2\text{O}$ on the FMQ buffer. At the same conditions another possible breakdown assemblage is $\text{Grun} + \text{Cpx} + \text{Qtz} + \text{H}_2\text{O}$, which is analogous to the decomposition products of actinolite found in this study. Ernst did not report the presence of grunerite in any of his runs, but the synthesis of grunerite is known to be difficult.

Natural assemblages should provide information on the decomposition products of ferrotremolite. Hytönen (1968) described samples which contain mineral assemblages relevant to this problem in drill cores taken from beneath the Gulf of Bothnia, Finland, and the author obtained some of these samples.

Seven of the nine samples examined contain the assemblage $\text{Ferrotre} + \text{Grun} + \text{Hd} + \text{Fa} + \text{Mag} + \text{sulfides}$; the assemblages lack quartz. The remaining two samples contain $\text{Ferrotre} + \text{Grun} + \text{Hd} + \text{Qtz} + \text{sulfides}$. All phases were observed in mutual contact, and both the ferrotremolite and grunerite contain fine ($0.01$) exsolution lamellae. Analyses of the ferromagnesian minerals are shown in Table 9.

To the knowledge of the author, the ferrotremolite in Table 9 is the most Fe-rich one which has been reported co-existing with a possible decomposition assemblage, and apparently the assemblage $\text{Ferrotre} + \text{Hd} + \text{Fa} + \text{Mag} + \text{Qtz}$ has never been reported. These observations suggest, but do not prove, that the stable reaction for the decomposition of the pure-Fe amphibole is $\text{Ferrotre} - \text{Grun} + \text{Hd} + \text{Qtz} + \text{H}_2\text{O}$ at $f_0$ defined by the FMQ buffer.

Table 9. Electron Microprobe Analyses of Coexisting Minerals from a Metamorphosed Iron Formation, Gulf of Bothnia, Finland

<table>
<thead>
<tr>
<th></th>
<th>Ca-pyroxene</th>
<th>Ferrotremolite</th>
<th>Grunerite</th>
</tr>
</thead>
<tbody>
<tr>
<td>$\text{SiO}_2$</td>
<td>50.94</td>
<td>47.48</td>
<td>50.04</td>
</tr>
<tr>
<td>$\text{Al}_2\text{O}_3$</td>
<td>0.33</td>
<td>4.04</td>
<td>2.46</td>
</tr>
<tr>
<td>FeO*</td>
<td>25.11</td>
<td>31.22</td>
<td>30.63</td>
</tr>
<tr>
<td>MgO</td>
<td>3.34</td>
<td>3.61</td>
<td>4.36</td>
</tr>
<tr>
<td>CaO</td>
<td>21.28</td>
<td>10.47</td>
<td>10.61</td>
</tr>
<tr>
<td>$\text{Na}_2\text{O}$</td>
<td>0.38</td>
<td>0.89</td>
<td>0.57</td>
</tr>
<tr>
<td>$\text{K}_2\text{O}$</td>
<td>0.00</td>
<td>0.33</td>
<td>0.18</td>
</tr>
<tr>
<td>Total</td>
<td>101.38</td>
<td>98.02</td>
<td>98.84</td>
</tr>
</tbody>
</table>

*Total Fe as FeO
Sample from drill core No. 1, depth 91.5 m.
Acknowledgments

This paper is based on a Ph.D. dissertation submitted to the Faculty of the Virginia Polytechnic Institute and State University. The writer gratefully acknowledges the guidance and advice of M. C. Gilbert who suggested this study. Discussions with D. M. Burt and H. S. Yoder were also particularly helpful. An earlier version of this manuscript was substantially improved by the constructive criticisms of F. D. Bloss, P. H. Ribbe, M. Cameron, and W. G. Ernst.

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