CUMMINGTONITE-GRUNERITE SERIES: A CHEMICAL, OPTICAL AND X-RAY STUDY

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ABSTRACT

Nine new chemical analyses of members of the cummingtonite-grunerite series are presented. The material is from the high grade metamorphic, Precambrian, Wabush Iron Formation, Labrador, Canada. These analyses and additional analyses from the literature are recalculated in terms of the components $Mg_7Si_8O_{22}(OH)_2$, $Fe_7Si_8O_{22}(OH)_2$ and $Mn_7Si_8O_{22}(OH)_2$. The solid solution within this ternary system is found to contain 35 to 100 mole per cent $Fe_7Si_8O_{22}(OH)_2$ and 0 to 34 mole per cent $Mn_7Si_8O_{22}(OH)_2$. Variation in optical properties, unit cell dimensions and density within this solid solution series is presented graphically in binary and ternary variation diagrams. Completely indexed powder diffraction patterns are given for three chemically dissimilar members of the cummingtonite-grunerite series.

The γ index and $Z \wedge c$ angle are the most easily obtainable optical parameters for determination of chemical compositions in the ternary system. The b cell dimension shows an appreciable and systematic variation with composition, whereas $a \sin \beta$ and c vary little. Manganoan cummingtonites (those containing 10 mole per cent or more $Mn_7Si_8O_{22}(OH)_2$) have physical properties which are so similar to those of the tremolite-actinolite series that the presence of manganese must be established chemically.

INTRODUCTION

The variation in optical properties as a function of chemical composition in the cummingtonite-grunerite series has been published by Sundius (1924, 1931) and Winchell (1938). Winchell's data refer to essentially iron-magnesian members of the grunerite series, whereas Sundius' work includes grunerites containing up to 10.95 weight per cent MnO. Bowen and Schairer (1935) show the variation of optical parameters with composition in synthetic fluorine analogues of the iron-magnesian members of the grunerite series. Jaffe *et al.* (1961) give chemical, optical and *x*-ray data for one manganoan cummingtonite, containing 19.2 weight per cent MnO.

The present study was undertaken to investigate the variations in optical and *x*-ray parameters with changes in the Mg, Fe and Mn content of the cummingtonite-grunerite series.

NOMENCLATURE

The nomenclature of the iron-magnesian cummingtonite-grunerite series used in this paper is similar to that suggested by Jaffe *et al.* (1961). Grunerite refers to $(Fe, Mg)_7Si_8O_{22}(OH)_2$ with Fe > Mg; cummingtonite refers to $(Mg, Fe)_7Si_8O_{22}(OH)_2$ with Mg > Fe. The break between cum-

mingtonite and grunerite thus occurs at 50 mole per cent $Mg_7Si_8O_{22}(OH)_2$. The prefix manganoan is used whenever the "Mn" component is present in amounts larger than 10 mole per cent. The prefixes ferroan and magnesian can be used whenever the amount of the "Fe" or "Mg" component exceeds 10 mole per cent. The $Mn_7Si_8O_{22}(OH)_2$ end-member has not been synthesized, is unknown in nature and is not named. The above classification is illustrated on a ternary diagram in Fig. 1.

It is suggested that the names cummingtonite and grunerite be used for all members of the series instead of varietal names such as dannemorite (Erdmann, 1851) and tirodite (Dunn and Roy, 1938; Bilgrami, 1955;
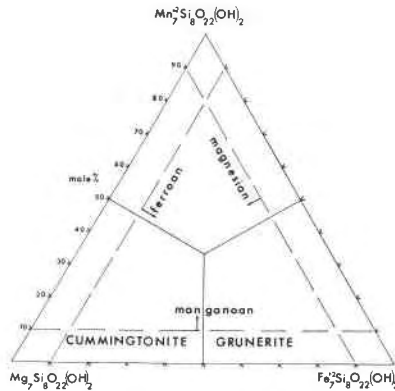

FIG. 1. Suggested nomenclature for the cummingtonite-grunerite series.

Segeler, 1961). Chemical suffixes as proposed by Schaller (1930) can be used to indicate any variation in composition, if known.

DESCRIPTION OF SPECIMENS

The six grunerites and three manganoan cummingtonites used in this study were collected by the writer during the summer field seasons of 1959 and 1961. All nine samples came from the Wabush Iron Formation, a high grade metamorphic Precambrian formation, which crops out in the Labrador City area, Labrador, Newfoundland, Canada (Klein, 1960). The grunerite samples are part of the "silicate" and "silicate-carbonate" horizons. The manganoan cummingtonites are found in the "oxide" horizons. All samples are coarse grained and completely free of alteration. The mineral assemblage and mode, as estimated in the hand specimen, are given in Table 1. A detailed study of the phase assemblages in these iron formations will be published later.

Sample Preparation and Purification

All samples were ground to between minus 50 and 100 mesh. Samples containing magnetite were then purified with a hand magnet (samples No. 11A, 7 and 4). Samples No. 3 and 4, showing a black MnO_2 stain, were soaked for one hour in HCl (1:1) to dissolve the MnO_2. All other samples were also soaked in HCl to remove any limonite coating. In all cases the Frantz Isodynamic separator was used to obtain a 90 to 95 per cent pure amphibole separation. Subsequent purification was ob-

TABLE 1. Mineral Assemblages and Estimated Modes for Nine Samples Containing Members of the Cummingtonite-Grunerite Series

Amphibole No.	Assemblage
1	grunerite (60%)—quartz (40%)schist
9A	grunerite (40%)—hypersthene—En_2Fs_8 (30%)—siderite (25%)—quartz (5%)gneiss
11A	grunerite (50%)—actinolite (30%)—diopside (15%)—magnetite (5%)—carbonate gneiss
7	grunerite (60%)—quartz (20%)—magnetite (15%)—calcite schist
8	grunerite (90%)—quartz (10%)schist (asbestoform variety of grunerite)
10A	quartz (40%)—diopside (30%)—grunerite (25%)—carbonate gneiss
2	Mn-cummingtonite (90%)—specularite (6%)—quartz (2%)gneiss
3	Mn-cummingtonite (80%)—specularite (20%)—schist (MnO_2 stained)
4	Mn-cummingtonite (60%)—quartz (30%)—Specularite (10%)schist (MnO_2 stained)

tained with heavy liquids using methylene iodide and acetone mixtures. The final purification of the samples was done by hand-picking under a binocular microscope. All analyzed samples were estimated to be 99.8 to 100 per cent pure. Optical, x-ray and density measurements were made on a separate part of the analyzed sample.

Chemical Composition

Complete chemical analyses for nine amphiboles are given in Table 2. The number of cations calculated on the basis of 24 (O, OH, F) is also given. In calculating the number of cations the $H_2O(-)$ values were ignored as these most probably represent adsorbed, non-structural water. Justification for this assumption is provided by the calculated values for (OH, F) which are larger than 2 in six out of nine samples. Table 2 also provides calculated molecular ratios for MgO:FeO:MnO.

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Upon inspection of the analyses it becomes obvious that Fe^{2+}, Mg^{2+} and Mn^{2+} ions are present in very large amounts as compared to Ca^{2+}, Na^{2+} and K^+ ions. As such the X and Y (M_4, M_1, M_2, M_3) positions in the general formula $X_2Y_5(Z_8O_{22})(OH)_2$ are almost solely occupied by Fe, Mg or

TABLE 2. GRUNERITE AND CUMMINGTONITE ANALYSES
(ANALYST JUN ITO, DEPT. OF GEOLOGICAL SCIENCES, HARVARD UNIVERSITY)

	1	9A	11A	7	8	10A	2	3	4
SiO_2	49.01	49.33	51.58	51.95	51.79	52.28	55.27	55.74	55.10
TiO_2	0.05	0.02	0.00	0.02	0.02	0.00	0.00	0.00	0.00
Al_2O_3	0.00	0.39	0.10	0.15	0.33	0.07	0.34	0.23	0.10
Fe_2O_3	—[1]	—[1]	—[1]	—[1]	—[1]	—[1]	—[1]	—[1]	—[1]
FeO	44.99	40.94	34.40	33.70	34.38	31.90	4.52	7.09	11.08
MnO	0.37	0.54	0.70	0.99	0.23	0.57	16.62	14.73	13.17
MgO	3.17	6.65	10.33	10.44	10.72	12.35	19.18	18.55	17.00
CaO	0.31	0.18	0.97	0.10	0.14	0.79	1.19	1.04	1.22
Na_2O	0.04	0.12	0.02	0.08	0.09	0.12	0.26	0.08	0.13
K_2O	0.00	0.20	0.05	0.05	0.18	0.08	0.00	0.02	0.02
H_2O+	1.28	1.54	1.99	2.54	1.97	1.62	2.16	2.26	2.05
H_2O-	0.31	—	—	0.22	0.35	—	0.30	0.43	0.43
F_2	1.00	—	—	—	—	—	0.40	0.28	0.23
P_2O_5	0.1	—	—	—	—	—	0.09	—	—
Total	100.63	99.91	100.14	100.24	100.20	99.78	100.33	100.45	100.53
$F_2=O$	0.42						0.17	0.11	0.10
Total	100.21						100.16	100.34	100.43
Spectrographically detected elements	Cu, Ag Sr, Yb	Cu, Cr Ag, Be Y	Cu, Ag B	Cu, Co Ag, Cr Y, Yb Ni	Cu, Co Ag, Cr Y, Yb	Cu, Pb Ag, Y	Cu, Ag Sr, Yb Co, Cr Mo	Cu, Co Mo, Ag	Co, Mo Ag

Numbers of ions on the basis of 24(O,OH,F)

	1	9A	11A	7	8	10A	2	3	4
Si	8.00	7.92 } 8.00	7.95 } 7.97	7.92 } 7.94	7.97 } 8.00	8.00	7.95 } 8.00	8.01	8.02
Al	—	0.08	0.02	0.02	0.03	—	0.05	—	—
Al	—	—	—	—	0.02	0.02	—	0.03	0.02
Ti	—	—	—	—	—	—	—	—	—
Fe	6.14	5.50	4.44	4.30	4.42	4.00	0.54	0.85	1.35
Mn	0.05 } 7.02	0.08 } 7.28	0.09 } 7.06	0.13 } 6.85	0.03 } 7.01	0.07 } 7.19	2.02 } 6.88	1.79 } 6.82	1.63 } 6.91
Mg	0.77	1.59	2.37	2.38	2.46	2.82	4.11	3.97	3.69
Ca	0.06	0.03	0.16	0.02	0.02	0.13	0.18	0.16	0.19
Na	—	0.04	—	0.02	0.02	0.04	0.03	0.02	0.03
K	—	0.04	—	—	0.04	0.02	—	—	—
OH	1.39 } 1.90	1.64	2.04	2.59	2.02	1.66	2.07 } 2.25	2.16 } 2.28	1.99 } 2.09
F	0.51						0.18	0.12	0.10

Molecular ratio FeO:MgO:MnO

	1	9A	11A	7	8	10A	2	3	4
FeO	88.17	76.81	64.26	63.22	64.02	58.56	8.14	12.88	20.25
MgO	11.12	22.12	34.38	34.90	35.58	40.40	61.55	60.03	55.36
MnO	0.70	1.08	1.34	1.89	0.40	1.05	30.31	27.10	24.39

[1] In all analyses iron was determined in two ways: as total Fe and as FeO. Fe_2O_3 was calculated by subtraction. In every analysis the so obtained value for Fe_2O_3 was 0.0% with a range of error of 0.1% (absolute) As the absence of Fe_2O_3 was not proven by direct method, — is used instead of nil. (Dr. Jun Ito).

Mn. In addition, it should be noted that Al_2O_3 is present only in very small amounts, the maximum Al_2O_3 content being 0.39 weight per cent in grunerite No. 9A. The Z-position in $X_2Y_5(Z_8O_{22})(OH)_2$ is thus almost solely filled by Si. TiO_2 and P_2O_5 are present in trace amounts or are not determinable. A few analyses show an appreciable F_2 content, the maximum being 1.00 weight per cent in grunerite No. 1.

Because MgO, FeO and MnO are present in large and variable amounts and because all other oxide components are present in very small amounts only, the nine chemical analyses can be represented graphically in terms of three end-members $Mg_7Si_8O_{22}(OH)_2$, $Fe_7Si_8O_{22}(OH)_2$ and $Mn_7Si_8O_{22}(OH)_2$, as in Fig. 2. It becomes clear from this diagram that samples No. 1, 9A, 11A, 7, 8 and 10A can be considered as phases in a two-component "Mg-Fe" system. Samples No. 2, 3 and 4, however, can only be considered in terms of three components "Mg-Fe-Mn." The completeness of representation of the total analysis of these amphiboles in terms of the "Fe-Mg-Mn" components can be expressed quantitatively by the ratio

$$\frac{Fe^{2+} + Mg^{2+} + Mn^{2+} \text{ (in XY positions)}}{\text{total cations in XY positions}} \times 100.$$

Values for this expression are tabulated in Fig. 2.

The CaO content of the six manganese-poor analyses varies from 0.10 weight per cent (0.02 Ca ions/half unit cell; sample No. 7) to 0.97 weight per cent (0.16 Ca ions/half unit cell; sample No. 11A). The grunerite of sample No. 11A coexists with actinolite (see Table 1). From this assemblage and from its occurrence in a high temperature metamorphic deposit one may conclude that a content of 0.16 Ca ions per half unit cell is close to the upper limit of calcium solubility in manganese-poor cummingtonite and grunerite. Green (1960) gives complete chemical analyses for a hornblende-cummingtonite assemblage belonging to the sillimanite zone. This cummingtonite contains 0.19 Ca ions per half unit cell, which is very similar to the value given earlier. These data lead one to conclude that the upper limit of CaO solubility in manganese-poor cummingtonites and grunerites is approximately 1.5 weight per cent; they also make higher CaO values recorded in the literature somewhat suspect. In this respect, it should be noted that the chemical analysis results presented in this study do not support Layton and Phillips' (1960) suggestion that Ca is essential to stabilize the lattice of cummingtonite.

The manganoan members of the series show a range from 1.04 weight per cent CaO (0.16 Ca ions/half unit cell; sample No. 3) to 1.22 weight per cent CaO (0.19 Ca ions/half unit cell; sample No. 4). These values indicate that Ca is somewhat more soluble in the manganoan than in the manganese-poor members of the cummingtonite-grunerite series.

Eight cummingtonite-grunerite analyses selected from the literature are also represented on Fig. 2. The analyses were selected on the basis of high contents of MgO, FeO and MnO, as compared to Fe_2O_3, K_2O, CaO and Na_2O (maximum Fe_2O_3 content being 1.80 weight per cent in analysis U; maximum K_2O content 0.11 weight per cent in analysis R; maximum CaO content 2.0 weight per cent in analysis U; maximum Na_2O content 0.29 weight per cent in analysis R). In addition, the analyses were selected on the basis of low Al_2O_3 content, the maximum being 2.37

Values for $\dfrac{Fe^{2+} + Mg^{2+} + Mn^{2+} \text{ (in XY)}}{\text{total cations in XY}} \times 100$:

No. 1 —99.14 per cent
No. 9A —98.49
No. 11A—97.73
No. 7 —99.42
No. 8 —98.72
No. 10A—97.36
No. 2 —96.95
No. 3 —97.36
No. 4 —96.82
N —96.61
M —92.58
R —97.06
D —95.10
V —90.88
O —97.80
S —95.24
U —91.97

N (Nsuta)—Jaffe *et al.* (1961)
M (Mikonui River)—Mason (1953)
R (Rockport, Mass.)—Bowen and Schairer (1935)
D (Dannemora)
V (V. Silvberg)   Sundius (1924)
O (Ö. Silvergruvan)
S (Strömshult)—Sundius (1924) and
                              Palmgren (1916)
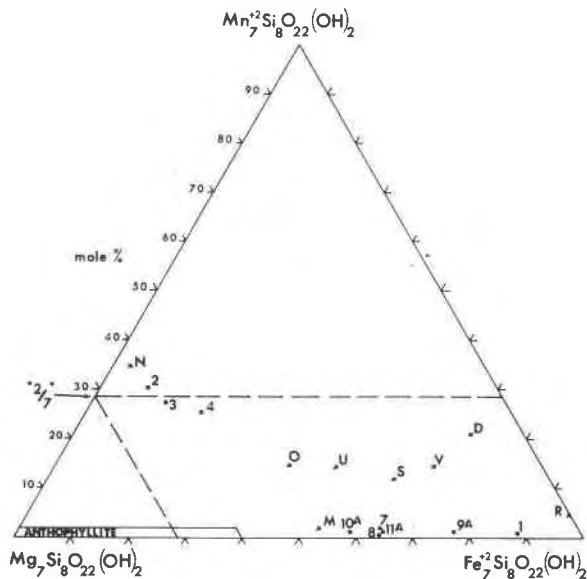U (Uttersvik)—Sundius (1931)

FIG. 2. Chemical composition of cummingtonites and grunerites. Numbered compositions refer to analyses in this study. Lettered compositions refer to analyses from the literature. The extent of the anthophyllite field is after Rabbitt (1948).

weight per cent in analysis M. This selection was necessary in order to permit graphical representation of the total analysis in terms of the components of the diagrams.

Figure 2 shows that the naturally occurring members of this series vary from 35 mole per cent to 100 mole per cent of the $Fe_7Si_8O_{22}(OH)_2$ component and from 0 to 34 mole per cent of the $Mn_7Si_8O_{22}(OH)_2$ component. These limits of solid solution within this amphibole series can probably be related to the crystal structure. In the general amphibole formula $X_2Y_7Z_8O_{22}(OH)_2$ the $X(M_4)$ position is invariably occupied by the larger cations whereas the smaller cations fill the various Y (M_1, M_2, M_3) positions. In the actinolite series Ca^{2+} always occupies the X position. In members of the manganese-poor cummingtonite-grunerite series Fe^{2+} is the principal occupant of the X position, whereas the remaining Fe^{2+} and Mg are distributed over the Y positions (Ghose, 1962). In analogy to this, Mn^{2+} will probably occupy the X position preferentially in the manganoan cummingtonites. If Mn^{2+} were to occupy the X position only, and were not to enter into the Y positions, only two out of seven cations per half unit-cell would be filled by Mn^{2+}. This "2/7" boundary is drawn in on Fig. 2. Mn-cummingtonite No. 2 contains 2.02 Mn^{2+} ions per half unit-cell, whereas Jaffe's analysis N contains 2.28 Mn^{2+} per half unit-cell. The latter analysis sample was not completely pure however, and corrections were made to subtract a certain amount of included spessartite. It is possible that not all spessartite was actually subtracted from the analysis. Jaffe's cummingtonite was associated with rhodochrosite, spressartite, rhodonite, talc and quartz, which reflects a very Mn-rich environment. However, although the Mn-content of the environment was very high, the number of Mn^{2+} ions per half unit-cell in the amphibole is still only about 2. From this one may conclude that the "2/7" line on the diagram may well represent the maximum amount of Mn^{2+} that can be housed in the cummingtonite or grunerite structure. This conclusion is in agreement with the non-existence of naturally occurring manganoan cummingtonites and grunerites which contain more than 35 mole per cent MnO; it also accounts for the impossibility of synthesizing this material in the laboratory.

Figure 2 shows one other line drawn at "2/7" in the anthophyllite corner. All amphiboles which have more than five out of seven cation positions occupied by Mg are known to occur as anthophyllites. Those with less than five out of seven cation positions occupied by Mg occur as members of the cummingtonite-grunerite series. This probably implies that the X position in monoclinic amphiboles is not suitable for housing Mg. As such the "2/7" line represents the maximum content of Mg in the cummingtonite-grunerite series.

Optical Properties

The manganese-poor members of the cummingtonite-grunerite series (No. 1, 9A, 11A, 7, 8 and 10A) are beige in color, showing very slight to no pleochroism under the microscope. The manganese-rich members (No. 2, 3 and 4) are light green in color, and show no pleochroism. All nine samples commonly show multiple twinning parallel to (100). In the hand specimen as well as under the microscope manganoan cummingtonites are easily confused with members of the tremolite-actinolite series; members of both series are light green in color and have very similar optical properties.

Table 3. Optical Properties and Cleavage Angles for Nine
Members of the Cummingtonite-Grunerite Series

Amphibole No.	1	9A	11A	7	8	10A	2	3	4	Variation
γ	1.719	1.708	1.694	1.693	1.693	1.688	1.652	1.661	1.665	± .001
β	1.700	1.690	1.675	1.675	1.675	1.671	1.644	1.648	1.651	± .001
α	1.679	1.667	1.660	1.659	1.659	1.656	1.630	1.634	1.638	± .001
$\gamma - \alpha$	0.040	0.041	0.034	0.034	0.034	0.032	0.022	0.027	0.027	± .002
$Z \wedge c$	13–14°	16°	17°	16–17°	17°	18–19°	19–20°	20°	19°	± 1°
$2V_\gamma$ calc.	93° 52′	98° 06′	83° 36′	88° 02′	88° 02′	87° 20′	107° 28′	92° 22′	89° 02′	—
$2V_\gamma$ meas.	94°	96°	—	85°	82°	—	—	—	—	—
Cleavage 110 $\wedge \bar{1}$10	53° 54′	53° 53′	54° 15′	54° 04′	54° 02′	54° 18′	54° 26′	54° 37′	54° 38′	± 1′

Indices of refraction and extinction angles were determined by means of a spindle stage (Wilcox, 1959) with oils checked at the time of measurement by a Leitz-Jelley refractometer, using a sodium light source. Axial angles were calculated from the expression:

$$\tan^2 V\gamma = \frac{1/\alpha^2 - 1/\beta^2}{1/\beta^2 - 1/\gamma^2}.$$

Several axial angles were also measured directly on a 4-axis universal stage. A complete listing of the optical properties is given in Table 3. Figure 3 is a graphical representation of the variation in optical properties in the manganese-poor grunerites. The slope and position of the curve for the γ index is very similar to curves published by Sundius (1931), Winchell (1938) and Tröger (1956). The deviation of the curve from the determined points is however less than in Sundius' and Winchell's diagrams. The position and slope of the β and α curves are a little different from published data. This is due to the fact that the β and α values in this study were measured directly, whereas many of the published values for β and α were calculated using two indices of refraction
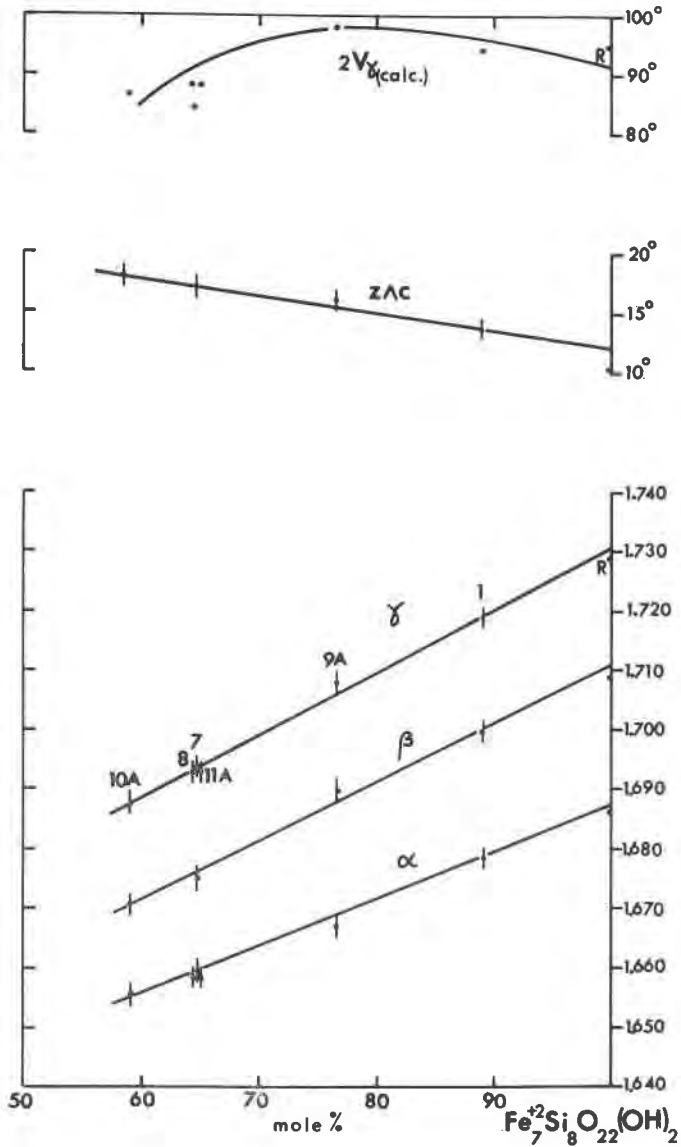
FIG. 3. Variation in optical properties as a function of chemical composition in gruner-ites. Values for R, on the extreme right refer to grunerite from Rockport, Mass. (Bowen and Schairer, 1935) which contains 3.02 mole per cent MnO.

Fig. 4. Relationship of α and γ indices with variation in Mg, Fe and Mn content in the cumingtonite-grunerite series. Crosses with index values refer to α indices from the literature.

and measured 2V. Index curves based upon direct measurement as in this study are considered to be more accurate than curves based upon calculations involving measured 2V. An accurate 2V measurement on a universal stage can be obtained only with extremely well controlled instrumental conditions (Fairbairn and Podolsky, 1951).

Figure 4 is a graphical representation of the variation in γ and α indices on a ternary diagram. Optical parameters for the cummingtonites and grunerites selected from the literature, and referred to earlier in Fig. 2,

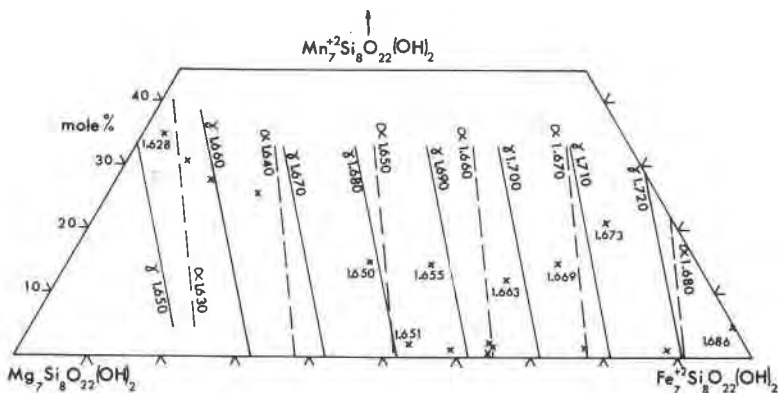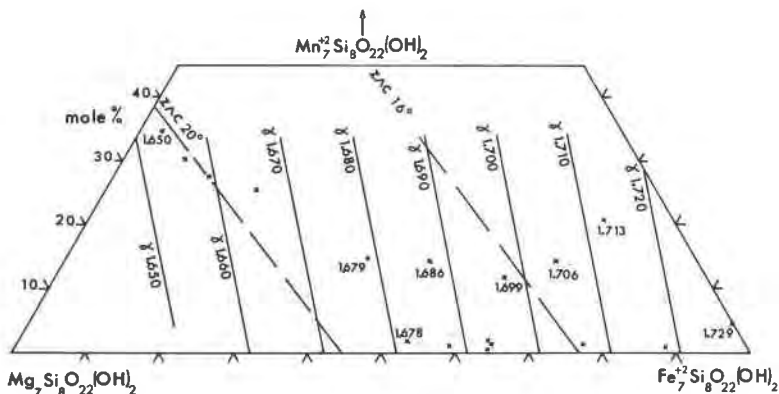Fig. 5. Relationship between γ and $Z\wedge c$ with variation in Mg, Fe and Mn content in the cummingtonite-grunerite series. Crosses with index values refer to γ indices from the literature.

have also been used in constructing Fig. 4. Figure 5 is a plot of the variation in γ and $Z \wedge c$. This diagram enables one to determine the molecular percentage of the three end members through measurement of the two most easily determined optical parameters.

Cleavage

Prismatic cleavage fragments used in single crystal x-ray measurements were mounted on an optical goniometer for orientation. The $110 \wedge \overline{1}10$ cleavage angle was measured and is recorded in Table 3. It is possible to find some correlation between cleavage angle and variation in chemical composition. The manganoan members consistently have a larger $110 \wedge \overline{1}10$ cleavage angle than the iron-magnesian grunerites.

Unit Cell Constants

All nine amphiboles were studied by the rotation, Weissenberg, precession and powder methods. Precession photographs along the b-axis yielded values for β, $a \sin \beta$ and $c \sin \beta$. Weissenberg photographs about c yielded values for b and $a \sin \beta$. Thus obtained values for a and b were improved by using $d_{6 \cdot 0 \cdot 0}$ and $d_{0 \cdot 12 \cdot 0}$ as measured on powder photographs (camera diameter 114.59 mm). The value for c could not be bettered in this manner as no high angle 00l reflections could be identified on the powder diagram. Using the directly measured value for β, values for a and b from powder diagrams, and c values from rotation or precession photographs the powder diffraction films were completely indexed using an IBM 7090 computer program for calculation of all possible d-values. With the aid of indexed single-crystal reflections and an estimate of their relative intensities, it was possible to assign single indices to a large number of the observed diffraction lines. Without relative intensity measurements of single-crystal reflections it would have been impossible to assign single indices, especially to many of the high angle diffraction lines. Approximately twenty to thirty singly indexed powder diffraction lines with 2θ above 30° were then used in a least square program for refinement of the unit cell parameters. The cell constants for cummingtonite No. 2 were refined in this manner, using the least-square refinement program developed at the U. S. Geological Survey. The unit cell parameters of the other eight samples were refined on the Harvard IBM 7090 computer using the program developed by C. W. Burnham at the Geophysical Laboratory, Washington.

The final values for a, b, c, β, V(unit cell) and $a \sin \beta$ compatible with the space group $C\ 2/m$ are given in Table 4. The accuracy of the values for grunerite No. 7 is relatively low because of the inherently broad and

fuzzy diffraction lines on its powder diagram. For this grunerite the accuracy ranges from 0.03 per cent in a, to 0.07 per cent in b and to 0.2 per cent in c. The accuracy for the other eight amphiboles is considerably better, ranging from 0.02 per cent in a of No. 2, to 0.13 per cent in c of No. 4. A higher accuracy would have resulted if the experimental conditions for obtaining the powder diffraction data had been more controlled (see powder diffraction data).

TABLE 4. UNIT CELL DIMENSIONS FOR NINE MEMBERS OF THE CUMMINGTONITE-GRUNERITE SERIES (SPACE GROUP $C2/m$)

	No. 1	No. 9A	No. 11A
a, Å	$9.562 \pm .0020$	$9.551 \pm .0012$	$9.538 \pm .0018$
b, Å	$18.380 \pm .0070$	$18.324 \pm .0061$	$18.248 \pm .0096$
c, Å	$5.338 \pm .0035$	$5.328 \pm .0040$	$5.349 \pm .0059$
β, deg.	$101.86 \pm .026$	$101.86 \pm .018$	$101.97 \pm .026$
V (Å³)	$918.2 \pm .7$	$912.5 \pm .62$	$910.74 \pm .89$
$a \sin \beta$, Å	9.359	9.347	9.330
	No. 7	No. 8	No. 10A
a, Å	$9.545 \pm .0038$	$9.527 \pm .0017$	$9.534 \pm .0019$
b, Å	$18.258 \pm .0137$	$18.238 \pm .0062$	$18.231 \pm .0047$
c, Å	$5.320 \pm .0112$	$5.326 \pm .0046$	$5.3235 \pm .0044$
β, deg.	$101.96 \pm .086$	$101.95 \pm .034$	$101.97 \pm .033$
V(Å³)	907.11 ± 1.4	$905.39 \pm .64$	$905.14 \pm .61$
$a \sin \beta$, Å	9.338	9.320	9.326
	No. 2	No. 3	No. 4
a, Å	$9.583 \pm .0023$	$9.560 \pm .0019$	$9.573 \pm .0026$
b, Å	$18.091 \pm .0050$	$18.089 \pm .0039$	$18.115 \pm .0054$
c, Å	$5.315 \pm .0043$	$5.309 \pm .0036$	$5.304 \pm .0073$
β, deg.	$102.63 \pm .023$	$102.36 \pm .031$	$102.35 \pm .059$
V (Å³)	$899.13 \pm .55$	$896.8 \pm .47$	$898.46 \pm .82$
$a \sin \beta$, Å	9.351	9.338	9.352

Figure 6 is a plot of the variation of b, $a \sin \beta$, and V as a function of composition for the managanese-poor members of the cummingtonite-grunerite series. From this figure it becomes clear that the b parameter is most influenced by changes in composition.

Figure 7 is a ternary diagram showing isodimensional lines for b as a function of Mn, Mg and Fe contents in the cummingtonite-grunerite series. When more such data become available the attitude of the isodimensional lines will become better substantiated.
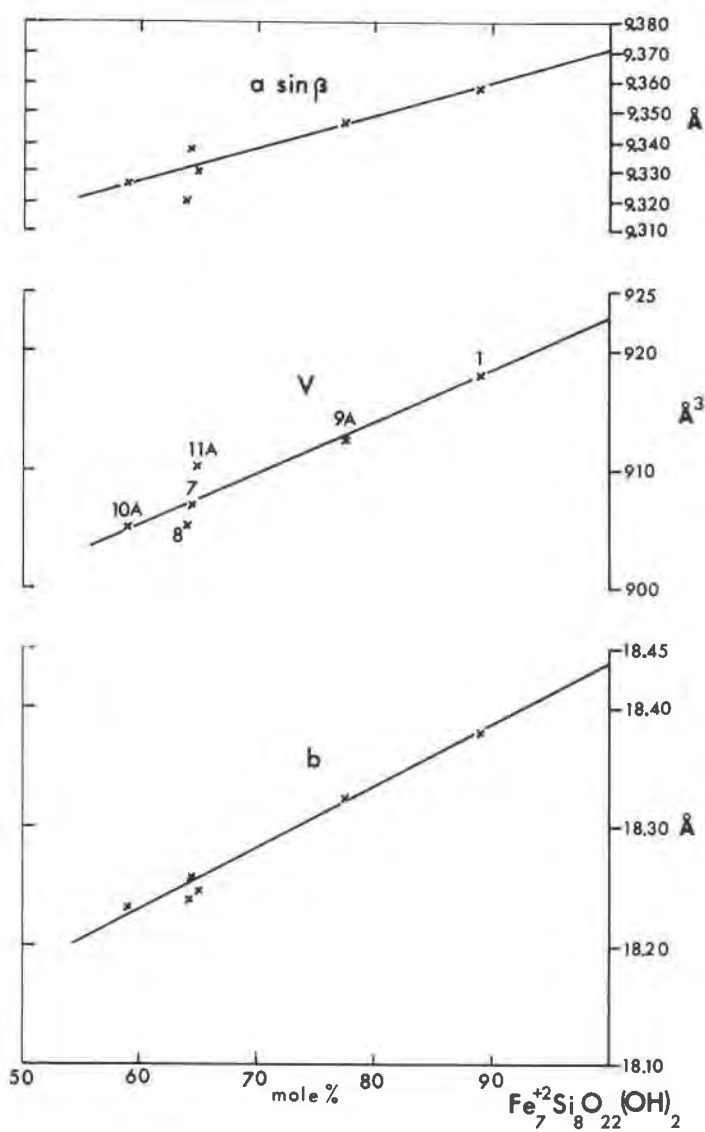
FIG. 6. Variation of *b*, V, and *a* sin β(d_{100}) as a function of the chemical composition of grunerites.

POWDER DIFFRACTION PATTERNS

Powder diffraction patterns for all nine amphiboles of this study were obtained with a camera of 114.59 mm diameter, using filtered iron radiation (Mn filter; Fe $k\bar{\alpha} = 1.93728$ Å; Fe $k\alpha_1 = 1.935970$ Å). All films were exposed for 48 hours in order to obtain relatively intense back reflections. During the exposure the temperature was not controlled. An internal standard was not mixed with the amphibole samples as it was found that some diffraction lines in the crowded amphibole pattern were being masked by the use of a standard. The spindles, rolled with collodion, were dilute and approximately 0.1 mm or less in diameter in order to reduce
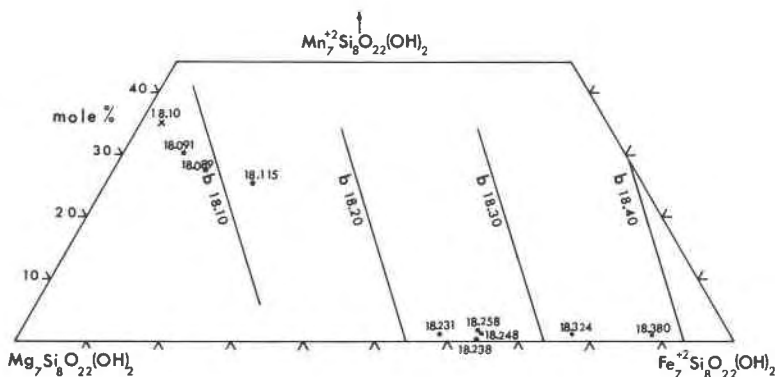
FIG. 7. Diagram showing isodimensional lines for b (attitudes somewhat tentative) as a function of chemical composition in the cummingtonite-grunerite series.

absorption effects in the low angle diffraction lines. The spindles were very accurately centered in the camera by means of a lens. The only correction applied to the resultant powder films was a shrinkage factor. The films were measured with a scale reading directly to 0.05 mm. It was not always possible to locate the diffraction lines to such accuracy, as many high angle reflection lines were fuzzy and not always well resolved in α_1 and α_2 doublets. The accuracy of the unit cell constants given in Table 4 is not only a function of the experimental conditions but also of the inherent broadness and fuzziness of many of the powder diffraction lines.

Table 5 gives completely indexed powder diffraction diagrams for two grunerites (No. 1 and 10A) and one manganoan cummingtonite (No. 2); measured and calculated d-values are given and compared. The estimated relative intensities of the diffraction lines are subject to error as the most intense lines were highly overexposed in order to obtain better definition for less intense high angle lines.

TABLE 5. X-RAY POWDER DIFFRACTION DATA FOR TWO GRUNERITES AND ONE MANGANOAN CUMMINGTONITE (FeK$\bar{\alpha}$=1.93728 Å; FeKα_1=1.935970 Å; Mn FILTER)

Grunerite No. 1							Grunerite No. 1					
I	hkl			d(obs)	d(calc)	d(obs-calc)	I	hkl			d(obs)	d(calc)	d(obs-calc)
50b	0	2	0	9.208	9.190	−.018	10bb	5	1	$\bar{2}$	1.6851	1.6882	−.0031
100bb	1	1	0	8.330	8.339	−.009	30	4	6	1	1.6664	1.6678	−.0014
vf	0	0	1	5.197	5.224	−.027	20	1	11	0	1.6440	1.6449	−.0009
30	1	1	$\bar{1}$	4.840	4.844	−.004	25	1	5	$\bar{3}$	1.6015	1.6013	+.0002
30	2	0	0	4.677	4.679	−.002	10	4	0	2	1.5875	1.5879	−.0004
35	0	4	0	4.582	4.595	−.013		2	10	1		1.5908	−.0033
40	2	2	0	4.160	4.169	−.009	10b	6	0	0	1.5586	1.5598	−.0012
50	1	3	$\bar{1}$	3.880	3.884	−.004	20	0	12	0	1.5314	1.5316	−.0002
vf	2	2	$\bar{1}$	3.592	3.596	−.004	15	3	5	$\bar{3}$	1.5230	1.5235	−.0005
55	1	3	1	3.466	3.468	−.002	10	5	5	1	1.5109	1.5108	−.0001
50	2	4	0	3.278	3.278	0	vf	3	7	2	1.5002	1.4993	+.0009
80b	0	6	0	3.071	3.063	+.008		4	4	2		1.5009	−.0007
	3	1	0		3.075	−.004	vf	6	0	$\bar{2}$	1.4779	1.4797	−.0018
40	2	2	1	2.9969	3.0019	−.0050	10	3	11	0	1.4720	1.4729	−.0009
90b	1	5	1	2.7655	2.7684	−.0029	vfb	4	10	$\bar{1}$	1.4409	1.4408	+.0001
70	0	6	1	2.6391	2.6425	−.0034	35b	6	6	$\bar{1}$	1.4074	1.4089	−.0015
60	2	0	$\bar{2}$	2.5070	2.5110	−.0040	25	5	1	2	1.3884	1.3879	+.0005
10	2	6	$\bar{1}$	2.4120	2.4108	+.0012		4	6	$\bar{3}$		1.3892	−.0008
vf	3	5	0	2.3713	2.3785	−.0072	20	1	11	2	1.3705	1.3697	+.0008
40	0	8	0	2.2995	2.2975	+.0020	5	1	13	1	1.3409	1.3402	+.0007
	3	5	$\bar{1}$		2.3053	−.0058	10	6	6	$\bar{2}$	1.3326	1.3325	+.0001
vf	4	2	$\bar{1}$	2.2480	2.2501	−.0021	35bb	2	12	$\bar{2}$	1.3081	1.3076	+.0005
40	3	1	$\bar{2}$	2.2245	2.2259	−.0014	10	4	12	0	1.2796	1.2815	−.0019
50	2	6	1	2.2019	2.2048	−.0029	vf	4	0	$\bar{4}$	1.2553	1.2555	−.0002
	2	4	$\bar{2}$		2.2035	−.0016	vf	3	7	3	1.2390	1.2372	+.0018
35	0	8	1	2.1015	2.1031	−.0016	20	6	4	2	1.1907	1.1902	+.0005
	2	0	2		2.1040	−.0025	vf	7	5	1	1.1721	1.1712	+.0009
30	3	5	1	2.0448	2.0471	−.0023	vfb	3	1	4	1.1235	1.1229	+.0006
10bb	1	9	0	1.9960	1.9953	+.0007	vfb	7	9	0	1.1195	1.1185	+.0010
	3	7	0		2.0088	−.0128	5	4	12	2	1.1029[1]	1.1024	+.0005
25bb	4	0	$\bar{2}$	1.9560	1.9538	+.0022	10	5	11	2	1.0699	1.0695	+.0004
vf	4	2	$\bar{2}$	1.9115	1.9111	+.0004	15	4	10	3	1.0489	1.0486	−.0003
vf	1	9	$\bar{1}$	1.8905	1.8918	−.0013	10	7	11	0	1.0441	1.0439	+.0002
vfb	4	6	0	1.8591	1.8593	−.0002	5	8	6	1	1.0309	1.0307	+.0002
vfb	1	9	1	1.8370	1.8373	−.0003	20	8	6	$\bar{3}$	1.0181	1.0178	+.0003
	0	10	0		1.8380	−.0010	10	9	5	0	1.0005	1.0006	−.0001
20	3	7	1	1.7966	1.7969	−.0003	15	8	0	2	.9940	.9941	−.0001
vf	0	8	2	1.7243	1.7251	−.0008	5	8	2	2	.9888	.9883	+.0005
vf	3	7	$\bar{2}$	1.7083	1.7052	+.0031	10	6	14	$\bar{2}$	.9821	.9821	0

[1] Reflection 4 12 2 (d obs.=1.1029) and higher are Kα_1 reflections

b =broad
bb =very broad
vf =very faint (I estimated ≤5)

TABLE 5—(*continued*)

I	h	k	l	d(obs)	d(calc)	d(obs-calc)	I	h	k	l	d(obs)	d(calc)	d(obs-calc)
50	0	2	0	9.115	9.115	0	25	6	0	0	1.5541	1.5544	−.0003
100b	1	1	0	8.302	8.303	−.001	40	0	12	0	1.5182	1.5192	−.0010
20	1	3	0	5.085	5.091	−.006	vf	5	5	1	1.5040	1.5037	+.0003
20	1	1	$\bar{1}$	4.827	4.832	−.005	vf	4	8	1	1.4931	1.4947	−.0016
40	0	4	0	4.549	4.558	−.009	vf	6	0	$\bar{2}$	1.4766	1.4764	+.0002
40	2	2	0	4.139	4.151	−.012	15	3	11	0	1.4627	1.4625	+.0002
30	1	3	$\bar{1}$	3.869	3.866	+.003	vf	4	10	$\bar{1}$	1.4305	1.4322	−.0017
30	1	3	1	3.446	3.450	−.004	60	6	6	$\bar{1}$	1.4028	1.4033	−.0005
80	2	4	0	3.258	3.259	−.001	10	5	1	2	1.3821	1.3822	−.0001
90	$\bar{3}$	1	0	3.0631	3.0645	−.0014	vf	1	11	2	1.3599	1.3606	−.0007
20	2	2	1	2.9873	2.9882	−.0009	20	7	1	0	1.3284	1.3288	−.0004
70	1	5	1	2.7538	2.7510	+.0028	40	7 / 2	3 / 12	0 / $\bar{2}$	1.2990	1.3014 / 1.2991	−.0024 / −.0001
50	0	6	1	2.6232	2.6245	−.0013	30	4	12	0	1.2730	1.2729	+.0001
10	2	6	0	2.5435	2.5457	−.0022	vf	7 / 2	1 / 12	1 / 2	1.2287	1.2280 / 1.2301	+.0007 / −.0014
30	0	2	$\bar{2}$	2.5040	2.5058	−.0018	vf	4	12	1	1.2057	1.2054	+.0003
10	3	5	0	2.3639	2.3656	−.0017	30	6	4	2	1.1846	1.1851	−.0005
30	3	5	$\bar{1}$	2.2932	2.2947	−.0015	15	8 / 7	0 / 5	0 / 1	1.1661	1.1658 / 1.1662	+.0003 / −.0001
15	4	2	$\bar{1}$	2.2408	2.2436	−.0024	vf	7	9	0	1.1128²	1.1131	−.0003
15	1	7	1	2.2129	2.2122	+.0 07	10	6 / 4	12 / 12	$\bar{1}$ / 2	1.0962	1.0958 / 1.0956	+.0004 / +.0006
50	2	6	1	2.1904	2.1912	−.0008	vf	8	6	0	1.0887	1.0884	+.0003
15	2	0	2	2.0950	2.0958	−.0008	15	5	11	2	1.0634	1.0633	+.0001
20b	$\bar{3}$	5	1	2.0375	2.0361	+.0014	vf	9	3	$\bar{1}$	1.0439	1.0436	+.0003
vf	3	7	0	1.9951	1.9964	−.0013	40	7	11	0	1.0387	1.0384	+.0003
vf	1	9	0	1.9756	1.9795	−.0038	10	8	6	1	1.0263	1.0263	0
15	3	7	$\bar{1}$	1.9510	1.9532	−.0022	10	8	6	$\bar{3}$	1.0152	1.0149	+.0003
5	1	9	$\bar{1}$	1.8771	1.8780	−.0009	10	4	4	$\bar{5}$	1.0083	1.0087	−.0004
10b	$\bar{5}$	1	0	1.8521	1.8556	−.0035	10	6	6	3	.99596	.99606	−.00010
vf	1 / 0	9 / 10	1 / 0	1.8203	1.8239 / 1.8231	−.0031 / −.0023	vf	8	0	2	.99035	.99011	+.00024
vf	5	1	$\bar{2}$	1.6839	1.6845	−.0006	vf	8	8	1	.98310	.98360	−.00050
vf	5	5	$\bar{1}$	1.6729	1.6742	−.0013							
50	4	6	1	1.6586	1.6590	−.0004							
40	1	11	0	1.6310	1.6318	−.0008							
15	1	5	$\bar{3}$	1.5951	1.5954	−.0003							
15	4 / 2	0 / 10	2 / 1	1.5800	1.5815 / 1.5795	−.0015 / +.0005							

² Reflection 7 9 0 (d obs. = 1.1128) and higher are Kα_1 reflections.

b = broad

bb = very broad

vf = very faint (I estimated ≤ 5)

SPECIFIC GRAVITY

Specific gravity determinations were made with the Berman Micro-balance (Berman, 1937) using 20 to 25 mg samples of minus 50 to 100 mesh grain size. Densities were calculated from the composition and unit cell volumes. Table 6 gives the measured and calculated values. The agreement between these is good except for samples No. 7 and No. 9A. These two grunerites show a difference of 1.7 per cent and 2 per cent, respectively, between the calculated and measured values. Deviations up to several per cent are not uncommon (Mason, 1944, p. 50); considering

TABLE 5--*(continued)*

Manganoan cummingtonite No. 2

I	h	k	l	d(obs)	d(calc)	d(obs-calc)
80b	0	2	0	9.034	9.045	−.011
90bb	1	1	0	8.296	8.307	−.011
vf	0	0	1	5.179	5.186	−.007
20b	1	3	0	5.040	5.068	−.028
20	1	1	$\bar{1}$	4.834	4.842	−.008
50	0	4	0	4.516	4.523	−.007
40	2	2	0	4.150	4.153	−.003
vf	1	1	1	4.054	4.059	−.005
35	1	3	$\bar{1}$	3.856	3.861	−.005
vf	2	2	$\bar{1}$	3.602	3.601	+.001
60b	1	3	1	3.426	3.472	−.006
80b	2	4	0	3.251	3.251	0
100b	3	1	0	3.071	3.027	+.005
30	2	2	1	2.968	2.972	−.004
15	1	5	$\bar{1}$	2.932	2.936	−.004
40	3	3	0	2.766	2.769	−.003
70	1	5	1	2.726	2.726	0
20	3	3	$\bar{1}$	2.661	2.667	−.005
50	0	6	1	2.6027	2.6066	−.0039
50	2	0	$\bar{2}$	2.5111	2.5127	−.0016
5	3	1	1	2.4205	2.4238	−.0033
	2	2	$\bar{2}$		2.4211	−.0006
20	3	5	0	2.3575	2.3615	−.0040
40	1	7	$\bar{1}$	2.2959	2.2983	−.0024
40	4	2	$\bar{1}$	2.2525	2.2570	−.0045
30	3	1	$\bar{2}$	2.2312	2.2328	−.0016
20	2	4	$\bar{2}$	2.1925	2.1965	−.0040
50	2	6	1	2.1742	2.1772	−.0030
vf	3	3	$\bar{2}$	2.1044	2.1081	−.0037
20	2	0	2	2.0799	2.0827	−.0028
30	3	5	1	2.0255	2.0263	−.0008
20	3	7	0	1.9850	1.9985	−.0045
30	4	0	$\bar{2}$	1.9606	1.9629	−.0023
	2	8	$\bar{1}$		1.9595	+.0011
50	5	1	0	1.8600	1.8603	−.0003
vf	4	6	$\bar{1}$	1.8410	1.8440	−.0030
10bb	3	1	2	1.8023	1.7995	+.0028
	0	10	0		1.8091	−.0068
10	5	3	0	1.7848	1.7863	−.0015
40	5	1	$\bar{2}$	1.6951	1.6964	−.0013
	3	7	$\bar{2}$		1.6970	−.0019
30	5	5	$\bar{1}$	1.6768	1.6798	−.0030
60	4	6	1	1.6494	1.6524	−.0030
40b	1	11	0	1.6189	1.6197	−.0008
30	5	3	1	1.5877	1.5897	−.0020
25	6	0	0	1.5566	1.5585	−.0019
vf	6	2	0	1.5311	1.5358	−.0047
10	5	7	0	1.5139	1.5151	−.0012

Manganoan cummingtonite No. 2

I	h	k	l	d(obs)	d(calc)	d(obs-calc)
40	0	12	0	1.5068	1.5076	−.0008
30	4	8	1	1.4885	1.4878	+.0007
	6	0	$\bar{2}$		1.4871	+.0014
30	3	11	0	1.4541	1.4546	+.0005
10	4	10	$\bar{1}$	1.4277	1.4291	−.0014
75	6	6	$\bar{1}$	1.4063	1.4075	−.0012
30	5	1	2	1.3766	1.3764	+.0002
10bb	1	11	2	1.3495	1.3509	−.0014
20	6	6	$\bar{2}$	1.3333	1.3337	−.0004
vf	1	13	1	1.3197	1.3199	−.0002
10	0	12	2	1.3030	1.3033	−.0003
35	2	12	$\bar{2}$	1.2921	1.2928	−.0007
	0	14	0		1.2922	−.0001
25	7	5	$\bar{1}$	1.2789	1.2795	−.0006
10	4	12	0	1.2671	1.2670	+.0001
5	4	0	$\bar{4}$	1.2570	1.2564	+.0006
5	6	0	2	1.2218	1.2229	−.0011
5b	3	13	1	1.2097	1.2095	+.0002
5	2	14	1	1.1952	1.1954	−.0002
5	4	10	2	1.1881	1.1873	+.0008
30	5	11	$\bar{2}$	1.1828[3]	1.1833	−.0005
15	8	0	0	1.1689	1.1689	0
10	8	4	$\bar{1}$	1.1571	1.1579	−.0008
10b	4	14	$\bar{1}$	1.1298	1.1314	−.0016
15	5	13	0	1.1166	1.1164	+.0002
25	8	6	$\bar{1}$	1.1132	1.1132	0
vf	3	13	2	1.1030	1.1029	+.0001
vf	5	5	3	1.0967	1.0962	+.0005
15	4	12	2	1.0888	1.0886	+.0002
10	4	14	1	1.0817	1.0814	+.0003
20	5	11	2	1.0576	1.0573	−.0003
5	5	7	3	1.0508	1.0509	−.0001
5	9	1	$\bar{2}$	1.0451	1.0450	+.0001
40	7	11	0	1.0371	1.0369	+.0002
35	9	3	$\bar{2}$	1.0309	1.0313	−.0004
	6	12	1		1.0298	+.0011
20	8	6	1	1.0253	1.0253	0
20b	8	6	$\bar{3}$	1.0206	1.0206	0
20	6	10	2	1.0132	1.0132	0
vf	6	14	$\bar{1}$	1.0030	1.0032	−.0002
20	2	6	$\bar{5}$	1.0018	1.0025	−.0007
30b	2	10	4	.9910	.9910	0
	6	6	3		.9908	+.0002
vf	7	11	1	.9860	.9851	+.0009
10	3	13	3	.9829	.9829	0
10	8	10	$\bar{2}$	.9792	.9797	−.0005
	4	6	$\bar{5}$		.9796	−.0004
5	6	14	$\bar{2}$	.9757	.9754	+.0003

[3] Reflection 5 11 $\bar{2}$ (d obs. =1.1828) and higher are Kα₁ reflections.

b =broad

bb =very broad

vf =very faint (I estimated ≤5)

Table 6. Measured Specific Gravity and Calculated Density Values
for Nine Members of the Cummingtonite-Grunerite Series

	No. 1	No. 9A	No. 11A	No. 7	No. 8	No. 10A
meas.	$3.54 \pm .02$	$3.45 \pm .02$	$3.38 \pm .02$	$3.41 \pm .02$	$3.39 \pm .02$	$3.37 \pm .02$
calc.	3.54	3.51	3.38	3.35	3.38	3.37

	No. 2	No. 3	No. 4
meas.	$3.13 \pm .02$	$3.21 \pm .02$	$3.22 \pm .02$
calc.	3.18	3.19	3.24

the many possible and difficult to evaluate sources of error in the calculated density values the correspondence is still satisfactory. Figure 8 is a plot of the variation in specific gravity as a function of chemical composition in the manganese-poor grunerites.

DISCUSSION OF RESULTS

The chemical analyses from the literature and the new analyses in this study show clearly that the cummingtonite-grunerite series should be con-
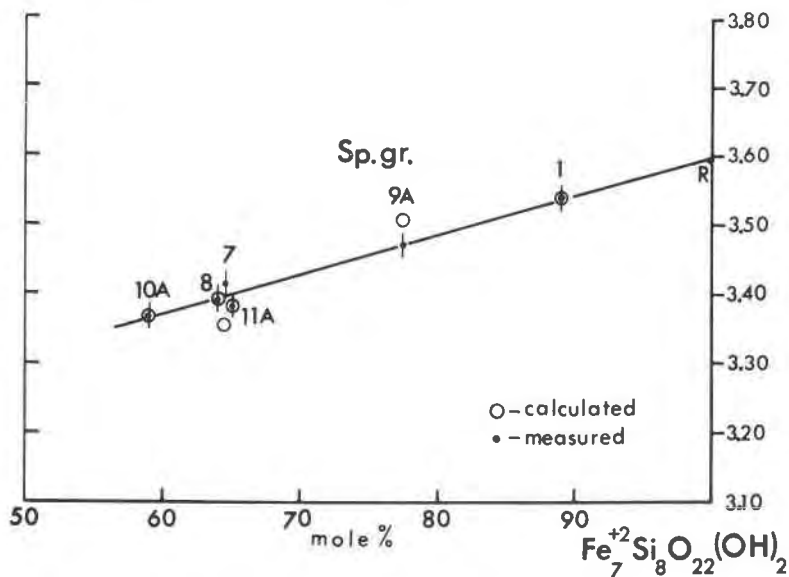
Fig. 8. Variation in specific gravity (measured and calculated) as a function of chemical composition of grunerites. Value for R refers to grunerite from Rockport, Mass. (Bowen and Schairer, 1935).

sidered in terms of at least three components "Mg-Fe-Mn." The Al_2O_3 component is generally small, ranging from zero to about three weight per cent, but may at times be present in amounts up to 8.65 weight per cent (Collins, 1942). The CaO content varies from trace amounts to a probable maximum of approximately 1.5 weight per cent. The effect of the presence of Al_2O_3, CaO and minor amounts of Fe_2O_3, K_2O and Na_2O has not been considered in this work.

The variation in optical parameters, unit-cell constants and specific gravity as a function of chemical composition in the "Mg-Fe-Mn" cummingtonite-grunerite series has been determined. It should be noted, however, that manganoan cummingtonite is still not readily identified as such by its physical properties. The manganoan cummingtonites of this study are light green, show no pleochroism and have optical properties, x-ray parameters and densities which are very similar to those of the tremolite-actinolite series. This means that the presence of manganese must be established chemically.

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