

VARIATIONS IN COMPOSITION AND PROPERTIES OF THE CALCIFEROUS AMPHIBOLES

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ABSTRACT

By plotting the position of about two hundred analyses of common hornblende analyses on a (partial) triangle with $\text{Ca}_2(\text{Mg, Fe})_6\text{Si}_8\text{O}_{22}(\text{OH})_2$, $\text{Ca}_2(\text{Mg, Fe})_3(\text{Al, Fe})_4\text{Si}_6\text{O}_{22}(\text{OH})_2$, $\text{NaCa}_2(\text{Mg, Fe})_5\text{AlSi}_7\text{O}_{22}(\text{OH})_2$, and $\text{NaCa}_2(\text{Mg, Fe})_4(\text{Al, Fe})_3\text{Si}_6\text{O}_{22}(\text{OH})_2$ as its end-members, Hallimond has shown very clearly that many hornblendes have a composition which can be expressed in terms of these four formulas. By using this (partial) triangle as the base of a (partial) triangular prism it is possible to represent (in a three dimensional figure) all the most important variables in the composition of hornblende, and this makes it possible to show the relations between variations in composition and in physical properties as done in Fig. 9. Since several (less important) variables are disregarded in developing this diagram it must be considered as only an approximation. The formulas used in this figure are:



The position of any hornblende analysis in this (partial) triangular prism can be obtained by calculating the number of each kind of atom for $2400(\text{O} + \text{OH} + \text{F})$, dividing each side of the triangle into 200 parts, and then using Si atoms -600 as one coordinate, $\text{Ca} + \text{Na} + \text{K} - 200$ as a second coordinate and $200(\text{Mg} + \text{Al})$ divided by $\text{Mg} + \text{Al}' + \text{Fe}'' + \text{Fe}''' + \text{Ti} + \text{Mn}$ as the third (vertical) coordinate (Al' being equal to total Al atoms minus Al atoms necessary to make $\text{Si} + \text{Al}$ equal to 800).

In a very similar way the composition of certain oxyhornblendes is shown in Fig. 10 and the relations between composition and optical properties are shown in Fig. 12, using the following formulas for the ferriiferous end-members:



This diagram is based on very incomplete data and should be used with caution.

For a long time the writer has emphasized the fact that many minerals vary considerably in composition and he has published diagrams showing the relations between variations in composition and corresponding variations in physical properties for most of the common minerals. In this work hornblende has been exceptionally difficult to deal with, first, because its composition was not fully understood, and second, because it varies in composition in so many ways that no ordinary diagram can be used to represent these variations.

Under these circumstances Hallimond's excellent study of the calciferous amphiboles¹ is of great assistance. It should be clearly understood that Hallimond's study does not include all the possible variations in composition in amphibole nor even all those for which there is now good

¹ Hallimond, A. F., *Am. Mineral.*, **28**, 65 (1943).

evidence of continuous variation. But it does include all the commonest variables and therefore includes most of the "common hornblendes" as well as ordinary tremolite, actinolite, ferrotremolite, pargasite, and hastingsite.

Hallimond includes oxyhornblendes ("basaltic hornblendes") in his study. However, the optic properties of oxyhornblende differ so much

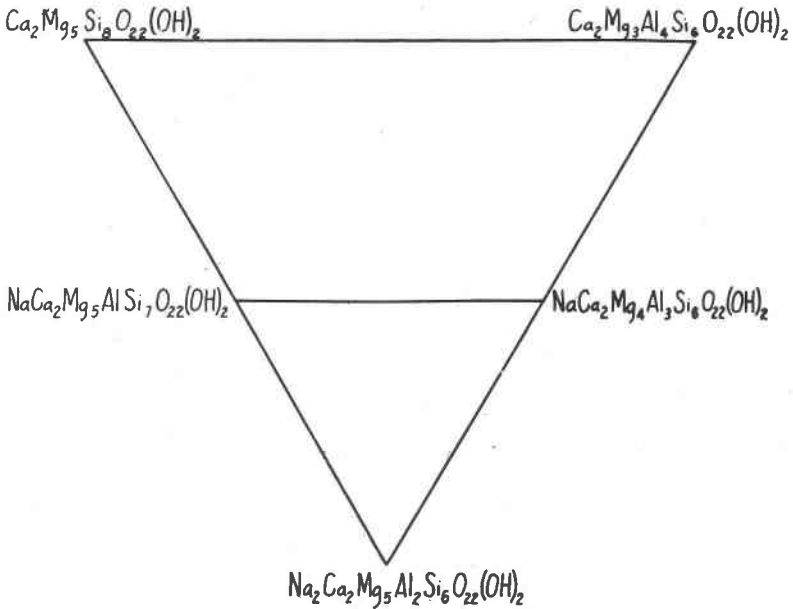


FIG. 1. Relations of some hornblende formulas.

from those of common hornblende that it is necessary to deal with them separately for the purposes of the present study. Moreover, oxyhornblendes differ also in composition from common hornblendes. It is desirable to begin with common hornblendes.

PART 1. COMMON HORNBLENDES

It is well known that Warren's work² established the correctness of Schaller's formula³ for tremolite and also gave us the contents of the unit

² Warren, B. E., *Zeit. Krist.*, **72**, 493 (1930).

³ Schaller, W. T., *U. S. Geol. Surv., Bull.* **610**, 133 (1916).

cell, as well as the presence of a vacant space which can be occupied by an atom of Na, thus permitting an Al atom to replace a Si atom, the total valencies being satisfied by the simultaneous introduction of one Na atom into the vacant space. Hallimond's study makes use of these facts and also shows very clearly that there is continuous variation from the tremolite formula in two different ways (disregarding the variation from Mg to Fe); these two ways are:

- | | |
|---|--|
| | Substitution |
| 1. $\text{Ca}_2\text{Mg}_5\text{Si}_3\text{O}_{22}(\text{OH})_2$ to $\text{Na}_2\text{Ca}_2\text{Mg}_5\text{Al}_2\text{Si}_6\text{O}_{22}(\text{OH})_2$ | Na_2Al_2 for Si_2 |
| 2. $\text{Ca}_2\text{Mg}_5\text{Si}_3\text{O}_{22}(\text{OH})_2$ to $\text{Ca}_2\text{Mg}_3\text{Al}_4\text{Si}_6\text{O}_{22}(\text{OH})_2$ | Al_4 for Mg_2Si_2 |

Thus three formulas are obtained all possible proportions of which may be represented by a triangle, the corners of which are used to represent these formulas. However, reliable analyses of amphibole show that the substitution of Na_2Al_2 for Si_2 extends only half as far as indicated, perhaps due to lack of more vacant spaces. Accordingly, $\text{Na}_2\text{Ca}_2\text{Mg}_5\text{Al}_2\text{Si}_6\text{O}_{22}(\text{OH})_2$ is not an amphibole formula; it is used merely for convenience to complete the triangle, as shown in Fig. 1. The chief end-member formulas for non-ferrous calciferous amphiboles are therefore the following four:

- | | |
|--|---|
| 1. $\text{Ca}_2\text{Mg}_5\text{Si}_3\text{O}_{22}(\text{OH})_2$ | 2. $\text{Ca}_2\text{Mg}_3\text{Al}_4\text{Si}_6\text{O}_{22}(\text{OH})_2$ |
| 3. $\text{NaCa}_2\text{Mg}_5\text{AlSi}_7\text{O}_{22}(\text{OH})_2$ | 4. $\text{NaCa}_2\text{Mg}_4\text{Al}_3\text{Si}_6\text{O}_{22}(\text{OH})_2$ |

Hallimond notes that the second of these formulas is often called the Tschermak molecule; the writer would suggest that it be called tschermakite and the corresponding ferrous formula may then be called ferrotschermakite.

Hallimond has plotted nearly two hundred of the best analyses of calciferous amphiboles on such a (partial) triangle (always assuming that Mg stands for Mg and also Mn, Fe'' , Fe''' and Ti). He uses the number of Si atoms {for $2400 (\text{O} + \text{OH}_2\text{F})$ } as one coordinate and the number of atoms in the "vacant space" as the other coordinate. This is not quite the same as using the formulas, because Na atoms can occupy such spaces as a result of other kinds of replacements, notably 2Na for Ca. The writer has attempted to plot analyses more strictly on the basis of the formulas, excluding all analyses containing more than about 10 per cent of other end-members (including soda-tremolite, glaucophane and all kinds of oxyhornblende) and also all analyses not accompanied by data on the physical properties of the samples concerned. The results are shown in Fig. 2.

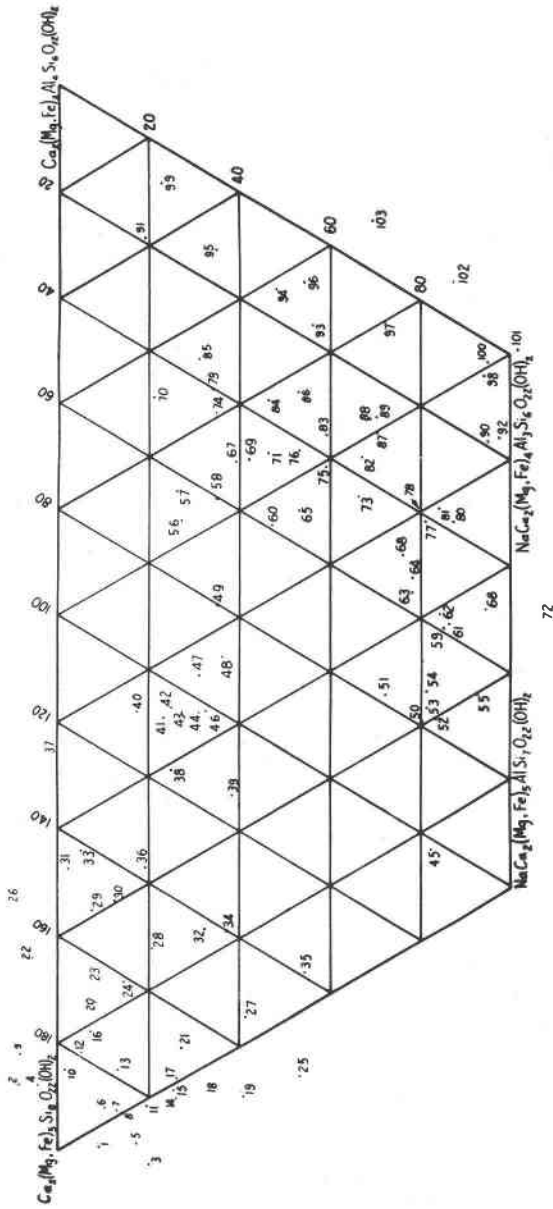


Fig. 2. Composition of selected hornblendes disregarding Mg-Fe variation. (Nos. 88 to 103 are Nos. 89 to 104 of Table 2)

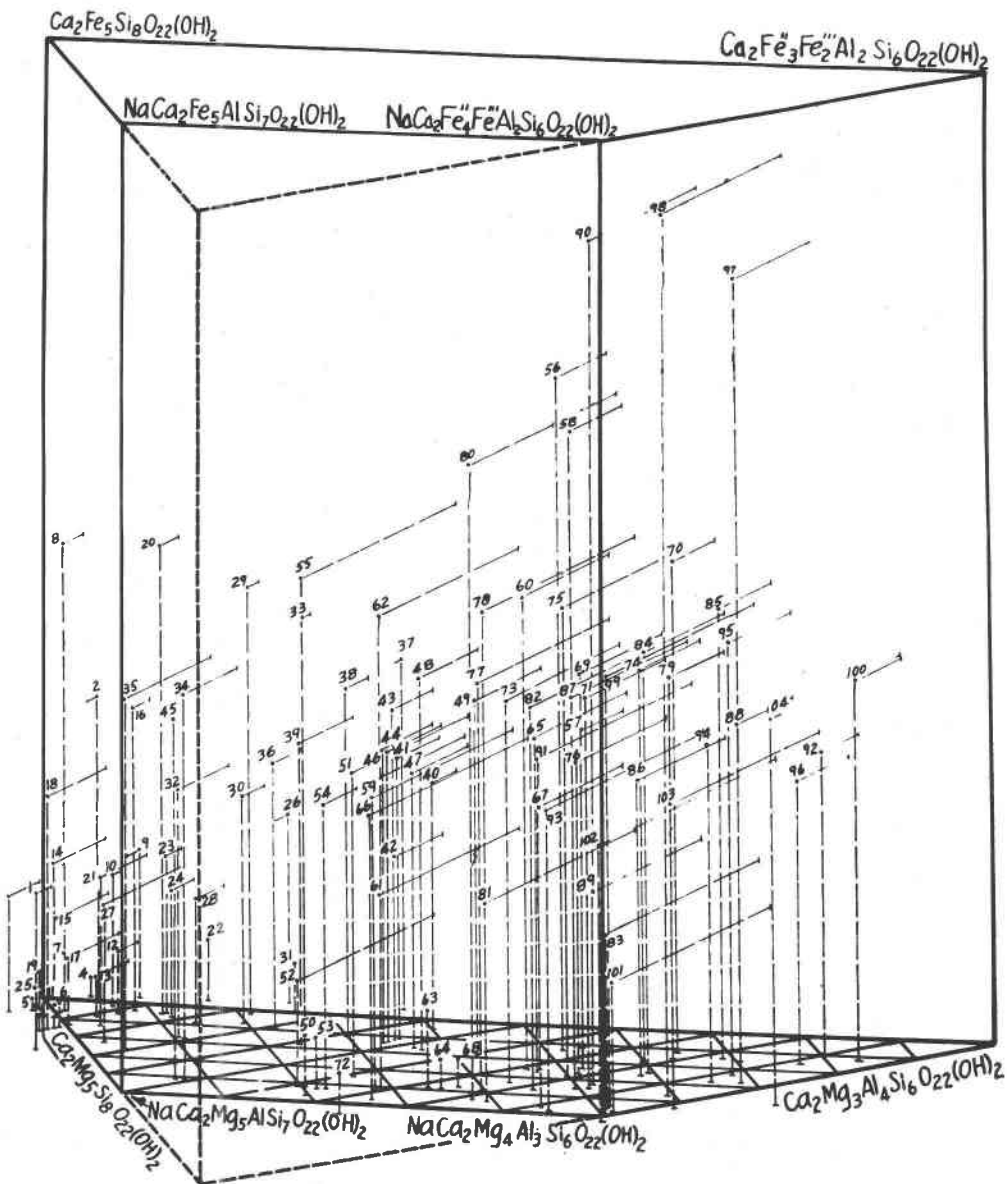


FIG. 3. Composition of selected hornblendes.

Of course physical properties vary with variations in composition but they vary more rapidly with variations in the amount of replacement of Mg by Fe than with the variations shown in Fig. 2. Accordingly it is almost impossible to discover the relations between variations in composition and variations in physical properties without considering the Mg-Fe variation. Since this variation is entirely independent of the variations shown in Fig. 2 and may affect all the end-members of that figure, it can be shown by adding a third coordinate at right angles to the others. This changes the (partial) triangle into (part of) a triangular column, as shown in Fig. 3.

To determine the position of any analysis on the vertical coordinate it is necessary to make certain assumptions. This coordinate may be considered to represent Y of Machatschki's groups⁴: X = (Ca, Na, K), Y = (Mg, Mn, Fe, Al, Ti) and Z = (Si, Al). It is impossible to represent more than one kind of variation along the vertical coordinate. It is thought that the relation between 200 (Mg + Al') and Mg + Al' + Fe'' + Mn + Fe''' + Ti will be the most useful. To find Al' it is only necessary to subtract from the total Al atoms the number necessary to be added to Si to make Si + Al = 800 (on the basis of 2400 (O + OH, F) atoms.) It is assumed that all Mn and Fe'' and Fe''' and Ti atoms belong to the Y group and play the role of Mg atoms. It is now rather generally accepted that AlAl may proxy for MgSi; it seems very probable that Fe''' and Ti do not proxy for Si, but replace Mg only; to keep the total valencies balanced this may be explained as a replacement of MgSi by Fe''' Al and of MgSiSi by TiAlAl. It is a fact that practically all analyses of hornblende contain enough Al atoms to permit this assumption. Accordingly, the vertical coordinate of Fig. 3 is obtained from the relation between Mg + Al' and Fe'' + Fe''' + Mn + Ti.

Even after thus obtaining the position of more than a hundred analyses of calciferous amphiboles in this (partial) triangular column, it is not a simple matter to derive nor to show the corresponding variations in physical properties. And the variations in physical characters can not correspond with the expressed variations in composition in any simple way because there are many unexpressed variations in composition which must have their effects upon the physical properties. For example, no attention is paid to the replacement of Fe'' by Fe''' or Fe'' by Mn or Fe'' by Ti, or Na by K, or OH by F. In spite of all these limitations the writer believes that an expression of the best approximation of

⁴ *Zeit. Krist.*, **70**, 214 (1929).

average conditions, which can be derived, will be useful, if only as something to be improved. After constructing about fifty diagrams showing the variations of physical properties along selected lines and planes in

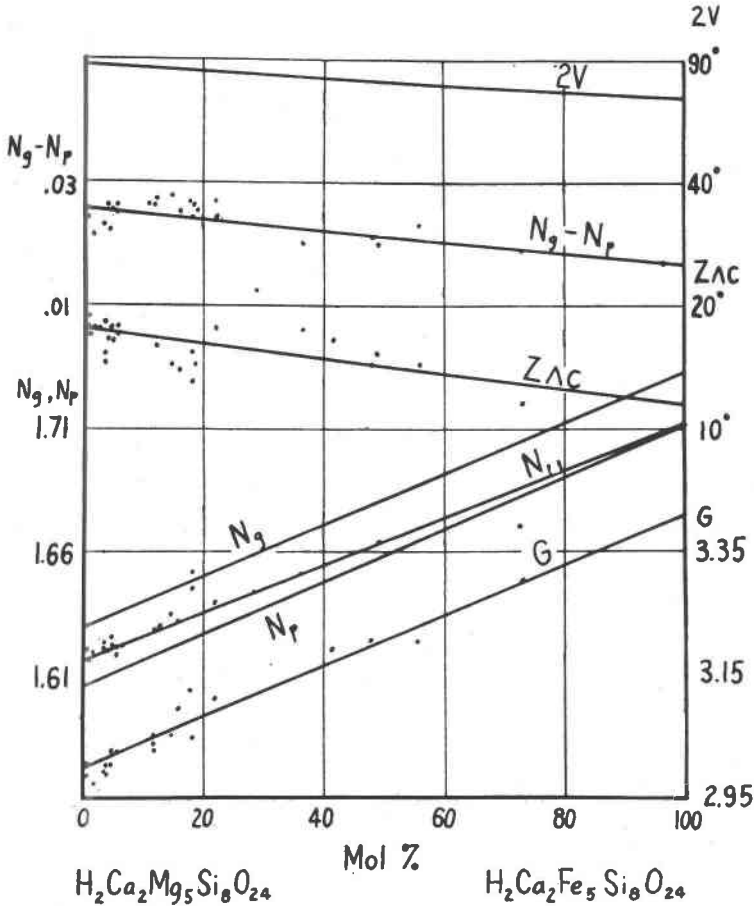


FIG. 4. The tremolite-ferrotremolite series.

this (partial) triangular column, and studying their mutual relations, certain conclusions have been reached.

First, it seems possible, now, to simplify and modify the diagram for the tremolite-ferrotremolite series first published⁵ in 1931 to give the results shown in Fig. 4. This diagram gives the properties of two end-

⁵ Winchell, A. N., *Am. Mineral*, **16**, 257 (1931).

members of the system. The writer is much indebted to Hallimond for an important correction⁶ of the published analysis of "pargasite" from Amity, N. Y., which vitiates entirely the former diagram⁷ for the series: $\text{Ca}_2\text{Mg}_5\text{Si}_8\text{O}_{22}(\text{OH})_2 - \text{NaCa}_2\text{Mg}_5\text{AlSi}_7\text{O}_{22}(\text{OH})_2$. Estimates of the physical properties of this and other end-members of the system follow, derived (in part by extrapolation) from data on analyzed samples inside the (partial) triangular column:

TABLE 1. APPROXIMATE PROPERTIES OF END-MEMBERS

End-member	Sign	2V	N_g	$N_g - N_p$	$Z/\wedge c$	G
$\text{Ca}_2\text{Mg}_5\text{Si}_8\text{O}_{22}(\text{OH})_2$	(-)	88°	1.628	.03	18°	2.98
$\text{NaCa}_2\text{Mg}_5\text{AlSi}_7\text{O}_{22}(\text{OH})_2$	(+)	50°	1.63	.02	25°	3.06
$\text{NaCa}_2\text{Mg}_4\text{Al}_3\text{Si}_6\text{O}_{22}(\text{OH})_2$	(+)	85°	1.64	.02	28°	3.15
$\text{Ca}_2\text{Mg}_3\text{Al}_4\text{Si}_6\text{O}_{22}(\text{OH})_2$	(-)	80°	1.657	.02	20°	3.13
$\text{Ca}_2\text{Fe}_5\text{Si}_8\text{O}_{22}(\text{OH})_2$	(-)	75°	1.735	.025	12°	3.40
$\text{NaCa}_2\text{Fe}_5\text{AlSi}_7\text{O}_{22}(\text{OH})_2$	(-)	20°	1.73	.02	15°	3.42
$\text{NaCa}_2\text{Fe}''_4\text{Fe}'''_2\text{Al}_2\text{Si}_6\text{O}_{22}(\text{OH})_2$	(-)	35°	1.74	.02	18°	3.45
$\text{Ca}_2\text{Fe}''_3\text{Fe}'''_2\text{Al}_2\text{Si}_6\text{O}_{22}(\text{OH})_2$	(-)	70°	1.75	.03	18°	3.42

From these data it is possible to construct diagrams showing the variations in properties on the front faces of the (partial) triangular column (Fig. 5), on the rear face of the column (Fig. 6), on the base of the column (Fig. 7), and on the top of the column (Fig. 8). Finally, it is possible to show (in clinographic projection) the variations in properties in all parts of the column, as done in Fig. 9.

The last two formulas in Table 1 show the replacement of the five atoms of Mg (+Al) by Fe''' as well as Fe''. It should be understood that in locating any analysis in the (partial) triangular column Mn is included with Fe'' and Ti is included with Fe'''. When Ti comes into the problem, the last formula may be written: $\text{Ca}_2\text{Fe}''_4\text{TiAl}_2\text{Si}_6\text{O}_{22}(\text{OH})_2$.

It is important to remember that this (partial) triangular column will not correspond with the facts in every case; indeed it will only rarely correspond with all the data—it is only an estimate of the *average* condition. But it is believed that it will give an *approximately* correct idea of the composition of a sample of amphibole belonging to this system. The amphiboles belonging to this system include tremolite, actinolite, ferro-tremolite, pargasite, hastingsite and "common hornblende," but do not include cummingtonite, grunerite, soda-tremolite, oxyhornblende, glauc-

⁶ Hallimond, A. F., *Am. Mineral.*, **28**, p. 79, note 107 (1943).

⁷ Winchell, A. N., *Am. Mineral.*, **16**, 260 (1931).

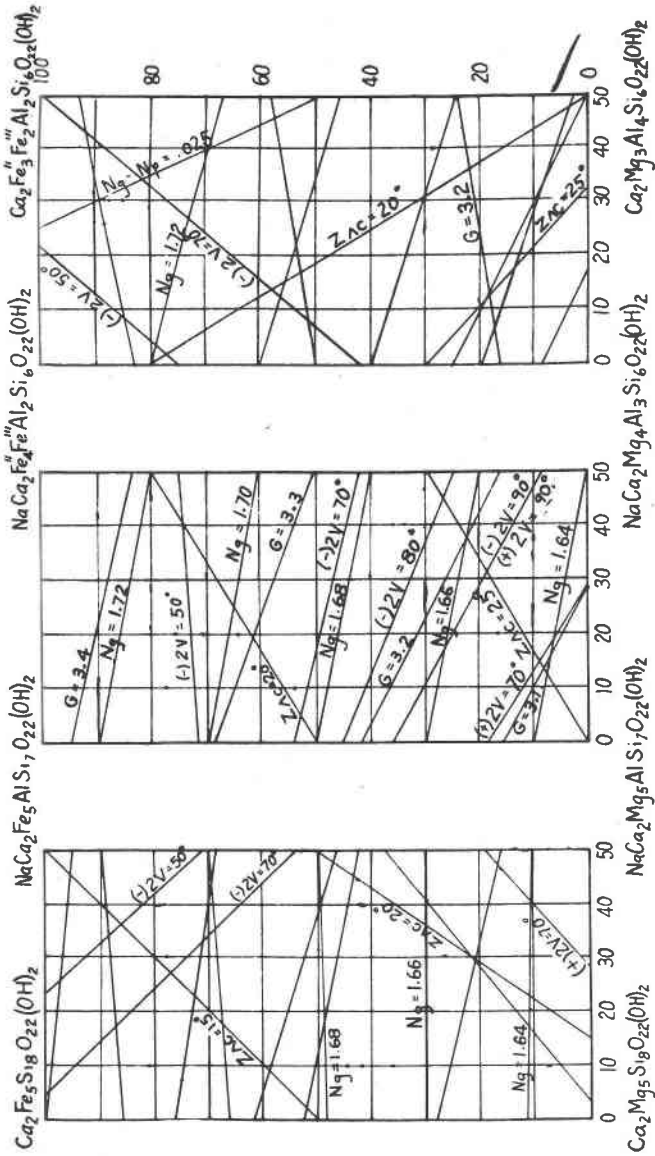


FIG. 5. Properties on the front faces of the (partial) triangular prism (hornblende).

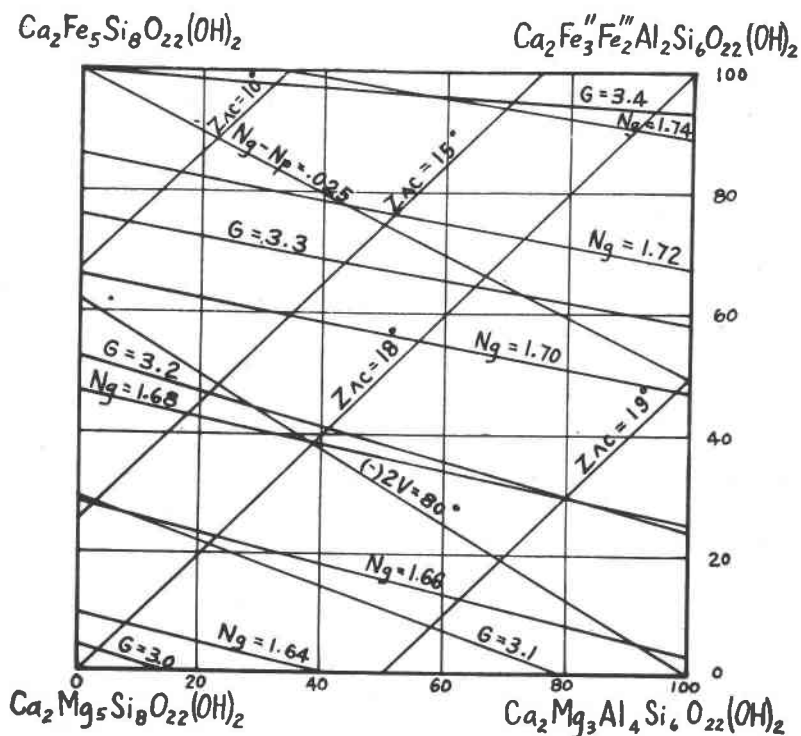


FIG. 6. Properties on the rear face of the (partial) triangular prism (hornblende).

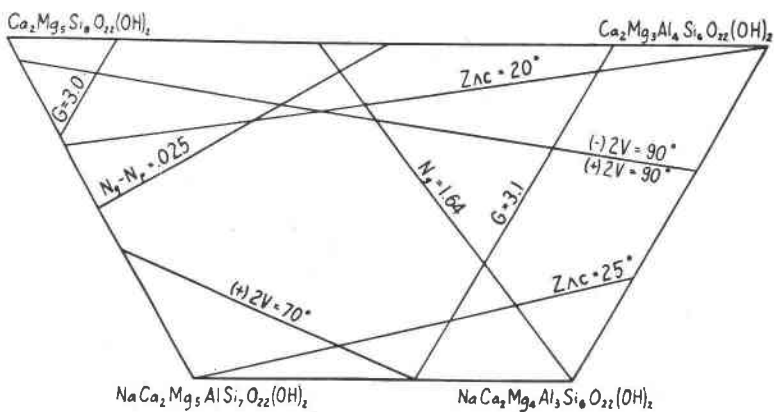


FIG. 7. Properties on the bottom of the (partial) triangular prism (hornblende).

phane, riebeckite, arfvedsonite and intermediate types sometimes called "soda-amphiboles."

For many years the writer has prepared diagrams showing the relations between physical properties and composition on the basis of molecular (or atomic) percentages. It was very surprising to him to find in a recent publication⁸ the statement: "Optic properties in general are more

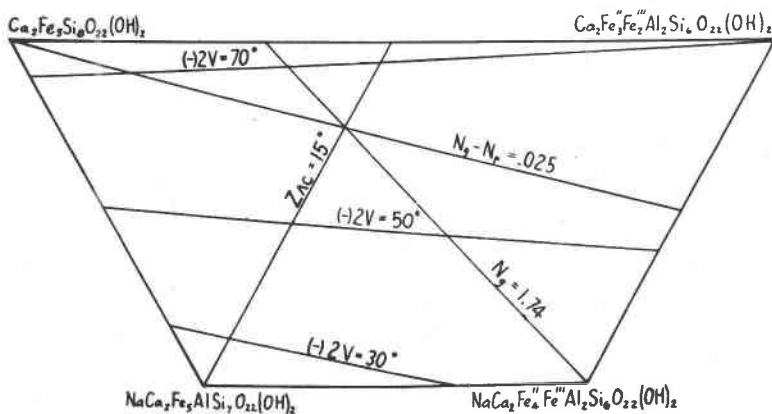


FIG. 8. Properties on the top of the (partial) triangular prism (hornblende).

nearly a function of density (weight percentage) than of number of ions (molecular percentage)." As long ago as 1878 Dufet⁹ proved that the contrary statement is true for orthorhombic crystals. In 1894 Lavenir¹⁰ confirmed Dufet's results and extended them.¹¹ In 1925 Porter¹² proved that the index of refraction is a rectilinear function of the composition *expressed in molecular percentage* for rays vibrating along the axis of symmetry in monoclinic minerals, and, in general, is more simply related to molecular percentage composition than to the weight percentage composition in all crystals. But the relationship is not necessarily nor usually exactly rectilinear in triclinic crystals nor in monoclinic crystals except along the *b* axis.

Table 2 gives the number of atoms (or ions) of each constituent in 104 analyses of calciferous amphiboles on the basis of 2400 atoms of oxygen

⁸ Folinsbee, R. E., *Am. Mineral.*, **26**, 498 (1941).

⁹ *Bull. Soc. Fr. Min.*, **1**, 58 (1878).

¹⁰ *Bull. Soc. Fr. Min.*, **17**, 153 (1894).

¹¹ Among minerals there seem to be some as yet unexplained exceptions to this rule, as illustrated in the triphylite-lithiophilite series and in the new data for the orthorhombic pyroxene, enstenite.

¹² Porter, Mary W., *Proc. Roy. Soc.*, **109**, 78 (1925).

TABLE 2. ATOMIC RATIOS OF CALCIFEROUS AMPHIBOLES FOR (O, OH, F) = 2400

No.	Si -600	Al	Fe'''	Fe''	Mn	Ti	Mg	Ca	Na	K	H	F	Ca+Na+K -200	200(Mg+Al) Mg+Al+Fe	Hallmond's No.	Reference
1	195	23	11	50	3	1	466	189	15	5	77		9	176	2	1
2	192	51	24	135	4		329	178	9	4	96		-9	139	4	1
3	192	20	15	48	2	2	468	190	21	9	63	7	20	176	5	1
4	190	22		7	1		485	188	6	1	197	7	-5	197	9	1
5	190	21	2	2	1		509	190	18	9	106	33	17	198	13	1
6	187	29		6		2	490	193	13	4	142	16	10	197	12	1
7	186	17		22		8	458	190	18	5	209		13	188	10	1
8	186	49	31	217	7		248	176	31	7	61	14	14	105	15	1
9	186	20	8	64	3		442	181	5	5	168	2	-9	171	16	1
10	184	20	2	61	3		450	185	13	3	182		1	175		2
11	182	20	2	2		1	485	205	11	3	202	5	19	198	18	1
12	179	21	6	20	6		477	186	15	4	161	41	5	188	20	1
13	178	2		17			483	200	10	3	136		13	193		3
14	177	57		88			398	158	68		164		26	166	19	1
15	176	37	14	43			463	186	25	14	56	58	25	179	22	1
16	174	45	12	139		3	325	190	15	2	194		7	138	25	1
17	173	21	8	27			474	181	33	12	183	27	26	186		4
18	173	24	0	114	2	2	378	193	39		214		32	152	26	1
19	169	75	5	22		3	450	194	35	12	80	11	41	188		5
20	168	34	32	211			257	192	16		194		8	103	29	1
21	167	34	23	50	2	2	446	184	32	11	36	111	27	171	30	1
22	167	22		27	2	1	484	192	2		123		-6	188		6
23	165	42	27	52		1	422	180	22	5	148	34	7	169	32	1
24	161	49	16	48			426	199	15		200		14	174	35	1
25	159	36	9	17	2		466	169	65	19	200	37	53	189	34	1
26	159	60	5	86	2	2	394	188			221		-12	162	36	1
27	154	62	13	51	Ni 3		401	196	31	14	212		41	172	42	1
28	151	106	7	56	2	5	429	203	13	4	42		20	175	38	1
29	151	40	28	185			280	171	26	10	254		7	114	33	1
30	147	45	33	80	9	3	410	182	21	9	136	41	12	153	44	1
31	146	38	2	21		1	486	187	8	2	148		2	190		7
32	142	124	23	68	3	4	349	210	12	10	91		32	162	48	1
33	141	68	31	176			284	195	10		217		5	117	50	1
34	140	83	33	147	2	4	325	192	33	12	98		37	130	49	1
35	139	160	2	194	4	11	279	203	13	38	17		54	128		8
36	137	21	32	103	2	8	391	187	32		243	19	19	146		9
37	124	118	37	151		3	304	183	9	5	140		-3	129	57	1
38	116	101	72	95	2	13	312	178	34	13	134	12	25	129	59	1
39	114	67	66	103	3	1	380	206	31	1	119		38	137		10
40	109	155	17	107	14	3	358	198	14	5	66		17	150		11
41	107	133	39	100	2	12	337	184	29	10	61	64	23	142	62	1
42	104	98	31	61		8	429	200	22	3	160		25	163	64	1
43	104	110	60	96	2	16	322	187	28	12	157		27	132	65	1
44	102	124	27	116	1	12	343	175	53	3	166		31	141	66	1
45	102	115	16	152		10	289	199	83		198		82	126		12
46	101	118	40	104		5	338	204	28		180		32	142	67	1
47	95	125	22	125			352	197	27	7	203		31	143	69	1
48	89	212	9	156		21	207	208	28		129		36	125		13
49	80	166	54	113	4	19	292	175	39	21	64	37	35	128	75	1
50	79	183	7	20		1	433	189	69	22	67		80	189	79	1
51	78	176	22	121		11	289	183	79	10	176		72	138	77	1
52	77	146	11	40	2	4	457	189	79	15	100	31	83	179		14
53	76	178	8	18			433	183	73	25	93	81	81	190	80	1

TABLE 2—Continued

No.	Si -600	Al	Fe ^{'''}	Fe ^{''}	Mn	Ti	Mg	Ca	Na	K	H	F	Ca+Na+K -200	200(Mg+Al) Mg+Al'+Fe	Hallimond's No.	Reference
54	73	138	50	101	1	1	379	211	35	35	25	57	81	143		15
55	68	100	141	150	1	13	271	213	68	11			92	94	84	1
56	68	163	28	294	17	22	128	184	30	13	176		27	61		16
57	63	165	66	84	2	19	317	175	34	17	168		26	134	88	1
58	60	154	17	228	1	23	166	172	47	16	229		35	80	93	1
59	59	141	46	94	3	16	378	206	39	40	32	66	85	140		17
60	59	227	21	221	2	15	195	193	31	23	88		47	104	95	1
61	58	203	13	27	1	5	430	197	74	15	94	11	86	182	98	1
62	57	203	43	187	2	21	202	230	33	21	52		84	102	101	1
63	56	162	10	20	1	9	463	193	67	18	43	138	78	185	99	1
64	53	227	4	12	1		430	190	78	10	127	12	78	194	103	1
65	53	198	53	112	2	20	304	177	60	16	79		53	131	102	1
66	51	196	52	93	4	10	324	185	72	37	73	23	94	140	106	1
67	51	194	34	98	1	17	348	189	44	16	116		49	145		18
68	50	233	3	9	1		426	192	75	9	128	14	76	195	107	1
69	49	183	56	109	2	41	269	186	36	21	93	41	42	118	108	1
70	48	174	45	177	2	6	213	177	18	26	336		21	101		19
71	46	181	27	153		15	295	191	55		203		46	125	110	1
72	45	204	3	9	1	7	437	202	76	32	68	125	110	192		20
73	44	201	54	143		10	277	186	57	23	163		66	122	113	1
74	44	189	36	160		18	272	181	41	13	200		35	117	115	1
75	42	207	78	158	5	9	221	190	49	20	122		59	104		21
76	42	258	25	132	4	4	259	181	60	12	145		53	137	116	1
77	41	209	31	182	3	17	254	189	73	19	62	39	81	113	109	1
78	40	217	33	185	2	33	209	210	47	23	70		80	102	118	1
79	40	219	116	98			279	207	27				34	122		22
80	38	178	108	215		12	167	178	83	26	96	19	87	71	119	1
81	37	238	19	59		14	328	175	88	21	192		84	163	121	1
82	36	186	70	103		28	289	188	79		136		67	121		23
83	36	214	3	72		8	409	197	38	24	49	105	59	169	125	1
84	36	208	51	162		5	251	176	47	23	210		46	115	122	1
85	36	194	41	187		13	254	181	40	10	204		31	109	120	1
86	31	228	33	133		2	309	186	53	14	156		53	137	130	1
87	30	209	57	137		15	276	197	50	23	132		70	120	132	1
88	29	206	37	111	2	31	305	185	31	19	136	39	35	131	135	1
89	28	267	18	74	1	2	295	189	58	22	224		69	161		24
90	27	209	82	372			34	197	39	34	138		70	27	137	1
91	19	252	29	128	2	31	259	201	67	27	77	17	95	127		25
92	19	233	65	96	2	11	317	185	19*	15	150		19	138	141	1
93	17	263	16	38		5	385	213	57	27	106	10	97	177	152	1
94	17	200	44	107	1	26	323	186	58	12	197		56	131	146	1
95	14	211	44	149		28	252	184	53	11	230		48	111	148	1
96	13	294	29	112	1	8	259	172	47	16	198		35	142	150	1
97	10	237	51	353	17	39	53	170	57	27	65		54	36	153	1
98	8	207	92	341	13	11	33	169	64	39	217		72	19	154	1
99	7	235	84	138	13	4	263	198	50	46	78	3	94	112	156	1
100	7	191	187	3	1	2	296	192	29	2	224		23	121		26
101	4	253	21	42	1	5	399	203	65	27	33	137	95	174	158	1
102	-2	251	53	72	3	20	330	203	64	35	60	35	102	142		27
103	-7	241	78	29	1	57	334	187	54	46	43		87	139		28
104	-10	239	56	86	2	24	304	196	67	6	203		69	133		29

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(plus hydroxyl plus fluorine). For convenience the number of Si atoms is given after subtracting 600, since it then gives directly one coordinate needed to locate the analysis in Figs. 2 and 3. Another coordinate is given in the column headed $Ca+Na+K-200$. And the third coordinate is in the column headed 200 ($Mg+Al'$) divided by $Mg+Al'+Fe$, in which Fe stands for Fe'' , Fe''' , Mn and Ti.

Table 3 gives the physical properties of these 104 analyzed samples of calciferous amphiboles, so far as they have been determined. More measurements of these properties on analyzed samples are much needed, especially in the case of hornblende rich in iron.

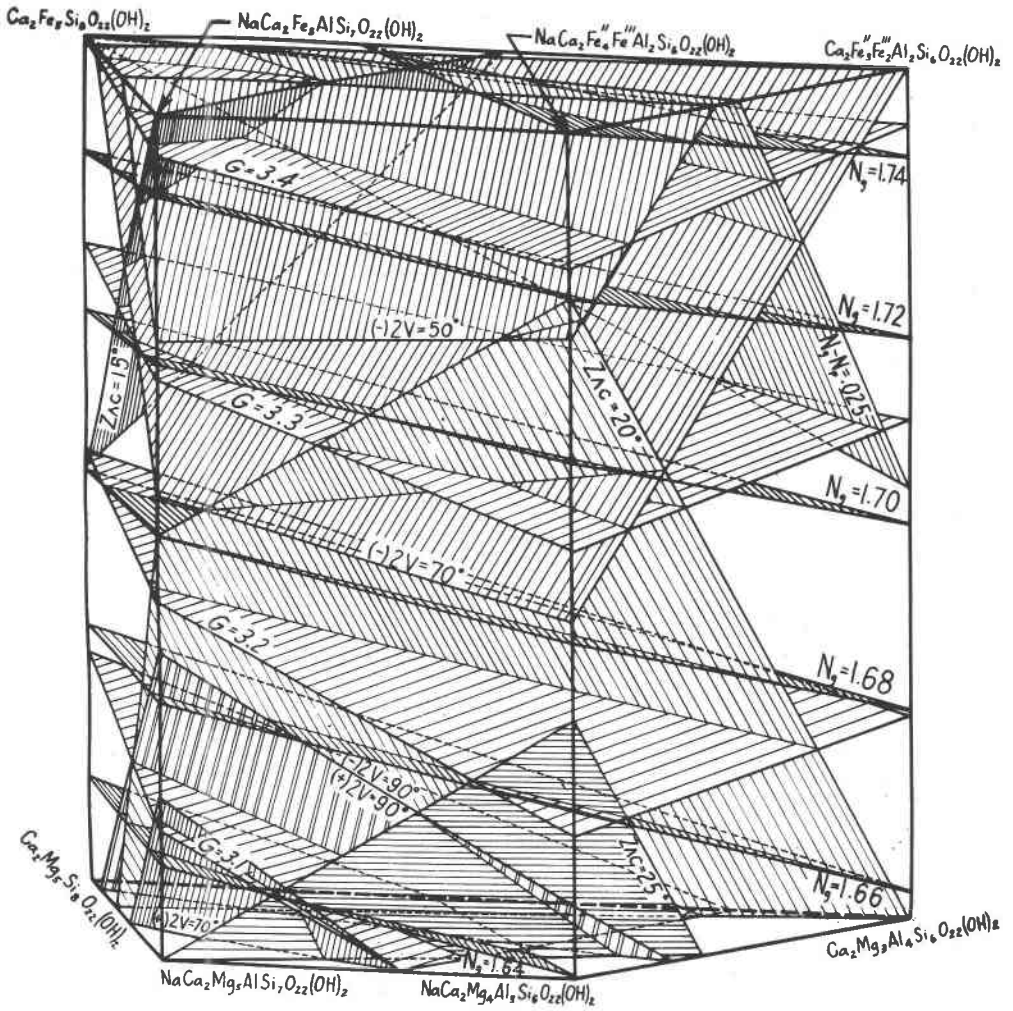


FIG. 9. Physical properties of calciferous hornblendes.

TABLE 3. PHYSICAL PROPERTIES OF CALCIFEROUS AMPHIBOLES

No.	Sign	2V	N_R	N_m	N_p	$N_R - N_p$	$Z \wedge c$	G
1	—		1.634	1.626	1.616	0.018	18°	3.044
2	—		1.653	1.642	1.628	0.025	15°	3.131
3	—		1.639	1.626	1.611	0.028	17°	3.051
4	—	79°38'	1.6272	1.6155	1.6000	0.0272	15°25'	2.98
5	—	88°23'	1.6246	1.6132	1.5992	0.0254	20°1'	2.997
6	—	86°29'	1.6347	1.6192	1.6022	0.0325	16°38'	2.980
7	—		1.632		1.609	9.023		
8	—		1.659	1.649	1.631	0.028	24–27°	3.126
9	—	81°38'	1.6450	1.6330	1.6173	0.0277	14°59'	3.047
10	—	79°49'	1.6410	1.6297	1.6139	0.0271	16°31'	3.047
11	—	82°	1.6307	1.6183	1.6024	0.0283	15°30'	
12	—	87°56'	1.6299	1.6171	1.6036	0.0263	18°18'	3.025
13	—	80°	1.631	1.620	1.608	0.023	17°	2.989
14	—		1.6529		1.6267	0.0262	14°34'	3.116
15	+		1.634	1.621	1.613	0.021	17°	3.056
16	—		1.650	1.641	1.627	0.023	20°	3.11
17	—	84°5'	1.6319	1.6210	1.6065	0.0254	16°54'	3.031
18	—		1.641	1.630	1.617	0.024	18°	3.09
19	—	83°				0.0241	16°	2.996
20	—		1.663		1.642	0.021	15°	3.211
21	—		1.637	1.628	1.618	0.019	19°	3.064
22	—	78°30'	1.632	1.620	1.607	0.025	17°30'	
23	—	81°30'	1.6412	1.6304	1.6162	0.0250	14°27'	3.092
24	—		1.638		1.615	0.023	18°	
25	—	86°14'	1.6244	1.6134	1.6017	0.0227	19°31'	3.03–3.04
26	—	Lg.	1.638	1.629	1.618	0.020	17°	3.06
27	—		1.650		1.628	0.022		
28	—		1.642	1.634	1.625	0.017	12°	3.054
29	—		1.664	1.652	1.637	0.027	20°	
30	—	84°8'	1.6503	1.6382	1.6237	0.0266		3.111
31	—	81°30'	1.634	1.624	1.612	0.022	18°	
32	—	Lg.	1.642	1.631	1.618	0.024	16°	3.147
33	—		1.659		1.638	0.021	17°	3.188
34	—		1.647	1.645	1.631	0.016	18°	3.171
35	+		1.672	1.650	1.649	0.023		3.18
36	—	76°	1.652	1.642	1.626	0.026	16°	3.11
37	—	83°57'	1.6678	1.6551	1.6416	0.0262	15°30'	
38	—	78°	1.664	1.655	1.643	0.021	19°	3.159
39	—	70°	1.654	1.646	1.627	0.027	17°	3.12
40	—	88°30'	1.661		1.636	0.025	16°	3.12
41	—	75°	1.670	1.662	1.651	0.019	19°	3.160
42	—		1.654		1.633	0.021		3.110
43	—	76°	1.671	1.663	1.653	0.018	20°	3.159
44	—	81°	1.652	1.642	1.629	0.023	25°	
45	—	77°	1.666	1.657	1.643	0.023	15°	3.15
46	—		1.661		1.641	0.020		3.18
47	—		1.658		1.640	0.018	17°	3.182
48	—	79°	1.675	1.665	1.650	0.025	19°	3.18
49	—		1.680	1.675	1.652	0.028	14°30'	3.188
50	+	60°29'	1.6351	1.6180	1.6131	0.0220	26°	3.069
51	—	78–82°	1.658		1.638	0.020	14°30'	
52	+	60°	1.645	1.630	1.622	0.023	27°	

TABLE 3—Continued

No.	Sign	2V	N_g	N_m	N_p	$N_g - N_p$	$Z \wedge c$	G
53	+	58°51'	1.6353	1.6205	1.6158	0.0195	26°57'	3.095
54	+	74°	1.654	1.644	1.638	0.016	21°	3.20
55	-	75°44'	1.6798	1.6729	1.6598	0.020	14°40'	3.21
56	-	59°	1.701	1.692	1.673	0.028	15°+	
57	-	77°	1.669	1.660	1.651	0.018	18°	3.164
58	-	77°	1.700	1.687	1.666	0.034	22°	
59	+	76°	1.654	1.643	1.636	0.018	22°	3.20
60	-	75°		1.675		0.016	18°	3.27
61	+	66°	1.6429	1.6284	1.6218	0.0211	24°	
62	-	70°	1.684	1.676	1.660	0.024	17°	3.254
63	-	83°	1.638	1.631	1.622	1.016	20-22°	3.090
64	+	70°	1.6430	1.6291	1.6221	1.0209	21°	
65	-	Lg.	1.683	1.673	1.658	0.025	13°30'	
66	-	88°	1.6665	1.6589	1.6511	0.0154	23°	
67	-	81°	1.666	1.652	1.633	0.033	15°	3.15
68	+	64°	1.6416	1.6265	1.6206	0.0210	22°	
69	-	71°	1.673	1.665	1.654	0.019	17°	3.176
70	-	77°6'	1.673	1.659	1.651	0.022	25°	3.15
71	-		1.574		1.653	0.021		
72	+	52°9'	1.6332	1.6180	1.6142	0.0190	27°30'	3.11
73	-		1.678		1.658	0.020	24°	3.268
74	-		1.676		1.657	0.019	18°	3.26
75	-		1.681		1.661	0.020		3.292
76	-	86°	1.674	1.663	1.651	0.023	16°15'	3.171
77	-	81°42'	1.6789	1.6701	1.6583	0.0206	23°48'	3.285
78	-	66°30'	1.679	1.674	1.661	0.018	13°30'	3.278
79	-	80°4'	1.6843	1.6753	1.6648	0.0195	16°	
80	-	38°	1.7000	1.6980	1.6804	0.0198	20°	
81	-	85°	1.659	1.647	1.636	0.023	21°22'	3.13
82	-	86°	1.673	1.662	1.647	0.026	20°	3.18
83	+	63°1'	1.6519	1.6380	1.6329	0.0190	26°15'	3.186
84	-		1.677		1.658	0.019		3.284
85	-	65°38'	1.681	1.673	1.659	0.022	16°30'	3.234
86	-		1.671		1.652	0.019		3.214
87	-		1.678		1.658	0.020	16°	3.258
88	-	70°	1.672	1.664	1.650	0.022	15°	3.170
89	-	72-74°	1.668		1.635	0.033	15-17°	3.258
90	-	Sm.	1.713	1.710	1.693	0.020	17°	
91	-	73°	1.697	1.691	1.680	0.017	19°	3.267
92				1.65			26°	3.13
93	+	64°5'	1.6530	1.6384	1.6327	0.0203	26°20'	3.18
94	-	75°	1.677	1.669	1.652	0.025	26°	
95	-	78°	1.683	1.674	1.658	0.025	23°	
96	-	84°	1.672	1.661	1.648	0.024	19°	3.20
97	-	47°	1.722	1.719	1.698	0.024	20°	3.375
98	-	16°	1.714	1.713	1.697	0.017	(Y) 15°	
99				1.68				3.283
100	-	79°38'	1.6823	1.6743	1.6576	0.0207	20°	3.224
101	-	88°	1.641	1.632	1.622	0.019	18-19°	3.163
102	-	80°	1.685	1.674	1.665	0.020	26°30'	3.189
103	-	83°	1.718	1.700	1.676	0.042	13°	3.221
104	-	80°	1.685	1.671	1.658	0.027	22°6'	3.187

PART 2. OXYHORNBLENDES

The careful study of the changes in hornblende at about 800°C by Barnes¹³ showed very clearly the relation between "common hornblende" and oxyhornblende. He proved that ordinary hornblende is changed to oxyhornblende by heating to 800°C. and that the chemical change is loss of hydrogen (not water) and oxidation of ferrous iron. He proved that amphibole with practically no iron undergoes practically no change. Therefore ordinary hornblendes containing no iron have the same composition and the same properties as oxyhornblendes free from iron. They are more appropriately known as ordinary hornblendes because they have suffered no oxidation, but they nevertheless are end-members of the oxyhornblende system. Consequently Fig. 7 shows the composition and properties of iron-free oxyhornblendes as well as those of common hornblendes. It may therefore be used as the base of a (partial) triangular prism showing the relations between composition and properties in all parts of that portion of the hornblende system which is the subject of this study—that is, those oxyhornblendes containing the normal tenor of Ca(+Na+K).

There are only a few samples of calciferous oxyhornblendes which have been analyzed and measured optically. In Table 4 available analyses are given after being recalculated to show atomic ratios for 2400 (O+OH, F), as well as the coordinates used in plotting these analyses, namely: Ca+Na+K-200, and 200 (Mg+Al') divided by Mg+Al'+Fe''+Mn+Fe''' + Ti, just as in the case of ordinary hornblendes. The optical properties of these oxyhornblendes are given in Table 5, which also shows the properties of the ordinary hornblendes from which some of these oxyhornblendes were derived by heating.

The sample of hornblende from Frankfort, Pa. (No. 13 of Table 4), which was studied optically by V. E. Barnes in 1930 was analyzed in 1935 by Ray Wilcox with the following results: SiO₂ 41.53, Al₂O₃ 9.31, Fe₂O₃ 9.54, FeO 13.69, MgO 8.14, CaO 10.60, Na₂O 1.04, K₂O 0.79, H₂O 5.35, TiO₂ 0.67, MnO 0.04, F 0.33; Sum 101.03-0.14 (for 2F=O)=100.89. This shows an extraordinarily high tenor of water, but supplies a hornblende very near to tschermakite, whose properties after oxidation are known.

The eighteen analyzed samples of oxyhornblendes of Table 4 are plotted in Fig. 10 according to the same rules used in plotting the analyses of ordinary hornblendes in Fig. 2. Most of these amphiboles were ordinary hornblendes as analyzed; their optical properties were measured, and then they were changed to oxyhornblendes by heating, and the optical properties remeasured. Therefore it seems reasonable to label the end-members as if the minerals were ordinary hornblendes. The proper formulas for oxyhornblendes will be discussed later.

¹³ Barnes, V. E., *Am. Mineral.*, 15, 393 (1930).

TABLE 4. ATOMIC RATIOS OF CALCIFEROUS OXYHORNBLENDES FOR (O, OH, F) = 2400

No.	Si -600	Al	Fe ^{'''}	Fe ^{''}	Mn	Ti	Mg	Ca	Na	K	H	F	Ca+Na+K -200	200(Mg+Al) Mg+Al+Fe	Reference
1	182	20	2 ^b	2 ^b		1	485	205	11	3	202 ^b	5	19	198	1
2	141	68	31 ^b	176 ^b		4	284	195	10		217 ^b		5	118	2
3	110	110	64 ^b	147 ^b		5	286	154	89	27	116 ^b	10	70	116	1
4	72	134	160	35	5	23	298	178	39	21	20	17	38	115	3
5	58	203	13 ^b	27 ^b	1	5	430	197	74	15	94 ^b	11	86	183	1, 4
6	53	227	4 ^b	12 ^b	1		430	190	78	10	127 ^b	12	78	194	1, 4
7	51	196	141	4	4	10	324	185	72	37	5	23	94	140	1
8	50	233	3 ^b	9 ^b			426	192	75	9	128 ^b	14	76	195	1, 4
9	45	204	3	9	1	7	437	202	76	32	68	125	110	192	1, 5
10	38	178	274	53	tr	12	167	178	83	26	8	19	87	70	1
11	36	208	51 ^b	162 ^b		5	251	176	47	23	210 ^b		46	115	2
12	7	222	137	102		28	222	200	67	20	46		87	97	2
13	1	158	104 ^b	164 ^b	1	7	176	164	29	14	516 ^b	15	7	78	1
14	0	210	131	3	9	33	315	187	70	29	122*		86	130	6
15	-1	226	85	80		46	289	194	66	28	72		88	120	2
16	-3	231	76	64		48	303	199	81	18	67		98	128	2
17	-7	241	78	29	1	57	334	187	54	46	43		87	138	7
18	-8	262	86	47		29	320	197	65	23	57		85	140	2

^b Before heating.

* From ignition.

REFERENCES FOR TABLE 4.

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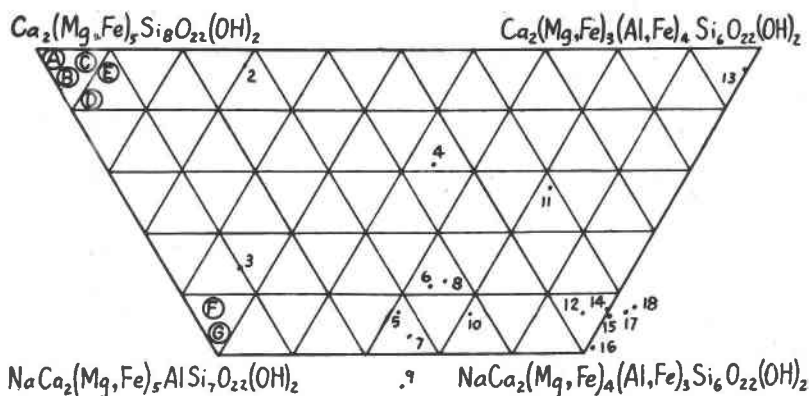


FIG. 10. Composition of selected oxyhornblendes.

TABLE 5. OPTICAL PROPERTIES OF CALCIFEROUS OXYHORNBLENDES

No.	Sign	2V	N_g	N_m	N_p	$N_g - N_p$	$Z \wedge c$	
1	—	82°	1.6307	1.6183	1.6024	0.0183	15½°	Before heating
1	—	82°	1.6308	1.6178	1.6026	0.0182	18°	After heating
2			1.659		1.638	0.021	17°	Before heating
2			1.676		1.655			After heating
3	—	63°	1.6702	1.6656	1.6539	0.0163	32°	Before heating
3	—	61°	1.7193	1.7060	1.6666	0.0527	4°	After heating
4	—	Lg.	1.735	1.715	1.675	0.060	5°	Natural
5	+	66°	1.6429	1.6284	1.6218	0.0211	24°	Before heating
5	+	86°	1.6526	1.6402	1.6287	0.0239	21°	After heating
6	+	70°	1.6410	1.6256	1.6188	0.0222	21°	Before heating
6	+	71°	1.6430	1.6291	1.6221	0.0209	21°	After heating
7	—	88°	1.6665	1.6589	1.6511	0.0154	23°	Before heating
7	—	66°	1.7100	1.6961	1.6626	0.0474	5°	After heating
8	+	64°	1.6416	1.6265	1.6206	0.0210	22°	Before heating
8	+	70°	1.6413	1.6265	1.6190	0.0223	28°	After heating
9	+	58°	1.6331	1.6186	1.6143	0.0188	27°	Before heating
9	+	58°	1.6317	1.6204	1.6170	0.0147	25°	After heating
10	—	38°	1.7000	1.6980	1.6804	0.0198	20°	Before heating
10	—	65°	1.7960	1.7690	1.7020	0.0940	0°	After heating
11			1.677		1.658			Before heating
11			1.697		1.676			After heating
12			1.700		1.681	0.019	0°	Natural
13	—	45°	1.6980	1.6950	1.6801	0.0179	22°	Before heating
13	—	69°	1.7825	1.7564	1.7003	0.0822	0°	After heating
14	—	82°	1.731	1.711	1.685	0.046	1°	Natural
15			1.701		1.679	0.022		Natural
16			1.700		1.677	0.023	6°	Natural
17	—	83°	1.718	1.700	1.676	0.042	13°	Natural
18			1.668		1.667	0.021	3°	Natural
A	—	81°	1.6278	1.6178	1.6042	0.0236	20°	Before heating
A	—	72°	1.6367	1.6262	1.6058	0.0309	13°	After heating
B	—	77°	1.6267	1.6181	1.6041	0.0226	16°	Before heating
B	—	83°	1.6284	1.6175	1.6014	0.0270	15°	After heating
C	—	82°	1.6301	1.6201	1.6067	0.0234	18°	Before heating
C	—	70°	1.6377	1.6270	1.6053	0.0324	12°	After heating
D	—	78°	1.6403	1.6278	1.6085	0.0318	16°	Before heating
D	—	56°	1.6617	1.6532	1.6220	0.0397	11°	After heating
E	—	73°	1.6413	1.6313	1.6133	0.0280	15°	Before heating
E	—	56°	1.6728	1.6634	1.6301	0.0427	10°	After heating
F	—	72°	1.6792	1.6715	1.6578	0.0214	20°	Before heating
F	—	65°	1.7477	1.7288	1.6759	0.0718	0°	After heating
G	—	86°	1.6640	1.6510	1.6370	0.0271	10°	Before heating
G	—	65°	1.7175	1.6980	1.6507	0.0668	1°	After heating

Barnes measured the optical properties of several hornblendes (both before and after heating till oxidation occurred) which have never been analyzed. It is possible to obtain the approximate composition of these samples from Fig. 9, and thus to plot their approximate position on Fig. 10. The fact that their composition is only roughly known is suggested by indicating their position by a small circle; it is *not* intended to suggest that the correct position must be inside the circle. These samples are labeled: *A* to *G*, as follows: *A*, tremolite, Russell, N.Y.; *B*, tremolite, Natural Bridge, N.Y.; *C*, actinolite, Pierrepont, N.Y.; *D*, actinolite, Zillertal, Tyrol; *E*, actinolite, Val Malenco, Italy; *F*, hornblende, Milford, Mass.; *G*, hornblende, Tyrol.

TABLE 6. APPROXIMATE PROPERTIES OF END-MEMBERS

	Sign	2V	N_g	$N_g - N_D$	$Z/\lambda c$
$\text{Ca}_2\text{Fe}''_3\text{Fe}'''_2\text{Si}_8\text{O}_{24}$	(-)	55°	1.78	.070	0°
$\text{NaCa}_2\text{Fe}''_3\text{Fe}'''_2\text{AlSi}_8\text{O}_{24}$	(-)	60°	1.79	.075	0°
$\text{NaCa}_2\text{Fe}''_2\text{Fe}'''_3\text{Al}_2\text{Si}_6\text{O}_{24}$	(-)	55°	1.80	.085	0°
$\text{Ca}_2\text{Fe}''\text{Fe}'''_4\text{Al}_2\text{Si}_6\text{O}_{24}$	(-)	55°	1.80	.095	0°

Since the chemical change involved in converting ordinary hornblende into oxyhornblende is loss of hydrogen from hydroxyl, the oxygen atom which remains in the crystal having its valence satisfied by the simultaneous change of one atom of iron from the ferrous to the ferric state, the process can result in the oxidation of only as many atoms of iron as the number of atoms of hydrogen which are driven out of the crystal. This conclusion is confirmed by the results of the analysis of the hornblende from Renfrew Co., Ont. (after complete oxidation at about 800°C) which show that even after driving out all the hydrogen an oxyhornblende may still contain ferrous iron. Therefore no oxyhornblende can contain more than two atoms of ferric iron which are due to the process of oxidation. Since the iron end-member formulas of ordinary calciferous hornblende are: $\text{Ca}_2\text{Fe}''_5\text{Si}_8\text{O}_{22}(\text{OH})_2$, $\text{NaCa}_2\text{Fe}''_5\text{AlSi}_7\text{O}_{22}(\text{OH})_2$, $\text{NaCa}_2\text{Fe}''_4\text{Fe}'''_1\text{Al}_2\text{Si}_6\text{O}_{22}(\text{OH})_2$ and $\text{Ca}_2\text{Fe}''_3\text{Fe}'''_2\text{Al}_2\text{Si}_6\text{O}_{22}(\text{OH})_2$, the corresponding formulas for oxyhornblende must be $\text{Ca}_2\text{Fe}''_3\text{Fe}'''_2\text{Si}_8\text{O}_{24}$, $\text{NaCa}_2\text{Fe}''_3\text{Fe}'''_2\text{AlSi}_7\text{O}_{24}$, $\text{NaCa}_2\text{Fe}''_2\text{Fe}'''_3\text{Al}_2\text{Si}_6\text{O}_{24}$ and $\text{Ca}_2\text{Fe}''\text{Fe}'''_4\text{Al}_2\text{Si}_6\text{O}_{24}$.

Knowing the approximately correct position of the hornblende samples *A* to *E* on Figs. 9 and 10, their content of iron can be obtained fairly closely from Figs 4, 5 and 6. In this way it is possible to find that actual data extend only to 20 per cent of the iron molecule in the $\text{Ca}_2\text{Mg}_5\text{Si}_8\text{O}_{22}$

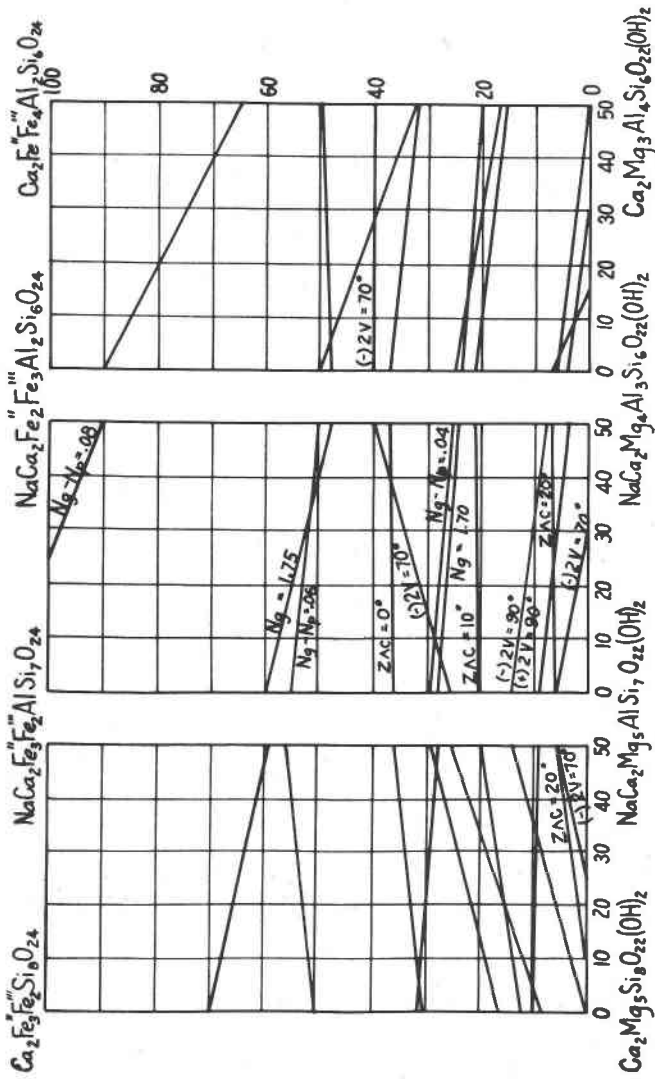


FIG. 11. Properties on the front faces of the (partial) triangular prism (oxyhornblende).

(OH)₂ -Ca₂Fe''₃Fe'''₂Si₈O₂₄ series. For all the rest of this series the properties are obtained by extrapolation, keeping in mind that only two of the five replacing Fe atoms can be oxidized, and also using the conclusions reached by Barnes that (1) the extinction angle decreases to zero, but

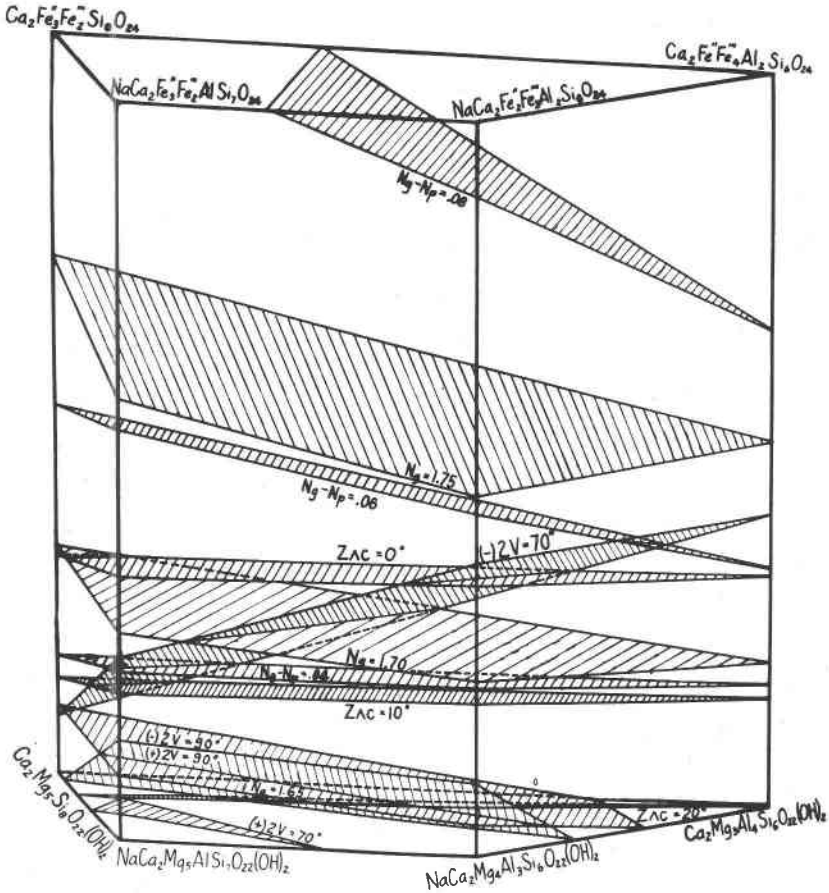


FIG. 12. Optical properties of calciferous oxyhornblends.

does not become minus, and (2) the optic angle about X decreases to about 55°, but not to any value materially less, no matter how much iron may be present. It is surprising to discover that both of these changes are completed before all the oxidation is completed.

In other parts of the system data are available to about 60 per cent of the iron molecules.

By such approximate and admittedly unsatisfactory methods the properties of the iron end-members of the calciferous oxyhornblende system have been derived as given in Table 6.

The variations in properties on the front faces of a (partial) triangular prism of the oxyhornblende system are shown in Fig. 11. Finally the variations in properties as related to variations in composition in all parts of the system are shown in clinographic projection in Fig. 12. It is obvious that this is only a first approximation.

In using this graph it should be remembered that it represents the conditions in oxyhornblendes after complete oxidation; many, if not all, natural oxyhornblendes are not completely oxidized; therefore their properties are intermediate between those of ordinary hornblende, and those of completely oxidized hornblende. It may be useful to note that the size of the extinction angle is a guide to the amount of oxidation, but, natural oxyhornblendes with an extinction angle of zero are not completely oxidized (at least in most cases). The size of the optic angle is also a guide to the amount of oxidation, but it can not be measured as easily as the extinction angle.

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