OPTIC PROPERTIES OF CORDIERITE IN RELATION TO ALKALIES IN THE CORDIERITE-BERYL STRUCTURE

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Abstract

The writer presents a chemical analysis of optically positive, gem quality cordierite from a new occurrence, north of Great Slave Lake. Anomalies in the Shibata (Winchell) variation diagrams, which relate the optic properties of cordierite to the iron-magnesium ratio are noted. The relation of the cordierite structure to that of beryl is clarified, and alkali ion positions in the beryl structure postulated. Alkalies have a pronounced effect on the optic properties of beryl, suggesting, by analogy, that anomalous variations in the optic properties of cordierite are due to variable alkali content. The suggestion is confirmed by graphical presentations of the optic properties of analyzed cordierites.

Introduction

Eight occurrences of optically positive cordierite are reported in the literature (4, 5, 10, 11, 13, 15, 16, 20). No such cordierite has been synthesized and at the time of Shibata's research (17), relating the optic sign to ferrous iron content, data regarding this optically anomalous group of cordierites were not available.

Cordierite from a new locality, analyzed by the writer (Table 1), is optically positive. This transparent gem-quality cordierite occurs north of Great Slave Lake in an injection and assimilation gneiss of the Scottish highlands type (14). The associated minerals are: garnet, sillimanite, green spinel, graphite, biotite, orthoclase, microcline and oligoclase feldspar, quartz and tourmaline. Single prismatic crystals of the cordierite attain a size of 4 by 2 by 2 inches. These crystals are fractured but yield many fragments of gem size and quality.

If the analysis of this cordierite and three other analyses of optically positive cordierite, for which there are reasonably complete chemical, optical and physical data, are plotted on the Shibata (Winchell) variation diagram (21), the following points are noteworthy (Fig. 1):

(1) Optically positive cordierites vary widely in ferrous iron content (from 15.7 to 44.0 per cent of the $Fe_2Al_4Si_5O_{18}$ molecule).

(2) These cordierites with optic angles greater than 90° occur over the range of iron content over which Winchell postulates both a maximum (83°) and minimum (43°) point on the 2V curve.

(3) The three indices of refraction of all optically positive cordierites are lower than the respective indices of optically negative cordierites of the same Fe/Mg ratio.

(4) The birefringence of optically positive cordierite is greater than that of optically negative cordierite of similar Fe/Mg ratio.
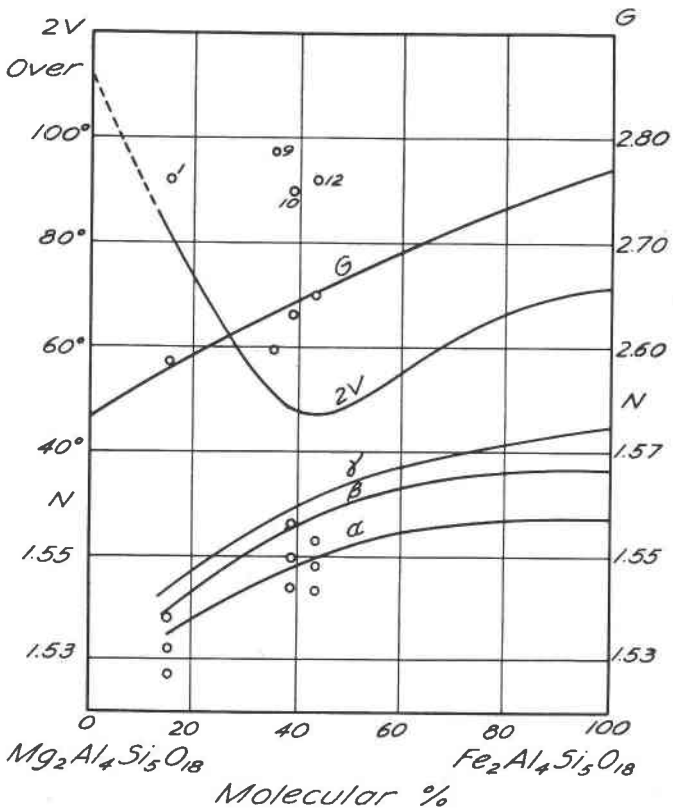
VARIATION DIAGRAM FOR CORDIERITE
(After Winchell)

FIG. 1. Optically positive cordierites (Table 4) plotted on the Shibata-Winchell variation diagram. The curves are Winchell's (21), the data from which the curves were derived are omitted.

TABLE 1. CHEMICAL ANALYSIS OF CORDIERITE FROM THE
GREAT SLAVE LAKE AREA

	Weight Per cent	Molecular Proportions
SiO_2	48.19	802
Al_2O_3	33.45	328
Fe_2O_3	0.55	3
FeO	8.40	117
MgO	7.95	197
CaO	0.17	3
Na_2O	0.22	3
K_2O	0.02	0
H_2O+	0.67	37
H_2O-	0.01	0
TiO_2	0.01	0
Li_2O	Trace	0
MnO	0.18	3

99.82

Analyst—R. E. Folinsbee

MOLECULAR COMPOSITION USING WINCHELL'S FORMULA (21)

	RO	Al_2O_3	SiO_2
MgO	98.5	197	492.5
FeO	58.5	117	292.5
MnO	1.5	3	7.5
Fe_2O_3	3.0	6	15.0
Calculated	161.5	323	807.5
Actual	161.5	328	802

61.0% $Mg_2Al_4Si_5O_{18}$
39.0% $(Fe, Mn)_2Al_4Si_5O_{18}$

It is clear that Winchell, following Shibata, in basing a variation diagram on the Fe/Mg ratio of cordierite, was forced to cull anomalous determinations. To quote Winchell (21):

The lack of correlation between chemical composition and optic properties is illustrated by the fact that there are almost as many (seven) samples of cordierite which have been analyzed and measured optically that cannot be used in the diagram as the number (eight) used in preparing it. . . . But it seems that some other factor (besides Fe + Mn) has a considerable influence on the optic angle.

The writer will endeavor to account for the variable optic angle and sign of cordierite on the basis of a hitherto undiscussed chemical constituent—the alkali content. The problem is approached first by an analogy based on structural relationships, then by inductive reasoning based on experimental evidence.

STRUCTURAL RELATIONSHIP OF CORDIERITE AND BERYL

The pseudo-hexagonal orthorhombic mineral cordierite is closely related to the hexagonal mineral beryl in atomic structure. The existence of this relationship, suggested by Gossner and Mussgnug (8, 9) on the basis of x-ray studies, is confirmed by the similarities in the chemical, crystallographic, optical, and physical properties of these two minerals.

The structural formulas are similar, though the relation involves the acceptance of an unusual type of substitution.

Beryl	Be_3	Al_2	Si_6	O_{18}
Cordierite	Mg_2Al	Al_2	Si_5Al	O_{18}
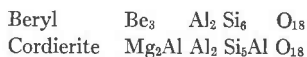
Translating freely from Gossner and Mussgnug (8, 9):

It is well known that the simple substitution of $2Be = 2Mg$ would ordinarily involve an essential change in the structure, since the ionic radius of beryllium is .34 Å while that of magnesium is .78 Å. However, for the two silicates cordierite and beryl, we have a special case where the ions Be and Mg, as constituents of a larger space group, can replace each other with no essential change in the structure.

The unit cell dimensions as given by Gossner (8) are comparable:

	Cordierite		Beryl	
Axis				Axis
$a' = b$	17.10 Å		16.0 Å	$(= a\sqrt{3})$
$b' = a$	9.78 Å		9.21 Å	$(= a)$
c	9.33 Å		9.17 Å	$(= c)$

Analogies in the chemical composition of the two minerals, as observed by the writer, are even more pronounced. In cordierite the H_2O+ content ranges from .50 to 3.72%, with the average of 1.50% H_2O+; in beryl it ranges from .37 to 3.00 with the average of 1.5%. The amount of water (or of OH ions) seems to have only a slight effect (19) on the density or optical properties, and to date its structural position has been ignored. It is the writer's opinion that the amount of water in these two minerals is an expression of the degree of alteration of the sample, and that the H_2O+ content forms a good basis for judging the purity and freedom from alteration of an analyzed sample (7, 12, 19).

The H_2O+ content of the Great Slave Lake cordierite is very low (0.67%). There is only one out of fifty analyses reported in the literature in which the water content was lower (7). This was also a gem cordierite (from Ceylon). Special care was taken by the writer in selecting the sample for analysis. A gem fragment was crushed, then hand picked. The fragments were checked under the microscope in oil of index 1.55 in order to ensure freedom from pinite (chlorite), and other alteration products or inclusions. An H_2O+ determination made on the reject

proved to be appreciably higher (0.95% H_2O+) than that of the analyzed portion (0.67% H_2O+), though to the unassisted eye the fragments appeared to be of comparable purity.
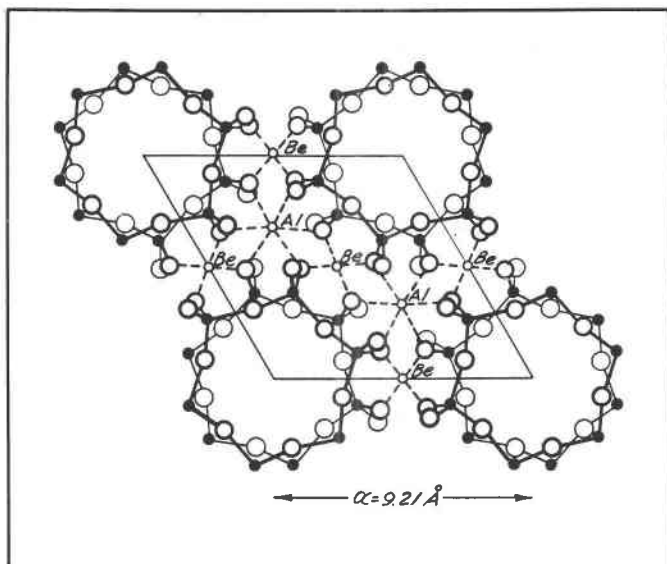

FIG. 2. The structure of beryl after Bragg and West (2, 3). The open hexagonal channels are the most striking feature of the structure.

The beryl-cordierite structure can accommodate variable amounts of the alkali ions Li, Na, and K (beryl may also contain Rb and Cs). The recorded range in alkali content is similar in the two minerals:

Beryl . . . 0.4 to 7.8 weight per cent alkalies (from 20 analyses).
Cordierite . . . 0.1 to 4.4 weight per cent alkalies (all recorded analyses).

The crystal habit of the two minerals is somewhat similar. Beryl occurs commonly as hexagonal prisms, cordierite as orthorhombic crystals cyclically twinned to a pseudo-hexagonal prism. Some beryl is orthorhombic and occurs as pseudo-hexagonal cyclic twins, identical with those of cordierite.

Beryl is normally uniaxial negative, but it may be biaxial with an optic angle (2E) up to 10°. Cordierite is usually biaxial with a wide range in optic angle. Tsuboi (20) reports a cordierite varying in optic angle from uniaxial positive to uniaxial negative within one crystal. Artificial iron-free cordierite may be uniaxial negative, like beryl (6). The indices likewise vary over an analogous range. Both minerals are

dichroic; in beryl the dichroism is slight and variable, in most cordierite it is very strong.

In the preceding paragraphs establishing the relationship of cordierite and beryl, the alkali content was shown to vary over a similar range. Since the alkali ions have a very significant effect on the optical properties of beryl (Fig. 3) and, as will be shown, on cordierite, their position in the structure becomes a matter of some theoretical interest. The alkali ions have large ionic radii (Table 2), and, lithium and sodium excepted, cannot substitute for the smaller ions Be, Si, Al, Mg and Fe. It follows that the structure of beryl and cordierite must provide special accommodation for variable amounts of K, Rb and Cs (the larger alkali ions), without loss of structural stability.

TABLE 2. IONIC RADII OF THE ALKALI METALS

Li	0.78 Å
Na	0.98 Å
K	1.33 Å
Rb	1.49 Å
Cs	1.65 Å

The most striking features of the beryl structure are the hexagonal rings formed by six silicon oxygen tetrahedra (Fig. 2). To quote Bragg (3):

These rings are stacked on each other along the hexagonal axis forming a series of open channels, for no atomic centre is nearer than 2.55 Å to a hexagonal axis. The whole structure is like a honeycomb with the hexagonal axes passing down the centres of the cells and within walls of closely packed oxygen atoms. The open channels in the structure are very striking and suggest investigation into properties they might confer on the crystal.

Bragg does not enlarge on this statement or discuss the structural relations of the alkali ions in beryl. His formulated structure is that of pure $Be_3Al_2Si_6O_{18}$ and he ignores the fact that beryl may contain up to 7.80% alkalies by weight.

Since K, Rb and Cs have such large ionic radii, the only available positions in the beryl structure are the hexagonal channels which Bragg mentions.

The possibility that the alkalies are located in these channels and the probable nature of the replacement which would necessarily accompany the introduction of positively charged ions were investigated by the writer for an alkali rich beryl (Table 3).

TABLE 3. IONS PRESENT IN ONE UNIT CELL OF CUSTER CO. BERYL*
Analyst R. E. Stevens (18)

Si		24.0
Fe		0.0
Be		8.3
Al		8.9
Na	1.23	
Li	1.43	
K	0.11	3.0
Rb	0.02	
Cs	0.23	
Ca		0.0
Mg		0.1
P		0.0
OH		6.0

Theoretically pure beryl	Be_{12}	$Al_2Si_{24}O_{72}$
Actual ions in Custer Co. beryl	Be_9 $Alk._3$	$Al_1Al_8Si_{24}O_{72}$
Proposed structural formulas		
(a) Custer Co. beryl	$(Na,K,Rb,Cs)_1Be_9$ $(Li, Na)_2$ $Al_1Al_8Si_{24}O_{72}$	
(b) Alkali beryl in general	$(Alkalies)_xBe_{(12-2x)}(Li, Na)_{2x}Al_xAl_8Si_{24}O_{72}$	

* The unit cell of beryl contains 24 silicon ions (4 molecules of $Be_3Al_2Si_6O_{18}$). Assuming the silicon content of each unit cell to be fixed at 24, the ionic ratios derived from the original analysis are recalculated into the number of ions in the unit cell by dividing each ratio by the number of unit cells statistically present. The following general equation is applied in making the recalculation:

$$\text{Number of ions in unit cell} = \text{Ionic ratio} \times \frac{24 \text{ (number Si ions in unit cell)}}{1054 \text{ (the ionic ratio of Si)}}.$$

The nature of the substitution is therefore:

$$3 \text{ Alkali ions}^{+1} + 1 \text{ Al}^{+3} = 3 \text{ Be}^{+2}.$$
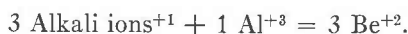
This substitution satisfies the structure electrostatically.

The probable position of the ions may be arrived at by comparing the cordierite structure with that of a conceivable alkali end member of the beryl series:

Pure beryl	Be_{12}	$Al_8 Si_{24}$	O_{72}
Alkali end member (Alkalies)$_4$	$(Li, Na)_8$ $Al_4 Al_8 Si_{24}$		O_{72}
Cordierite	$(Fe, Mg)_8 Al_4 Al_8 Si_{20} Al_4 O_{72}$		

In cordierite four of the aluminum ions take four of the beryllium positions of beryl. This applies also to aluminum in the alkali end member of the beryl series, four of the beryllium positions being filled by aluminum ions.

The substitution of 8 magnesium ions in cordierite for the remaining 8 beryllium positions as postulated by Gossner and Mussgnug (8, 9) has

an analogy in the alkali beryls. Magnesium has an ionic radius of .78 Å, identical with the ionic radius of lithium (.78 Å), slightly smaller than the ionic radius of sodium (.98 Å). Since magnesium (or iron) can take the position of the smaller ion beryllium without an essential change in the structure, it follows that 8 lithium and sodium ions can and do fill the remaining 8 beryllium positions of the alkali beryl.

The third alkali ion, needed for electrical stability in the alkali beryl structure, is in some of the unit cells cesium (1.65 Å), rubidium (1.49 Å) or potassium (1.33 Å), ions too large to substitute for beryllium. This third ion, then, must lie in one of the open hexagonal channels in the structure (Fig. 2).

It is felt that the substitution outlined above gives a very satisfactory explanation of the distribution of the alkali ions, and forms one more link in the chain of evidence relating the structures of beryl and cordierite.

EFFECT OF ALKALIES ON THE OPTICAL PROPERTIES OF BERYL

The intimate structural and optical relationship between cordierite and beryl has been established to validate an analogy between the effects of variable amounts of the alkalies on beryl and on cordierite.
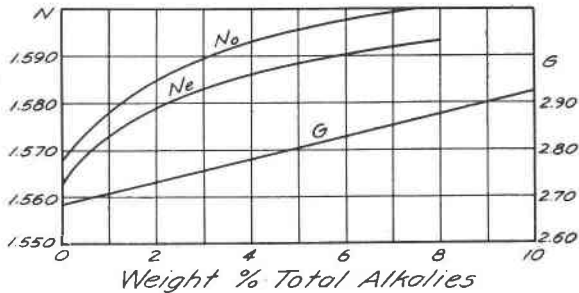

FIG. 3. The effect of alkalies on the optical and physical properties of beryl—after Winchell (22).

Figure 3 shows the effect of an increase in the alkali content on the properties of beryl, and from the figure inferences may be drawn as to the probable effect on cordierite.

An increase in alkali content in beryl is accompanied by:

(1) An increase in the indices.

(2) A change in the birefringence.

(3) The effect on the optical properties of any given increase in alkali content is greater for a low alkali beryl than for an alkali-rich beryl. It is not a straight line function like the increase in density with increasing alkalies.

EFFECT OF ALKALIES ON THE OPTICAL PROPERTIES OF CORDIERITE

By analogy with Fig. 3 an increase in alkali content in cordierite should:

(1) Increase the indices.
(2) Change the birefringence.
(3) Since cordierite is biaxial, change the optic angle.

The argument that the optical properties of cordierite are related in part to alkali content, to this point based on analogy, is confirmed by inductive reasoning based on actual analyses.

Unfortunately there are relatively few cordierite analyses in which the alkalies and calcium were determined. There are only fourteen complete analyses in the literature (Table 4) accompanied by sufficient optical data to use as evidence that the optical properties are determined in part by alkali and calcium content. The optical and chemical data are presented graphically in Figs. 4, 5, 6, 7, directly from this table.

TABLE 4. CORDIERITE—CHEMICAL ANALYSES AND OPTICAL PROPERTIES

Number	1	2	3	4	5
Author Occurrence	Pehrman Attu	Oppenheimer Twedestrand	Thiele Twedestrand	Thiele Orijarvi	Thiele Twedestrand
SiO_2	50.15	49.18	48.05	49.60	48.11
TiO_2	0.38	—	—	—	Trace
Al_2O_3	33.07	32.28	33.37	31.19	33.02
FeO	2.22	4.05	4.01	4.10	4.49
Fe_2O_3	1.52	0.08	0.09	1.19	0.81
MgO	11.01	11.25	11.11	11.13	10.95
MnO	0.12	—	Trace	—	0.07
Na_2O	0.14	0.21	0.26	0.21	0.12
K_2O	0.08	0.44	0.39	0.06	Trace
CaO	0.29	—	0.05	0.23	Trace
H_2O-	0.09	0.27	—	0.08	0.11
H_2O+	1.37	2.04	2.75	2.33	2.72
	100.44	99.80	100.08	100.22	100.16
% $Fe_2Al_4Si_5O_{18}$	15.7	17.0	17.1	20.4	21.2
Analyst	Pehrman	Dittrich	Thiele	Thiele	Thiele
Density	2.588	2.581	2.581	2.598	2.589
α	1.527	1.536	1.536	1.541	1.542
β	1.532	1.541	—	1.545	—
γ	1.538	1.543	1.543	1.547	1.548
2 V	92°	69° 26′	65° 36′	78° 12′	(74° 9′)
Optic sign	+	—	—	—	—

TABLE 4—*Continued*

Number	6	7	8	9	10
Author Occurrence	Thiele Bodenmais	Oppenheimer Haddam	Thiele Langdorf	Krishnan Madura	Folinsbee Great Slave
SiO_2	48.65	48.17	48.77	48.37	48.19
TiO_2	—	—	0.07	—	0.01
Al_2O_3	32.58	32.53	31.01	29.22	33.45
FeO	4.79	6.63	5.19	7.07	8.40
Fe_2O_3	1.75	1.21	2.83	2.20	0.55
MgO	10.02	8.86	8.91	9.54	7.95
MnO	—	—	0.31	0.42	0.18
Na_2O	0.15	1.39	0.16	—	0.22
K_2O	0.04	0.50	0.06	—	0.02
CaO	—	Trace	0.43	1.92	0.17
H_2O-	0.10	0.10	0.29	—	0.01
H_2O+	2.39	1.12	1.99	1.84	0.67
	100.26	100.51	100.02	100.58	99.82
% $Fe_2Al_4Si_5O_{18}$	26.2	32.3	33.3	35.6	39.0
Analyst	Thiele	Dittrich	Thiele	Krishnan	Folinsbee
Density	2.614	2.660	2.629	2.598	2.631
α	1.541	1.552	1.546	—	1.544
β	1.547	1.560	—	—	1.550
γ	1.550	1.561	1.558	—	1.556
2 V	(66° 24′)	43° 26′	—	85–99°	85–95°
Optic sign	—	—	—	±	±

TABLE 4—*Continued*

Number	11	12	13	14
Author Occurrence	Thiele Brocken	Pehrman Ilmajoki	Shibata Dôsi	Shibata Sasago
SiO_2	48.88	50.09	43.55	43.27
TiO_2	—	0.00	—	—
Al_2O_3	33.07	31.78	30.68	30.25
FeO	8.05	8.71	13.17	15.31
Fe_2O_3	0.53	0.78	1.50	1.09
MgO	7.04	6.69	2.70	1.48
MnO	0.06	0.00	2.88	1.20
Na_2O	0.09	0.00	0.32	0.90
K_2O	—	0.07	1.32	3.53
CaO	0.04	0.00	0.16	0.10
H_2O-	0.20	—	—	—
H_2O+	2.21	1.43	3.72	2.89
	100.17	99.55	100.00	100.00
$\%\ Fe_2Al_4Si_5O_{18}$	40.0	44.0	78.0	87.0
Analyst	Thiele	Pehrman	Tanaka	Tanaka
Density	2.647	2.650	2.72	2.75
α	1.549	1.543	1.555	1.558
β	(1.555)	1.548	1.566	1.568
γ	1.560	1.553	1.571	1.573
2 V	74° 36′	91° 29′	66°	68.5°
Optic sign	—	+	—	—
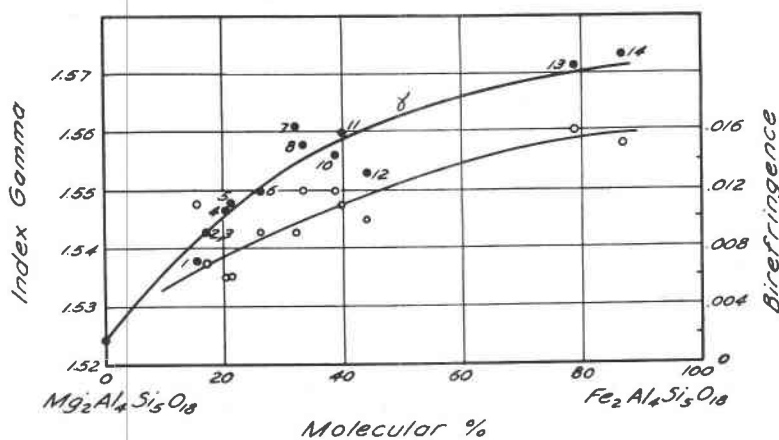
FIG. 4. The relation of index (upper curve) and birefringence (lower curve), in cordierite, to the Fe/Mg ratio.

Figure 4 shows clearly that an increase as in the Fe/Mg ratio increases the indices and birefringence of cordierite. The magnesian end member is a synthetic product (6).

The index of number 7 is anomalously high and it has a high alkali content. Numbers 1, 10, and 12 (anomalously low) are optically positive cordierite from the old shield areas. It is believed that their low index is a reflection of low alkali, high alkali-earth content.
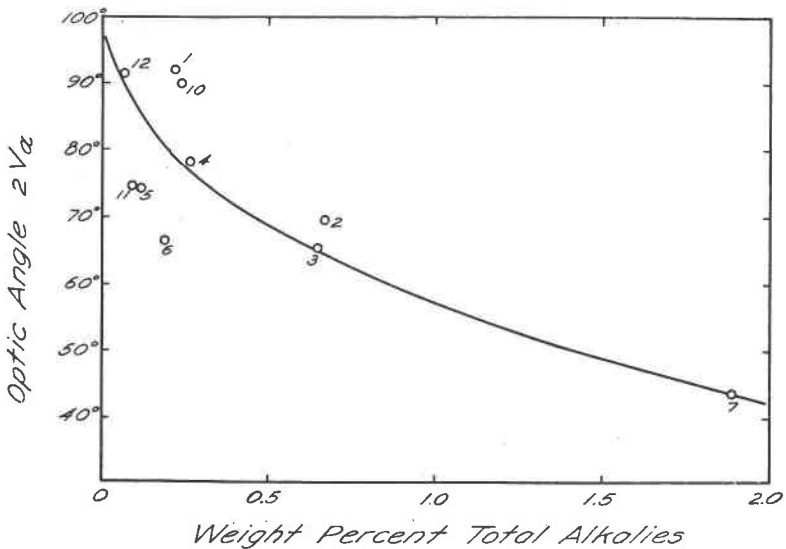

FIG. 5. The relation of optic angle in cordierite to the alkali content.

As indicated in Fig. 5 the optic angle in cordierite decreases with an increase in alkali content. Some of the points do not correspond closely with the curve, probably because of the effect of the variable CaO. Numbers 1 and 10 are high it is believed because of high CaO content. Numbers 11, 5 and 6 are low in CaO and therefore low in optic angle.

Numbers 13 and 14 (Table 4) are extremely iron-rich, alkali-rich cordierites. Their indices, birefringences and densities are very high, apparently related to the high iron content, possibly in part to the very high alkali content. They are not plotted on Figs. 5 or 7, and there are insufficient data to advance an explanation for their optic angles, other than that they belong to a distinct and rare group of the cordierite family.
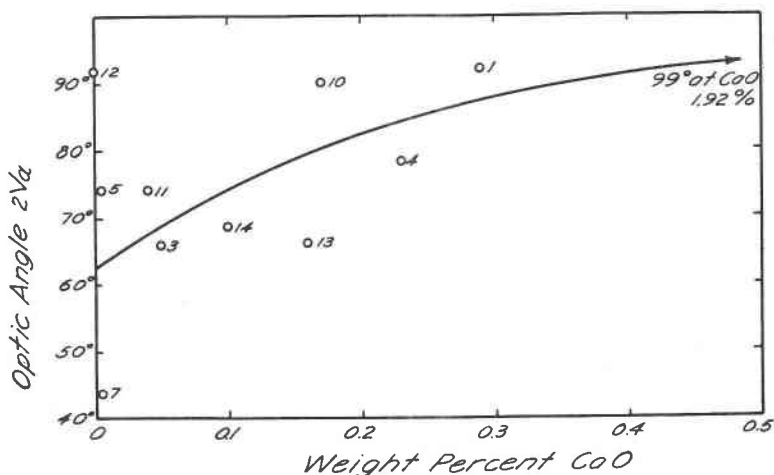
Fig. 6. The optic angle of cordierite tends to increase with an increase in CaO content.

An increase in the small amounts of CaO found in cordierite tends to increase the optic angle (Fig. 6). The anomalies arise from the subordination of the effect of CaO content to the effect of alkali content.
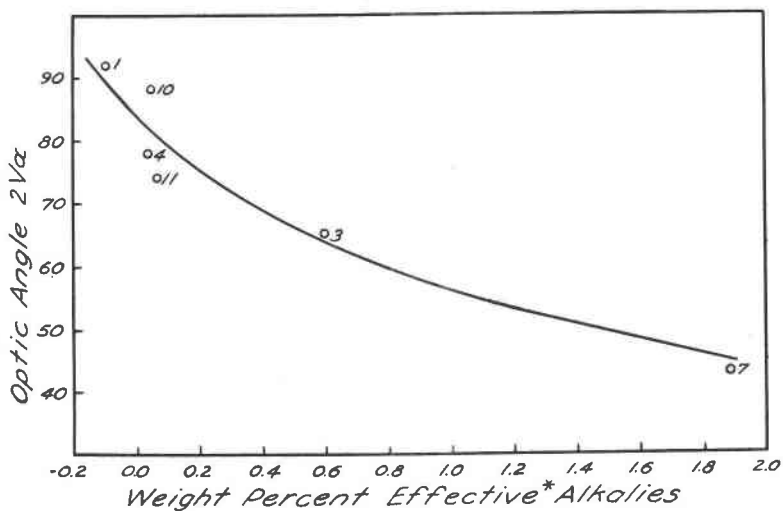

Fig. 7. The optic angle of cordierite plotted as a function of "effective" alkali content.

In Fig. 7 an attempt is made to eliminate the factor CaO in plotting the optic angle as a function of alkali content. The "effective" alkali content is arrived at by subtracting the weight percentage CaO from the weight percentage of total alkalies. Weight percentages are used in making this calculation rather than molecular percentages since optic properties in general are more nearly a function of density (weight percentage) than of number of ions (molecular percentage).

It is to be regretted that not all of the twelve analyses of Table 4 are complete. It is felt, however, that the graph establishes clearly the very close relation of effective alkali content and optic angle.

The effect of variables other than the per cent of $Fe_2Al_4Si_5O_{18}$, the alkalies, and the alkali earths should be considered. Thiele (19) in a recent paper presents seven new analyses and gives a very complete discussion of two variables hitherto unrecognized.

He is of the opinion:

(a) That an increase in water content affects the optic properties slightly, decreasing the indices and increasing the optic angle.

(b) That there may be an increase in the molecular percentage of silicon and magnesium as compared to aluminum due to isomorphous substitution of silicon and magnesium for aluminum.

$$Si^{+4} + Mg^{+2} = Al^{+3} + Al^{+3}$$
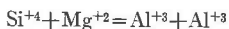
(c) That this substitution results in a decrease in indices and a sharp increase in the optic angle.

The writer cannot agree with these inferences of Thiele's after critical examination of the data upon which they were based.

Summary and Conclusions

The significant points brought out in this study of gem cordierite from the Great Slave Lake area are:

(1) A complete chemical analysis of cordierite from a new occurrence, especially significant since the analysis was made on selected fragments of exceptionally pure material, indicates that pure cordierite is very low in H_2O+.

(2) The analogy of the cordierite structure with that of beryl, as suggested by Gossner and Mussgnug on the basis of x-ray investigation is clarified by showing the close relationship of cordierite and beryl chemically, optically, and structurally.

(3) The nature of the isomorphous substitution accompanying the introduction of alkalies into the beryl structure is established for an alkali-rich beryl and confirmed by examination of the relation of the structural formulas of alkali beryl and cordierite.

(4) The Great Slave Lake cordierite is optically positive in character. The existence of optically positive cordierite has been questioned but the present results are carefully checked. A possible explanation of this anomaly is offered on the basis of a hitherto undiscussed variable in the chemical composition of cordierite—the alkali content.

(5) Alkalies have a pronounced effect on the optical properties of beryl, increasing the indices and birefringence. The variation in indices and birefringence of cordierite have previously been considered to be functions only of the Fe/Mg ratio (17, 21). The anomalies arising from this assumption are shown to be due to variations in the alkali content, the analogy between cordierite and beryl explaining the effects of this variable.

(6) An increase in the alkali content of cordierite is shown to:
> Increase the indices.
>
> Decrease the birefringence.
>
> Lower the optic angle.

Optically positive cordierites are in general a group of cordierites comparatively low in alkalies and high in calcium.

This paper emphasizes the importance of complete chemical analyses. In making analyses all the common elements should be determined, even if present in small amounts. Rare elements also, whose presence might be indicated on the basis of mineral structure, should be determined. The ideal formula, arrived at by structural investigation, will then form a basis for distributing the atoms as determined by the chemical analysis (2).

Complete analyses are of enduring value in solving problems of mineral chemistry, optics, and physics. They solve not only an immediate problem, but may find useful application in future investigations.

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