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VARIATIONS IN THE DELTA INDEX OF CORDIERITE AROUND THE CUPSUPTIC PLUTON, WEST-CENTRAL MAINE[1]

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Abstract

The Δ-index of 52 specimens of cordierite from an aluminous hornfels increases from 0.23 to 0.29 inward toward a quartz monzonite pluton. The MgO/(MgO+FeO) ratio in cordierite from the assemblage, cordierite-biotite-Al silicate-muscovite-quartz-plagioclase-magnetite±ilmenite±pyrrhotite±graphite(?), does not vary significantly toward the intrusive body. The Δ-index is independent of slight changes in MgO/(MgO+FeO) ratio and in the $Al_2O_3/(Al_2O_3+SiO_2)$ ratio.

Beryllium content in five samples ranges from 7 to 15 ppm and varies inversely with the Δ-index. The minor variations in beryllium content may reflect the ability of various structural states of cordierite to accommodate beryllium. Local variations in the Δ-index are related in a qualitative way to variations in the thermal environment, (1) near a large dike, and (2) at one corner of the intrusive body.

Introduction

In recent years considerable research has been done on the polymorphism of natural and synthetic cordierite. Much of this work stems from the investigations of Miyashiro and others (1955) and Miyashiro (1957), who found that cordierite forms a hexagonal polymorph and a continuous series of orthohombic pseudohexagonal forms through slight distortion of the hexagonal structure. Miyashiro proposed a distortion index of the type:

$$\Delta = 2\theta_{131} - \frac{(2\theta_{511} + 2\theta_{421})}{2} \qquad (\text{CuK}_{\alpha 1} \text{ radiation})$$

to relate the separation of three peaks on the X-ray powder diffraction pattern of the orthorhombic forms to the degree of distortion from the hexagonal end member (Δ=0.0). On the basis of Miyashiro's distortion index, Schreyer and Schairer (1961) subdivided alleged structural states of synthetic Mg-cordierite into high cordierite (Δ=0.0), intermediate-state cordierites (0.0<Δ<0.20) and low cordierite (0.20<Δ<0.30).

Meagher and Gibbs (1966, 1967) and Gibbs (1966) have shown that the polymorphic transformation involves four Al and five Si atoms in a tetrahedral framework structure of $Al_4Si_5O_{18}$ composition and may involve disordering between octahedral Mg and tetrahedral Al atoms at high temperatures. Analyses of the crystal structure, however, have cast serious doubt on the Δ-index as a reliable measure of the structural state

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of cordierite. Meagher and Gibbs (1966) found that hexagonal cordierite from the Bokaro coal field in India, thought to be completely disordered because of its Δ-index of 0.0 was, in fact, partly ordered. Pegmatitic cordierite from Haddam, Connecticut, with a Δ-index of 0.12 suggesting an intermediate state of order, had a completely ordered framework structure the same as cordierite from Guilford, Conn., with a Δ-index of 0.24 (Gibbs, 1966). Newton (1966) suggested that the apparently anomalous Δ-index values may be due in part to rather large amounts of BeO ($\approx$0.5 weight percent) in the cordierite from Haddam, Connecticut.

We have found, however, that variations in the Δ-index of cordierite from a contact aureole around a quartz monzonite stock are more closely related, at least qualitatively, to variations in the thermal history of the environment rather than to variations in the bulk chemistry of the cordierite.

Geologic Setting

The geology of the Cupsuptic quadrangle has been reported in detail by Harwood (1966) and summarized by Harwood and Berry (1967); therefore, only a brief description will be given here. Green and locally purple and black slate interbedded with feldspathic quartzite form the Albee Formation of Middle Ordovician age (Billings, 1937). The Albee is overlain by sulfidic black slate, greenstone, and graywacke of the Dixville Formation of late Middle Ordovician age (Green, 1964). The pre-Silurian rocks are unconformably overlain by three widely separated patches of conglomerate, limestone, and thin-bedded gray slate and feldspathic quartzite of Silurian and Devonian age.

All these Paleozoic rocks are in the chlorite zone of regional metamorphism. The argillaceous beds consist of quartz, muscovite, chlorite, albite, magnetite, zircon, and tourmaline; hematite is present in the purple slate, and pyrite and carbonaceous material, in the black slate.

Three small stocks of nonfoliated, equigranular to subporphyritic quartz monzonite, composed of quartz, oligoclase, microcline perthite, biotite, muscovite, magnetite, zircon, and apatite intrude the pre-Silurian rocks and generally form a maculose cordierite-andalusite-biotite hornfels. Adjacent to the igneous contact, however, a very fine-grained, equigranular, cordierite-sillimanite-biotite hornfels is formed. Along the sharp and vertical or steeply dipping contact between the hornfels and the quartz monzonite, the igneous rock is locally chilled to a fine-grained porphyritic facies as much as ten feet thick. Although the biotite isograd cannot be mapped continuously due to glacial overburden, all the contact aureoles are essentially conformable to the shape of the intrusive bodies and are separated from each other by slate and phyllite of chlorite grade.

All the Δ-index determinations, mineral assemblages, and chemical data reported herein are from cordierite-bearing rocks in the contact aureole around the Cupsuptic pluton of Middle or Late Devonian age (Fig. 1).

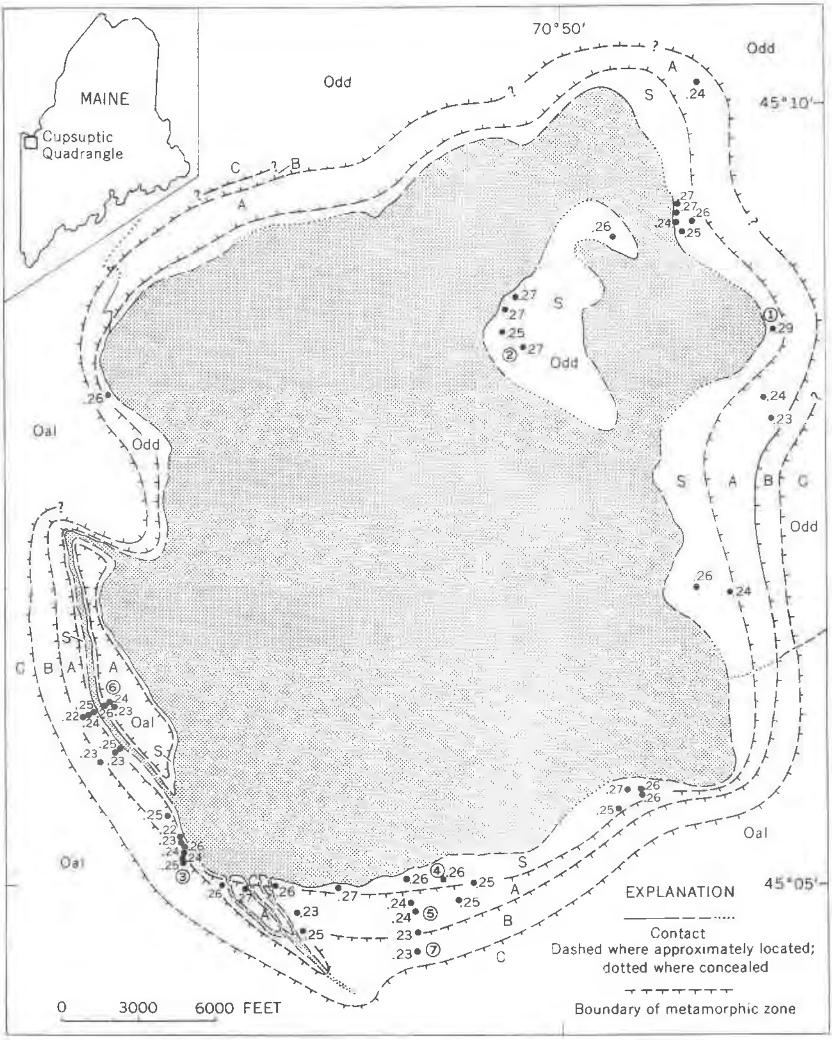

FIG. 1. Generalized geologic map of the Cupsuptic pluton (shaded) showing distribution of Δ-index values of cordierite. Oal=Albee Formation, Odd=Dixville Formation. Hachured lines separate metamorphic zones: S=sillimanite; A=andalusite; B=biotite; C=chlorite. Numbered localities correspond to samples in table 1.

Cordierite

Petrography and association. Cordierite porphyroblasts as much as 3.0 mm in diameter make up as much as 15 modal percent of the contact-metamorphosed slate of the Albee and Dixville Formations. The porphyroblasts are commonly black or bluish black and ovoid and weather to rusty-tan pits.

Microscopically, cordierite in the biotite zone and outer part of the andalusite zone appears as rounded, partly to completely altered, light and dark knots set in a very fine grained matrix of muscovite, quartz, albite, biotite, and chlorite. Some cordierite has faint sector twinning, yellow pleochroic haloes around zircon inclusions, and countless very fine grained inclusions of magnetite, quartz, plagioclase, and muscovite. Within the inner part of the andalusite zone, cordierite is generally fresh and has pronounced sector twinning. Twinning parallel to (110) is locally pronounced, and inclusions similar to those indicated above are abundant.

In the contact aureoles, the progressive textural change toward the igneous contact, from a maculose hornfels to an equigranular hornfels, is accompanied by progressive changes in the mineral assemblages, the most common of which are listed below in order. Each of the following assemblages contains cordierite, quartz, muscovite, albite/sodic oligoclase, biotite, zircon, and tourmaline in addition to the minerals listed; minerals in parentheses may or may not be present.

chlorite-(magnetite)-(ilmenite)-(pyrite)-(graphite)

andalusite-(chlorite)-(magnetite)-(ilmenite)-(pyrite)-(graphite)

sillimanite-(Mn-rich garnet)-(K-feldspar)-(magnetite)-(ilmenite)-(chlorite)-(pyrite)-(graphite)

Δ-index determination. Cordierite was concentrated to measure the Δ-index. Techniques were varied by trial and error to obtain a suitable concentrate but, in general, each sample was (1) ground and sieved to remove the $-80+100$-mesh fraction; (2) quartz, feldspar, and muscovite were separated from cordierite (with inclusions), biotite, and opaque minerals using a mixture of acetone and bromoform; and (3) cordierite with magnetite inclusions was separated from most of the biotite by a magnetic separator.

The Δ-index was determined from X-ray powder diffraction patterns using $CuK_{\alpha 1}$ radiation; scale-factor, multiplier, and time-constant settings of 4, 1, 1, respectively; a scanning speed of 1/4° per minute; and a chart speed of 1° per minute. The Geiger tube detector was set in oscillating mode and the region between 28.5° and 30.5° 2θ was scanned an average of six times. The Δ-index was determined to ±0.005° 2θ from the forward and backward scans measuring with a standard engineers scale and averaging the measurements. The standard deviation of the mean in each case is equal to or less than the uncertainty in reading the scale.

Distribution of the Δ-index. The distribution of the Δ-index of cordierite around the Cupsuptic pluton is shown in Figure 1 and is summarized below:

Metamorphic zone	Number of determinations	Δ-index range	Δ-index average
Sillimanite	28	0.24–0.29	0.26
Andalusite	20	0.22–0.25	0.24
Biotite	4	0.23–0.25	0.24

Although the total range of the Δ-index in this area is rather small, falling within the range of low cordierite ($0.20 < \Delta < 0.30$) of Schreyer and Schairer (1961), there is, in general, a progressive increase in the values toward the igneous contact as predicted by Schreyer (1966, p. 241, no. 2). The apparent reversal in this trend at the southwest corner of the pluton, near sample 3, is believed related to a large dike that is largely covered by glacial overburden.

A plot of the Δ-index values versus the map distance to the contact of the main intrusive body, shown by open circles in Figure 2, indicates a trend of increasing Δ-index as the contact is approached. A similar trend is more clearly shown by the solid circles in Figure 2 which represent nine samples plotted against map distance to the large dike near sample 6 on the west side of the pluton (Fig. 1). The Δ-index for sample 6 plots equally well against the map distance to both the main body (Fig. 2, no. 6) and the dike (Fig. 2, no. 6′).

Using the curve established for the Δ-index of cordierite adjacent to the dike as a model, we have sketched a hypothetical dashed curve through the scattered values for the main aureole. If we assume that the Δ-index values should lie on a smooth curve, such as the one drawn for the main aureole (Fig. 2) then values lying above the dashed curve represent samples that are, in effect, too far away from the igneous contact. A reasonable explanation for these points is that the shape of the igneous body may vary at depth, so that the map distance is not the shortest distance to the igneous contact. Conversely, the open circles below the dashed curve are, in effect, too close to the igneous contact if the above assumption holds. The position of these values cannot be improved by

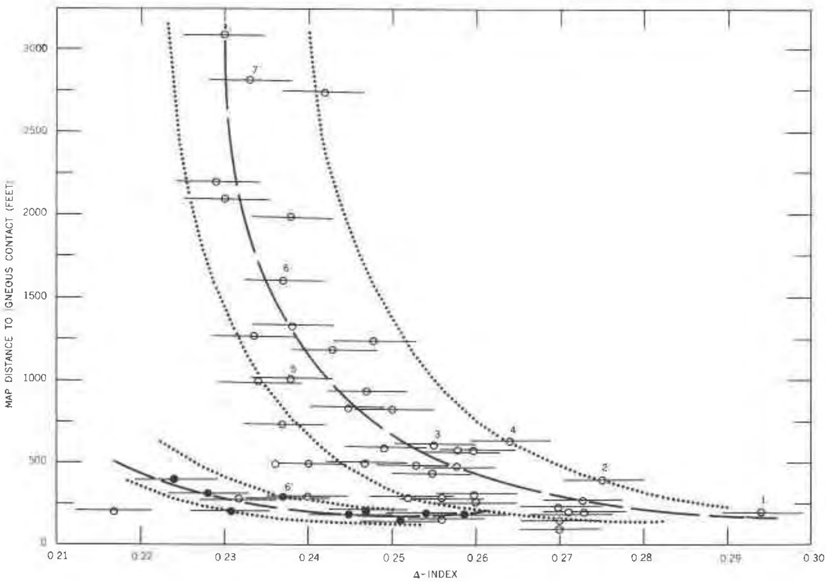

FIG. 2. Graph showing the relation between Δ-index of cordierite and map distance of the samples to the igneous contact; open circles represent samples influenced by the thermal history of the Cupsuptic pluton; solid circles represent samples affected by the dike on west side of pluton. Dotted lines outline trends of scattered points; dashed lines approximately bisect trends. Numbered points correspond to samples in table 1, and numbers 6 and 6′ represent same sample plotted against distance to main pluton and to dike, respectively.

varying the dip of the igneous contact, however, and there is no indication that the rocks have been faulted against the southwest corner of the quartz monzonite near sample 3 (Fig. 1), where most of the apparently anomalous values are found. These Δ-index values may reflect a very irregular thermal environment produced by the small and possibly overlapping thermal aureoles adjacent to the dikes at the southwest corner of the pluton.

In addition to local thermal anomalies, the overall shape of the main intrusive body probably influenced the apparently anomalous Δ-index values. Jaeger (1964) has shown in a simple heat-flow model that, disregarding latent heat, the contact temperature (T_c) is related to the shape of the contact as follows:

$$T_c = T_i \frac{\alpha}{2\pi}$$

where T_i is the temperature of the intrusive body and α is the interior angle measured between planar contacts. From this relation, it appears that the country rocks at the contact of the southwest corner of the Cupsuptic pluton (essentially a 90° outside corner) reached only 25 percent of the temperature of the intrusive body, whereas those at the contact along the essentially planar sides reached 50 percent of the temperature of the intrusive body. Thus, if the annealing history is proportional to the maximum contact temperature, the anomalous Δ-index values at the southwest corner of the pluton are apparently from cordierites that formed at a lower temperature and were not annealed to the same degree as most samples in the contact aureole.

Chemical variations. The above explanations for variations in the Δ-index are valid only if the Δ-index is independent of chemical variations in cordierite. The Δ-index of synthetic Mg-cordierites studied by Schreyer and Schairer (1961) varied over the known range (0.0 to 0.30), but data have not been presented to show whether rather small variations in the Δ-index of natural cordierite are independent of variations in the iron content.

To establish the relationship between the Δ-index and the MgO/(MgO+FeO) ratio, seven cordierite samples were analysed by Larson with the electron probe for MgO and total iron (reported as FeO). These data (Table 1, Fig. 3) show that cordierite from the assemblage, cordierite-biotite-Al silicate-muscovite-quartz-plagioclase-magnetite, does not vary significantly with distance to the intrusive body. The vertical regression line in Figure 3 was fit by the method of least squares using samples 1, 4, 5 and 6 (Table 1). The MgO/MgO+FeO ratio in samples 2, 3, and 7 in Table 1 cannot be compared to this trend because these cordierites are from different mineral assemblages.

A plot of the Δ-index versus the MgO/(MgO+FeO) ratio is given in Figure 4. Again, samples 1, 4, 5, and 6 are the only ones that are comparable, and these show the Δ-index to be essentially independent of variations in the iron content. A similar relation holds between these variables for cordierite from the contact aureole around the Onawa pluton in west-central Maine (Moore, 1960), as well as from high-grade gneisses in the Sturbridge area of Massachusetts (Barker, 1962; Paul Hess, 1967, written communications).

The analyses of cordierite compiled by Leake (1960) show that the aluminum content varies from 28.72 to 35.21 weight percent and the silicon content varies from 43.27 to 54.63 weight percent. A plot of the Δ-index versus the $Al_2O_3/(Al_2O_3+SiO_2)$ ratio, calculated from Table 1 of Leake (1960), is given in Figure 5. It is apparent from Figure 5 that the Δ-index is independent of variations in the $Al_2O_3/(Al_2O_3+SiO_2)$ ratio in cordierite.

TABLE 1.—CHEMICAL AND PHYSICAL PROPERTIES OF CORDIERITE FROM THE CUPSUPTIC AREA, MAINE

Sample	MgO (weight percent)	FeO[a] (weight percent)	MgO/(MgO+FeO) (mol proportion)	Δ-index[b]	Distance to igneous contact (feet)	Mineral Assemblage[c]
1	6.0	8.5	0.59	0.29 (4)	200	sil-cor-qtz-pla-bio-mus-pyr-gra.
2	6.5 6.5	13.0 12.2	.47 .48	.27 (5)	400	sil-cor-qtz-pla-ksp-mag-pyr-gra-(bio)-(and)-(mus).
3	8.5	4.5	.78	.25 (5)	600	cor-bio-mus-qtz-pla-gar-mag-ilm.
4	7.3 7.4	6.5(?) 8.5	.64(?) .60	.26(4)	650	sil-cor-mus-bio-qtz-pla-mag-ilm-(and).
5	6.3 7.3	9.3(?) 8.5	.55(?) .60	.23(8)	1,000	and-cor-bio-mus-qtz-pla-mag-ilm.
6	7.5	7.3	.64	.23 (6)	1,600 (300 to dike)	and-cor-bio-mus-qtz-pla-mag-ilm.
7	7.4	7.8	.62	.23 (3)	2,800	cor-bio-chl-qtz-mus-pla-mag-ilm.

[a] Total iron from electron-probe analyses reported as FeO.
[b] Numbers in parentheses are less precise, third-place figures.
[c] Mineral abbreviations: *sil*limanite, *and*alusite, *cor*dierite, *bio*tite, *mus*covite, *pla*gioclase, *gar*net, *chl*orite, *mag*netite, *ilm*enite, *gra*phite, *pyr*rhotite, qtz=quartz, ksp=potassium feldspar. Parentheses indicate mineral is present but surrounded or partly replaced by another phase or phases.

Variations in the minor element content of cordierite were determined by semiquantitative spectrographic analysis for five samples (Table 2). However, the analysed material was not clean cordierite, for it contained as much as 15 modal percent of the other mineral phases listed in Table 1. Therefore, the absolute values of the elements are meaningless and only changes in the relative amounts should be considered, and then not too rigorously because of the unequal amount of contaminants.

Beryllium is the only minor element in these 5 samples that shows any significant trend, and beryllium content is plotted against the Δ-index in Figure 6. Even if all the beryllium is in cordierite, the maximum con-

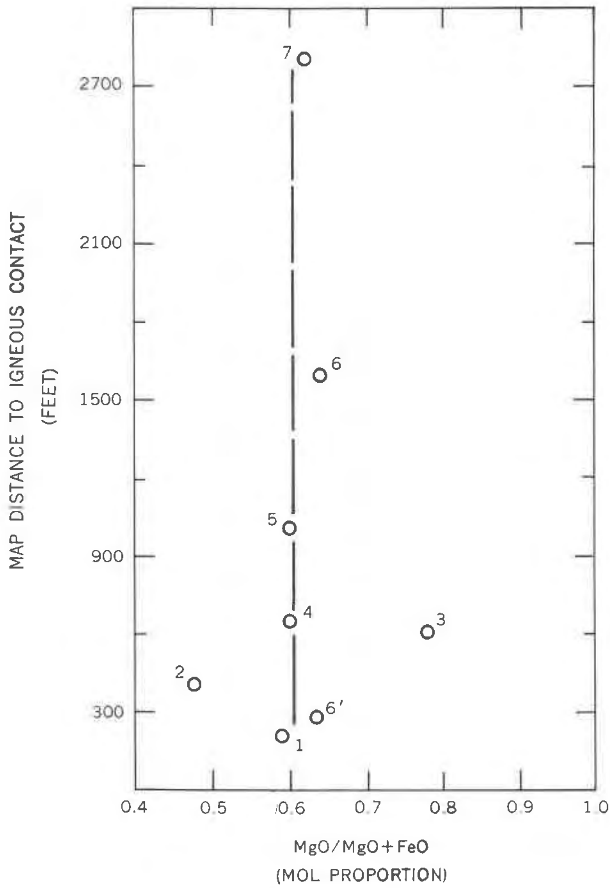

FIG. 3. Graph showing the relation between MgO/(MgO+FeO) ratio of cordierite and map distance of sample to igneous contact. Numbers correspond to samples in table 1, numbers 6 and 6′ represent same sample plotted against distance to main intrusive and to dike, respectively.

centration of 15 ppm and the change from 15 to 7 ppm is too small to affect the cordierite structure. Thus beryllium should have no affect on the spacing of reflections of the X-ray diffraction pattern used to define the Δ-index. The trend in Figure 6, however, may reflect the ability of various structural states to accommodate beryllium.

CONCLUSIONS

Within the contact aureole of the Cupsuptic pluton, the Δ-index of cordierite increases from about 0.23 to a maximum of about 0.29 as the

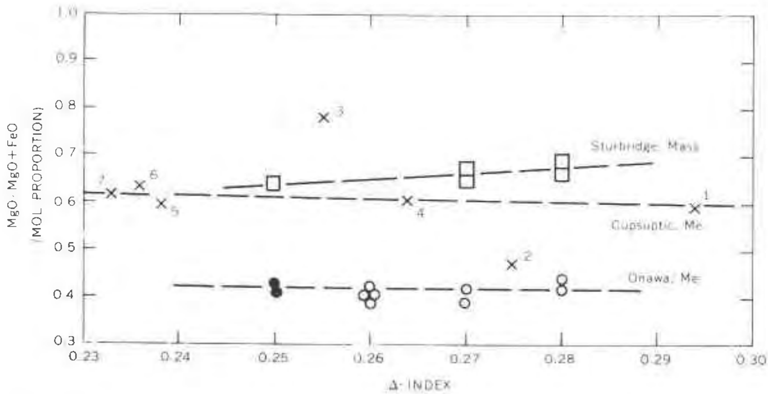

FIG. 4. Graph showing the relation between Δ-index and MgO/(MgO+FeO) ratio of cordierite. Numbered X's correspond to samples in table 1. Open circles represent cordierite-biotite-sillimanite-potassium feldspar-plagioclase-quartz assemblages; solid circles represent same assemblages with muscovite instead of potassium feldspar; squares represent cordierite-biotite-quartz-microcline-sillimanite-garnet-plagioclase assemblages. Data for Sturbridge, Mass., supplied by Paul Hess (1967, written communication), and data for Owana, Me., is from Moore (1960).

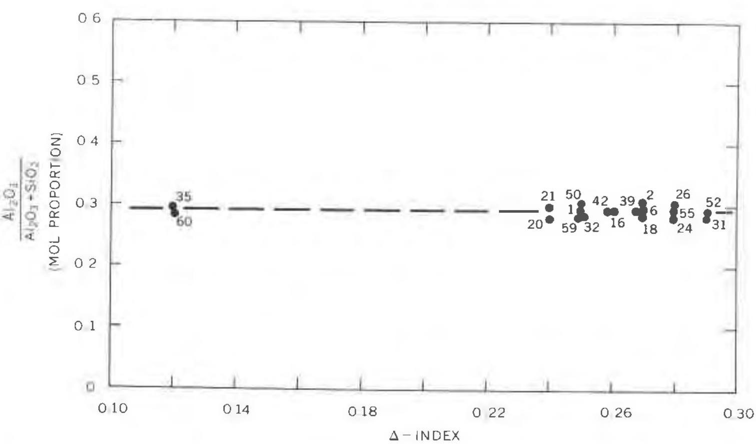

FIG. 5. Graph showing the relation between Δ-index and $Al_2O_3/(Al_2O_3+SiO_2)$ ratio of cordierite in analyses compiled by Leake (1960). Numbers refer to analyses in Table 1 of Leake. The Δ-indices were obtained from Leake (1960) and Schreyer (1966).

igneous contact is approached. The iron content of cordierite from a given assemblage of sufficient phases to eliminate variations due to changes in the bulk chemistry of the rocks varies slightly through the contact aureole but does not affect the Δ-index. The beryllium content of five cordierite samples decreases from 15 to 7 ppm with increasing Δ-index.

TABLE 2.—SEMIQUANTITATIVE SPECTROGRAPHIC ANALYSES[a] FOR MINOR ELEMENT CONTENT OF CORDIERITE FROM THE CONTACT AUREOLE AROUND THE CUPSUPTIC PLUTON, MAINE

Minor element	Sample[b]				
	2	3	4	6	7
B	0.0	0.003	0.0	0.003	0.003
Ba	.015	.02	.015	.02	.02
Be	.0007	.001	.0007	.0015	.0015
Ca	.2	.5	.3	.5	.3
Ce	.03	.03	.03	.03	.03
Co	.0015	.003	.003	.002	.002
Cr	.005	.01	.01	.01	.01
Cu	.003	.0005	.005	.003	.015
Ga	.0015	.002	.002	.002	.002
K	1.5	1.5	1.5	1.5	1.5
La	.01	.01	.01	.01	.01
Li	.0	.0	.0	.0	.02
Mn	.7	.7	.7	.5	.5
Mo	.0005	.005	.0003	.0005	.0003
Na	1.5	1.5	1.5	1.0	1.5
Nb	.0007	.001	.0007	.001	.001
Ni	.003	.007	.007	.005	.005
Sc	.001	.001	.001	.001	.001
Sr	.007	.005	.005	.007	.005
Ti	.5	.5	.5	.5	.5
V	.01	.015	.015	.01	.01
Y	.005	.007	.005	.007	.007
Yb	.005	.0007	.0005	.0007	.0007
Zr	.015	.03	.03	.03	.015

[a] Analyst: J. L. Harris. Results are reported in weight percent to the nearest number in the series 1, 0.7, 0.5, 0.3, 0.2, 0.15 and 0.1., etc., which represent approximate midpoints of group data on a geometric scale. The assigned group for semiquantitative results will include the quantitative value about 30 percent of the time.

[b] Sample numbers correspond to those in Table 1. Semiquantitative spectrographic analyses were run on cordierite concentrates that contained as much as 15 modal percent of the other mineral phases in the assemblages listed in Table 1.

We conclude, however, that such minor amounts of beryllium do not affect the Δ-index but instead may reflect the ability of various structural states of cordierite to accommodate beryllium.

Local variations in the Δ-index can be related qualitatively to variations in the thermal environment of the contact aureole, near a large dike and one corner of the igneous body.

Because changes in the Δ-index appear to be independent of chemical variations in this case and are related to the thermal environment, we

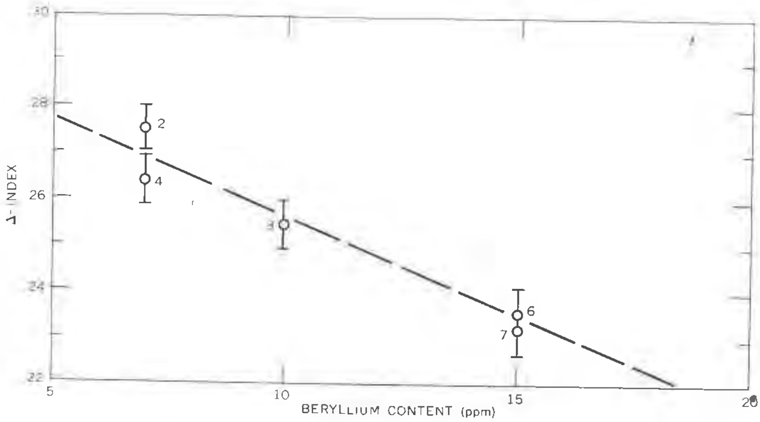

FIG. 6. Graph showing the inverse relation between beryllium content and Δ-index of cordierite, assuming beryllium is all in cordierite. Numbers refer to samples in table 1.

conclude that the Δ-index studied here is a measure of the Al-Si distribution in cordierite. Whether the cordierite studied here becomes more ordered or more disordered near the intrusive body cannot be unequivocally established by our present data. The progressive increase in the Δ-index suggests that cordierite becomes more ordered nearer the Cupsuptic pluton and thus shows an opposite trend to structural ordering in potassium feldspars around the Eldora stock, Colorado (Steiger and Hart, 1967; Wright, 1967). A study of the change in structural state of plagioclase from the Cupsuptic area is being initiated. If the feldspar becomes more ordered toward the pluton, the relation would suggest a higher water pressure or a longer annealing time, or both, for the contact aureole around the Cupsuptic pluton relative to that around the Eldora stock. If, on the other hand, the feldspar becomes more disordered toward the Cupsuptic pluton then (1) the cordierite may have formed initially as a metastable polymorph with a Δ-index lower than the annealed variety as concluded by Schreyer and Schairer (1961) and Schreyer (1966), or (2) the initial cordierite was in a stable structural state for a given temperature and became more ordered as it was annealed next to the pluton. There is no indication in this study, however, that the first cordierite to form was a hexagonal polymorph or even a cordierite with a Δ-index lower than 0.22.

Alternatively, if all of the beryllium is in the aluminum sites in this cordierite as appears to be the case in the pegmatitic cordierite studied by Cerny and Povondra (1966) and Meagher (1967), the smaller amounts of beryllium in cordierite next to the igneous body suggests that the cordierite becomes more disordered near the intrusive.

Acknowledgments

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