VARIATIONS IN THE DELTA INDEX OF CORDIERITE
AROUND THE CUPSUPTIC PLUTON,
WEST-CENTRAL MAINE¹

DAVID S. HARWOOD AND RICHARD R. LARSON, U. S. Geological
Survey, Washington D. C. 20242

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-alkali 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 Al₂O₃/(Al₂O₃+SiO₂) 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 orthorhombic pseudohexagonal forms through slight distortion of the hexagonal structure. Miyashiro proposed a distortion index of the type:

\[ \Delta = 2\theta_{131} - \frac{(2\theta_{511} + 2\theta_{131})}{2} \quad \text{(CuKα radiation)} \]

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 Schaier (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 Al₂Si₆O₁₈ 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.

¹ Publication authorized by the Director, U. S. Geological Survey.
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 (≈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 plagioclase, 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 CuKα 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 oscil-
lating 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:

<table>
<thead>
<tr>
<th>Metamorphic zone</th>
<th>Number of determinations</th>
<th>Δ-index range</th>
<th>Δ-index average</th>
</tr>
</thead>
<tbody>
<tr>
<td>Sillimanite</td>
<td>28</td>
<td>0.24-0.29</td>
<td>0.26</td>
</tr>
<tr>
<td>Andalusite</td>
<td>20</td>
<td>0.22-0.25</td>
<td>0.24</td>
</tr>
<tr>
<td>Biotite</td>
<td>4</td>
<td>0.23-0.25</td>
<td>0.24</td>
</tr>
</tbody>
</table>

Although the total range of the Δ-index in this area is rather small,
falling within the range of low cordierite (0.20 < Δ < 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 \( \alpha \) 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 Cup-
suptic 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 max-
imum contact temperature, the anomalous Δ-index values at the south-
west 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 regres-
sion 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 com-
parable, 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 plu-
ton 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 Al₂O₃/(Al₂O₃+SiO₂) 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 Al₂O₃/(Al₂O₃+SiO₂) ratio in
cordierite.
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-

### Table 1.—Chemical and Physical Properties of Cordierite from the Cusupctic Area, Maine

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

\(^a\) Total iron from electron-probe analyses reported as FeO.  
\(^b\) Numbers in parentheses are less precise, third-place figures.  
\(^c\) Mineral abbreviations: sillimanite, andalusite, cordierite, biotite, muscovite, plagioclase, garnet, chlorite, magnetite, ilmenite, graphite, pyrrhotite, qtz = quartz, ksp = potassium feldspar. Parentheses indicate mineral is present but surrounded or partly replaced by another phase or phases.
Concentration 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 $\Delta$-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 $\Delta$-index of cordierite increases from about 0.23 to a maximum of about 0.29 as the
Variations of Cordierite

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 Al₂O₃/(Al₂O₃ + SiO₂) 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.
We conclude, however, that such minor amounts of beryllium do not affect the A-index but instead may reflect the ability of various structural states of cordierite to accommodate beryllium.

Local variations in the A-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 A-index appear to be independent of chemical variations in this case and are related to the thermal environment, we
conclude that the $\Delta$-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 $\Delta$-index suggests that cordierite becomes more ordered nearer the Cupsuptic platon 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 platon, the relation would suggest a higher water pressure or a longer annealing time, or both, for the contact aureole around the Cupsuptic platon relative to that around the Eldora stock. If, on the other hand, the feldspar becomes more disordered toward the Cupsuptic platon then (1) the cordierite may have formed initially as a metastable polymorph with a $\Delta$-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 platon. There is no indication in this study, however, that the first cordierite to form was a hexagonal polymorph or even a cordierite with a $\Delta$-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

We are indebted to Professor John Moore of Carleton University and Mr. Paul Hess of Harvard University for allowing us to use their unpublished data and to Dr. Cornelis Klein of Harvard for making some earlier determinations of the iron-magnesium ratios and for supplying the probe standards used here.

Many helpful discussions with colleagues of the U. S. Geological Survey and Professor J. B. Thompson of Harvard are gratefully acknowledged. We also thank Louis Pavlides and J. J. Papke, U. S. Geological Survey, for reviewing the manuscript and making numerous suggestions for its improvement.

REFERENCES


AND J. F. SCHRAIER (1961) Compositions and structural states of anhydrous Mg-cordierites; a re-investigation of the central part of the system MgO-Al₂O₃-SiO₂. J. Petrology, 2, 324–406.


Manuscript received, October 29, 1968; accepted for publication, February 27, 1969.