

# SYMMETRY, CRYSTAL STRUCTURE, POLYMORPHISM, CRYSTALLOGRAPHIC ORIEN- TATION AND AXIAL RATIO OF CORDIERITE -

## A literature review

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Cordierite was first defined as a separate mineral in the beginning of the 19th century. After initially being classified as hexagonal, its orthorhombic symmetry was established in 1824. About a century later it was found that a hexagonal modification of cordierite could exist after all: it could be synthesized in the laboratory. In 1955 the only natural occurrence of hexagonal cordierite found hitherto was described from a buchite from India. Orthorhombic cordierites appear not in one specific structure: a series of structures is possible, defined by a varying distortion from the hexagonal structure. This structural variation is dependent on a two-step ordering process of (Al, Si), and possibly (Al, Mg).

Some confusion prevails as to the crystallographic orientation of cordierite. Both  $a > b$  and  $b > a$  have been used. It is proposed to use  $b > a > c$  exclusively. Attention is drawn to the fact that two series of axial ratios are currently used, which differ appreciably. It is proposed to use those based on X-ray data.

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Cordierite,  $(\text{Mg, Fe, Mn})_2\text{Al}_4\text{Si}_5\text{O}_{18}$ , was first described by Herr Kammerath und Kammerjunker von Schlotheim (1801), under the name *spanisches Lazulith* (Hintze, 1897, p. 922); it was classified by von Schlotheim as "... entweder ein eigenes Fossil oder bloss eine merkwürdige Abänderung des Quarzes ...". Von Schlotheim's material originated from the Cabo de Gata region, SE Spain (there is some evidence that it came from the Cerro del Hoyazo, a small hill in the area, see Zeck, 1968, pp. 12-13). Material from the same region was mentioned a few years later by Professor A. G. Werner (Freiberg) as being a distinct mineral species under the name *Iolith*—from the Greek for "violet"—(Schneider, 1807, pp. 266-267; Leonhard

1809, p. 101). Cordier (1809) was the first to give a detailed description of the same material, naming it *dichroïte* (from the Greek for “two colours”). To honour his compatriot's contribution Lucas (1813, pp. 219–222) introduced the name *cordierite* for the mineral. From about 1810 onwards the mineral also became known from regions other than Cabo de Gata, albeit initially under different names, for instance as *Peliom* (from the Greek for “blue spot”) from Bodenmais (Germany) and as *Steinheilit* (after the then governor of Finland, General-Lieutenant Graf von Steinheil, “. . . einem wahrhaft wissenschaftlich gebildeten und ausserordentlich thätigen Manne . . .”, Pansner, 1815, p. 301) from Orijärvi (Finland), (Steffens, 1811, pp. 369–371; Häüy, 1822, pp. 5–14; and Hintze, 1897, pp. 922–923).

### Crystal structure

Until 1824 cordierite was described as hexagonal. Mohs (1824, pp. 366–368) who termed the mineral *prismatischer Quarz*, was the first to establish its orthorhombic symmetry (Tamnau, 1828, p. 495).

Gossner & Mussnug (1928) reported the first structural analysis of common natural cordierite; the orthorhombic (pseudo-hexagonal) symmetry was confirmed, the homotypical (isomorphic, isostructural) relation with

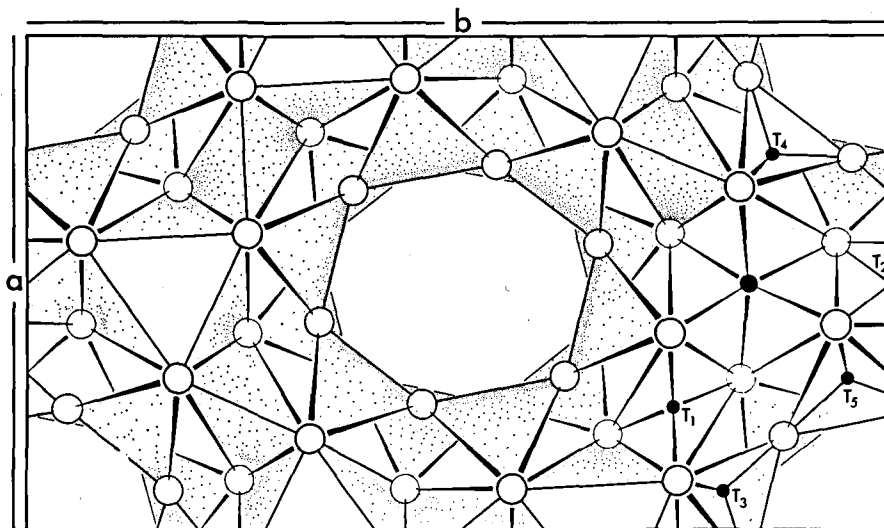


Fig. 1. The crystal structure of low cordierite viewed along the *c*-axis. The large open circles represent oxygen atoms, the large solid circle centering the octahedron represents the Mg,Fe atoms, the small circles centering  $T_1$  and  $T_5$  represent Al-rich sites and those centering  $T_2$ ,  $T_3$  and  $T_4$  Si-rich sites. After Gibbs (1966, fig. 4, p. 1080). Cell edges *a* and *b* added to conform with the  $b > a > c$  orientation (see pp. 43–45).

beryl was stressed and *Cmcm* was suggested as the space group. Byström (1942), and probably also Takane & Takeuchi (1936) in a paper composed in Japanese characters, redetermined the space group as *Cccm*, and gave a refined model for the crystal structure. Gibbs (1966) further refined the crystal structure model (see fig. 1); it was shown that all the Si and Al tetrahedra, both within and outside the hexagonal rings which figure in the structure, are involved in the order-disorder transformations and it was consequently concluded that cordierite should be classified with the framework silicates rather than with the ring silicates (see also Zoltai, 1960).

### Polymorphism

Experimental work on the  $\text{Al}_2\text{O}_3\text{-MgO-SiO}_2\text{-(-H}_2\text{O)}$  system showed that a hexagonal form of Mg-cordierite could be synthesized (Rankin & Merwin, 1917, p. 317: optically uniaxial negative crystals, no X-ray proof; Miyashiro & Iiyama, 1954, pp. 746-748, X-ray proof of hexagonal structure; and Schreyer & Schairer, 1961, p. 365, ditto, space group *P6/mcc*). A natural occurrence of hexagonal cordierite was reported by Miyashiro & Iiyama (1954, pp. 748-749) and Miyashiro, Iiyama, Yamasaki & Miyashiro (1955, pp. 193-194), (under the name indialite) from buchites (produced under the influence of adjacent burning coal seams) from the Bokaro coal field in India. Single crystal X-ray diffraction studies on Bokaro material by Meagher (1967, pp. 29-39) confirm that the material is hexagonal, with space group *P6/mcc*. Since the Bokaro find no well established hexagonal cordierite has been reported from elsewhere. An uncertain occurrence in the Allende meteorite (Mexico, 1969) has been pointed out by Fuchs (1969).

Later work by Miyashiro established the occurrence in natural rocks and experimental charges of cordierites, which, though all orthorhombic, show a varying degree of deviation from the hexagonal structure (Miyashiro, 1957). Schreyer & Schairer (1961) and Schreyer & Yoder (1964) further clarified these polymorphic relations. The latter authors explained the role of  $\text{H}_2\text{O}$  in the cordierite structure (op.cit. pp. 314-327) and it was shown that in some respects effects of (de)hydration had been misinterpreted by Miyashiro and his coworkers as being due to the polymorphic transition.

The structure of the orthorhombic form can be derived by a relatively slight distortion of the hexagonal structure. The degree of distortion is tied to a definite maximum. The variation in structure can be demonstrated in X-ray diffraction diagrams. As a means of measuring the deviation of the hexagonal structure, Miyashiro (1957, pp. 43-45) proposed the distortion index

$$\Delta = 2\theta_{311} - \frac{2\theta_{151} + 2\theta_{241}}{2}, \text{Cu K}\alpha_1\text{-radiation (indexing in } b > a > c \text{ orientation)}$$

$\Delta = 0^\circ$  for the hexagonal form, and has a maximum value of  $0.29^\circ$ – $0.31^\circ$  for any Fe/Mg ratio of the mineral. Details of the measuring procedure of  $\Delta$  have been discussed previously by Zeck (1969). In thin section the hexagonal form and the orthorhombic form are readily distinguished from each other by measuring the optical axial angle. The size of the axial angle and the value of  $\Delta$  are closely related.  $\Delta = 0^\circ$  corresponds to  $2V_x = 0^\circ$ , while, when  $\Delta$  increase to  $0.29^\circ$ – $0.31^\circ$ ,  $2V_x$  rises gradually to  $90^\circ$  or values close thereto (Miyashiro, 1957, p. 47; Schreyer & Yoder, 1961, p. 311; Meagher, 1967, p. 30). Schreyer & Schairer (1961, fig. 1) subdivided the polymorphic series in terms of  $\Delta$ -values: high cordierite ( $\Delta = 0.00^\circ$ ), intermediate state cordierite ( $0.00^\circ < \Delta \leq 0.20^\circ$ ) and low cordierite ( $0.20^\circ < \Delta$ ). This classification will not be used in the present paper. The terms high cordierite or hexagonal cordierite will be used for forms in which  $\Delta = 0.00^\circ$ , while all other forms ( $0.00^\circ < \Delta$ ) will be denoted as low or orthorhombic cordierite.

Application of experimental results concerning the  $\text{Al}_2\text{O}_3$ -MgO-SiO<sub>2</sub> (-H<sub>2</sub>O) system (Schreyer & Schairer, 1961, and Schreyer & Yoder, 1964) and additional, concurrent information on Fe-cordierite (Schreyer, 1965, p. 303; 1966, pp. 227–228) lead to the conclusion that the degree of  $\Delta$  in any cordierite would be mainly dependent on PT conditions during and subsequent to its growth. The experiments showed that the hexagonal form is formed first (in most cases metastably) under all PT conditions and that during prolonged heating, preferably at high T, the mineral turns orthorhombic and the distortion index increases. (Note, however, that for certain bulk compositions and  $P_{(\text{H}_2\text{O})}$ , the stability field of high cordierite can be reached at very high temperature, and inversion upon subsequent heating would not take place under these conditions; see Schreyer & Schairer, 1961, pp. 363–393, see also Schreyer & Yoder, 1964, pp. 310–314). The highest distortion indices may be expected to be found for cordierites from rocks that have endured the highest (but thus, under certain conditions not extreme) temperatures for the longest periods of time.

The increase in  $\Delta$ , from  $0.23^\circ$  to  $0.29^\circ$ , with increasing proximity to the Cupsupitic pluton (Maine, USA) in cordierites in its contact aureole (Harwood & Larson, 1969) is in agreement with such a model. The occurrence in the Hoyazo lava of cordierite of deeper origin with  $\Delta = 0.29^\circ$  ( $\pm 0.01^\circ$ ) and cordierite trillings precipitated from the volcanic melt with  $\Delta = 0.235^\circ$  ( $\pm 0.01^\circ$ ) can be explained accordingly (cf. Zeck, 1968, p. 146).

Miyashiro (1957, p. 45), Schreyer & Yoder (1964, p. 311) and Gibbs (1966, p. 1072) suggested that the variation in the cordierite structure reflects long range order-disorder relations of Al-Si (and perhaps Al-Mg). The disorder arrangement would produce high cordierite whereas an increase in ordering would result in an orthorhombic crystal structure. Meagher &

Gibbs (1966) found that the cordierite from Haddam (Connecticut, USA) with  $\Delta = 0.12^\circ$ , is completely ordered, and that the high cordierite ( $\Delta = 0.00^\circ$ ) from the Bokaro coal field in India has a 35 % Al-Si ordering. On the basis of this evidence it was suggested that "... the polymorphic transformation is not simple and direct"—the ordered Haddam form would then have  $\Delta \simeq 0.30^\circ$  and the Bokaro form  $\Delta \simeq 0.10^\circ$ —"but is a two step ordering process like that found in the igneous potassium feldspars" (Meagher & Gibbs, 1966, p. 108). Infra-red and powder X-ray diffraction studies by Langer & Schreyer (1969) confirmed this concept. These authors concluded that the structural transition may involve two different ordering processes: firstly, a short-range ordering process, (Al, Si) and possibly (Al, Mg), resulting in a domain structure, and including a marked increase in  $\Delta$  (up to about  $0.17^\circ$  in the synthetic Mg-cordierite studied by Langer and Schreyer), and subsequently, a long-range ordering process abolishing the domain structure and causing the final increase in  $\Delta$  to its maximum value.

It has been suggested that in some cases the  $\Delta$  value of a cordierite is not only a function of its ordering, but is also influenced by its chemical composition. Gibbs (1966, p. 1072), basing his argument on the findings of Newton (1966), suggested that the small distortion indices ( $\sim 0.1^\circ$ ) found for pegmatitic cordierites might be caused by their comparatively high Be content ( $\sim 0.5$  wt %). Harwood & Larson (1969) found that for cordierites from the contact aureole of the Cupsuptic pluton in Maine (USA) the variation in  $\Delta$  is independent of the MgO/(MgO + FeO) and  $\text{Al}_2\text{O}_3/(\text{Al}_2\text{O}_3 + \text{SiO}_2)$  ratios of the cordierites, but  $\Delta$  varies inversely with the Be content ( $\Delta$  ranges from  $0.27^\circ$  to  $0.23^\circ$  and Be ranges from 7 to 15 ppm concomitantly). However, the small variations in Be content are thought not to have caused the small variations in  $\Delta$ , but to reflect the fact that Be is more readily accommodated in cordierite with lower  $\Delta$  values.

### Crystallographic orientation

Two orientations were used at an early date to describe the mineral: Tamnau (1828) and Breithaupt (1832, p. 180; 1847, pp. 711–712) took  $b > a > c$ , and Brooke & Miller (1852, p. 325), interchanging the a and b axis, used  $a > b > c$ . The Tamnau and Breithaupt orientation found common acceptance in mineralogical articles and textbooks, see e.g. Des Cloizeaux (1862, pp. 354–364), Hintze (1897, pp. 918–943), Doelter (1917, p. 614), Gossner (1928), Winchell & Winchell (1959, p. 470), observing as it did the rule—initiated probably by C. S. Weiss and C. F. Naumann, the renowned 19th century German mineralogists (see also Zirkel, 1898, p. 93, and Klockmann, 1922, pp. 110–111)—that the b-axis was to be chosen as the longest of the horizontal axes (hence macro-axis). Strunz (1970, pp. 405–407; also earlier editions), who takes  $a > b$ , is an exception. However, in papers reporting X-ray results, the  $a > b > c$  orientation

was commonly used, see e.g. Gossner & Mussgnug (1928), Byström (1942), Gibbs (1966), Wyckoff (1968, p. 437). Wyckoff (1960, ch. 12, table p. 18) is an exception. Note that the difference between the two orientations lies solely in the interchanging of the *a* and *b* axes; the ratio between the axial units has been left unchanged.

In some mineralogical textbooks the unhappy situation is encountered that the classical  $b > a$  orientation is used to present the more “classical” crystallographic information such as the orientation of the indicatrix and indices of cleavage and twin planes, the  $a > b$  orientation being employed to present the X-ray data, such as cell edge dimensions and indices in X-ray diffractograms. This is the case in Deer, Howie & Zussman, 1962, first impression, pp. 268–299 (in the 2nd–5th impressions, 1963–1967, the cell edge dimensions are given in the  $b > a > c$  orientation on p. 268, but for the rest the situation is the same as in the first impression; in Deer, Howie & Zussman, 1966, the condensed version of this textbook, the  $b > a$  orientation is used throughout, except in their fig. 27) and in Ramdohr & Strunz (1967, p. 672). Even for the indices of crystal faces these latter authors used both orientations concurrently: fig. 523 (p. 672) is indexed for  $a > b$ , while in the corresponding text the same planes are indexed for the  $b > a$  orientation. (In the same work different sources for the numerical values of the axial ratio and the cell edge dimensions are used also, as though these were unrelated parameters: for the axial ratio the goniometer data of Brooke & Miller, 1852, were taken, while for the cell edges 20th century X-ray based values were used, which are essentially different, see pp. 45–46).

Both orientations ( $b > a$  and  $a > b$ ) are in principle equally well fitted to describe the mineral, but for practical reasons it is recommended that one and the same orientation be used to present all the various crystallographic parameters of the mineral, especially in the same text. Confronted therefore with the need to make a choice, the present author proposes to use

$$b > a > c$$

as it has priority, complies with a frequently observed rule and the use of the  $a > b$  orientation offers no advantage.

Strunz, Tennyson & Uebel (1970, 1971) and Strunz (1970, pp. 405–407; also earlier editions) proposed using the  $a > b > c$  orientation to bring out more clearly the structural similarities between cordierite and beryl. The present author does not consider this a sufficient reason for reversing an age old practice. Besides, as mentioned before, Gibbs (1966) showed that cordierite should be described as a tecto-silicate, and not, like beryl, as a soro-silicate (cyclo-silicate).

In fig. 1 the cell edges *a* and *b* are indicated in the crystal structure

display by Gibbs (1966). Correlation of optical directions and crystallographic axes for cordierite:  $a = Y$ ,  $b = Z$ ,  $c = X$ . Pleochroic scheme:  $X =$  faintly yellow,  $Z =$  faintly blue,  $Y =$  faintly blue, a shade darker than  $Z$ .

### Numerical axial ratio and cell edge data

(all data are given in the  $b > a > c$  orientation)

The earliest axial ratio data were based on measurements of interfacial angles of crystals. Some of the best known data are:

Breithaupt (1832, p. 180; 1847, pp. 711–712): (contact goniometer)	$a:b = 0.587:1$
Brooke & Miller (1852, p. 325): (reflection goniometer)	$a:b:c = 0.587:1:0.558$

A few of the interfacial angles on which the axial ratios are based:  $(110) \wedge (110) = 60^\circ 50'$  and  $(130) \wedge (\bar{1}30) = 120^\circ 50'$ . The Brooke & Miller data were taken over in numerous textbooks and papers, e.g. Des Cloizeaux (1862, pp. 354–364), Dana & Brush (1868, p. 299), Hintze (1897, pp. 918–943), Doelter (1917, p. 614), Venkatesh (1954, p. 636), Ramdohr & Strunz (1967, p. 672).

In more recent years it has become possible to calculate the axial ratio from cell edge dimensions determined by X-ray methods. Some of the thus derived values are given below; more are given by Iiyama (1956, p. 386). The axial ratio being related to  $\Delta$ , the respective values for this parameter are given with the examples stated below. The axial ratio by Breithaupt, and probably also the one by Brooke & Miller, refer to material from Bodenmais, Bavaria, Germany, which is known to have  $\Delta \simeq 0.26^\circ$ .

Gossner & Mussgnug (1928) ( $\Delta$ is probably about $0.26^\circ$ )	$a:b:c = 0.571:1:0.546$
Byström (1942) ( $\Delta = 0.25^\circ$ , Iiyama, 1956, p. 386)	$= 0.568:1:0.549$
Iiyama (1956, table 9, No. 13) ( $\Delta = 0.23_5^\circ$ )	$= 0.571:1:0.546$
Gibbs (1966) ( $\Delta = 0.24^\circ$ )	$= 0.570:1:0.546$

A clear difference appears to exist between the X-ray based data and the classical data based on geometrical measurements. One could suggest that the old measurements are correct and hence conclude that the crystals were misoriented in the X-ray investigations ( $[130]$  and  $[110]$  could be taken for the a-axis and b-axis, see Zeck, 1968, pp. 40–41). This possibility, however, may be disregarded since Gibbs (1966, p. 1073) specifically mentioned that

this pitfall was foreseen and that care was taken to avoid it. It is concluded therefore that the old goniometer measurements are incorrect, and that the X-ray based data should be used instead. This implies that the interfacial angles mentioned above become different:  $(110) \wedge (\bar{1}\bar{1}0) = 59^\circ 18'$  and  $(130) \wedge (\bar{1}\bar{3}0) = 119^\circ 18'$ .

This conclusion was reached earlier by Zeck (1968, pp. 39–41) and has since been sustained by Strunz et al. (1970, pp. 2–3; 1971, pp. 3–4). The discrepancy might be explained by misorientation of the crystal(s) having taken place during the measurements on the goniometer,  $\{110\}$  and  $\{130\}$  being taken for  $\{010\}$  and  $\{100\}$ , or perhaps some other more peculiar mistake was made (Zeck, 1968, p. 41). Strunz et al. (1971, p. 3) stated that it is “obviously caused by inaccuracies in morphologic angular measurements, and poor development of the crystal faces”.

It is surprising that the old axial ratio and interfacial angle data have been used for more than a century without their erroneous nature being discovered. This is all the more curious in papers written after 1928, as the X-ray based values were then available.

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## Dansk sammendrag

Cordierit defineredes som et separat mineral i begyndelsen af det 19. århundrede. Efter først at have været klassificeret som hexagonalt, blev dets rhombiske symmetri fastslået i 1824. Omtrent 100 år senere fandt man dog ud af, at en hexagonal modifikation af cordierit kunne eksistere alligevel: en sådan kunne syntetiseres i laboratoriet. I 1953 blev den indtil nu eneste naturlige forekomst af hexagonal cordierit beskrevet fra en buchit fra Indien. Rhombiske cordieriter fremtræder ikke med én specifik struktur: en serie af strukturer er mulige, som har en varierende deformation fra den hexagonale struktur. Denne strukturelle variation afhænger af en to-trins “ordering” proces af (Si, Al) og sandsynligvis (Al, Mg).

Der har hersket en del forvirring med hensyn til den krystallografiske orientering af cordierit. Både  $a > b$  og  $b > a$  har været brugt. Det foreslås udelukkende at anvende  $b > a > c$ . Endvidere gøres der opmærksom på, at man faktisk i dag opererer med to sæt af akse-forhold, som er mærkbart forskellige. Det foreslås at anvende de, der er baseret på røntgen-data.

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