Canadian Mineralogist
Vol. 15, pp. 43-49 (1977)

THE POLYMORPHISM OF CORDIERITE:
II. THE CRYSTAL STRUCTURE OF INDIALITE

E. P. MEAGHER
Department of Geological Sciences, University of British Columbia, Vancouver 8, B.C., Canada V6T 1W5

G. V. GIBBS
Department of Geological Sciences, Virginia Polytechnic Institute and State University, Blacksburg, Virginia 24061 U.S.A.

ABSTRACT

A least-squares refinement of type-locality indialite [$Mg_{1.40}Fe_{0.66}Al_{4.11}Si_{4.89}O_{18}$; *P6/mcc; a* 9.800(3), *c* 9.345(3)Å] shows the T_2 tetrahedra in the six-membered ring of the framework to be significantly smaller than the linking T_1 tetrahedra. By comparison of $\langle T\text{-O}\rangle$ data for cordierite the contents of these tetrahedra are estimated to be (T_1: 0.72 Al, 0.28 Si; T_2: 0.30 Al, 0.70 Si). Because the Al content of the T_1 tetrahedra differs significantly from 1.0, the order-disorder changes in cordierite involve all the tetrahedral atoms in the framework rather than just those in the six-membered ring. The degree of long-range order estimated with the Bragg-Williams equation is 0.38. The constraints of *P6/mcc* symmetry permit a range of intermediate structural states, and a domain structure is considered in terms of the Al, Si contents of the tetrahedra. Statistically featureless Fourier (F_o-F_c) maps indicate an absence of atoms in the structural channels parallel to the *c* axis.

SOMMAIRE

La structure de l'indialite du gisement type [$Mg_{1.40}Fe_{0.66}Al_{4.11}Si_{4.89}O_{18}$; *P6/mcc; a* 9.800(3), *c* 9.345(3)Å], affinée par moindres carrés, révèle que les tétraèdres T_2 des anneaux hexagonaux de la charpente sont nettement plus petits que les tétraèdres T_1 intercalaires. En comparant aux données $\langle T\text{-O}\rangle$ de la cordiérite, on estime la population des tétraèdres: 0.72Al, 0.28Si en T_1; 0.30Al, 0.70Si en T_2. Comme la teneur en Al des tétraèdres T_1 diffère notablement de 1.0, les changements ordre-désordre, dans la cordiérite, impliquent tous les atomes tétracoordonnés de la charpente et non pas seulement ceux des anneaux. Le degré d'ordre à longue distance, estimé au moyen de l'équation de Bragg-Williams, est égal à 0.38. Les contraintes imposées par le groupe spatial permettent toute une série d'états structuraux intermédiaires, et une structure en domaines est concevable en fonction de la population (Al,Si) des tétraèdres. Les séries différences (F_o-F_c), statistiquement sans relief, indiquent l'absence d'atomes dans les tunnels structuraux parallèles à l'axe *c*.

(Traduit par la Rédaction)

INTRODUCTION

The earliest quantitative information on the phase relations of cordierite was provided by a combined microscopic and thermal study of the system MgO-Al_2O_3-SiO_2 by Rankin & Merwin (1918) who discovered two forms of $Mg_2Al_4Si_5O_{18}$, referred to as α and μ forms. The μ form was considered to be a metastable product because, after crystallization from glass at temperatures below 950°C, it transformed irreversibly upon heating to the α form and because repeated attempts to transform the α form to the μ form at temperatures below 950°C failed. On the other hand, the α form was found to possess a well-defined stability field and crystallized as six-sided prisms. A conoscopic examination of this material yielded "nearly or quite uniaxial negative interference figures" indicating hexagonal or quasi-hexagonal symmetry. Although the paucity of data on natural cordierite precluded a definitive comparison between it and the α and μ forms, the physical properties of the α form were found to be similar to those of the natural material.

Crystallographic, optical and chemical data subsequently obtained for natural cordierite showed the mineral to be orthorhombic and to possess a crystal structure similar to that of hexagonal beryl (Fig. 1; Gossner & Mussgnug 1928; Takane & Takeuchi 1936; Byström 1942; Folinsbee 1941). A later study of the polymorphism of cordierite using optical and powder diffractometer data led Miyashiro & Iiyama (1954) to assert that the α form is truly hexagonal and therefore distinct from natural orthorhombic cordierite. A natural analogue of the α form was subsequently discovered in the fused sediments of the Bokaro coalfield, India, and named indialite (Miyashiro & Iiyama 1954). Although the structural relationship between indialite and cordierite was unknown, it was recognized that indialite is a hexagonal polymorph of Mg_2Al_4

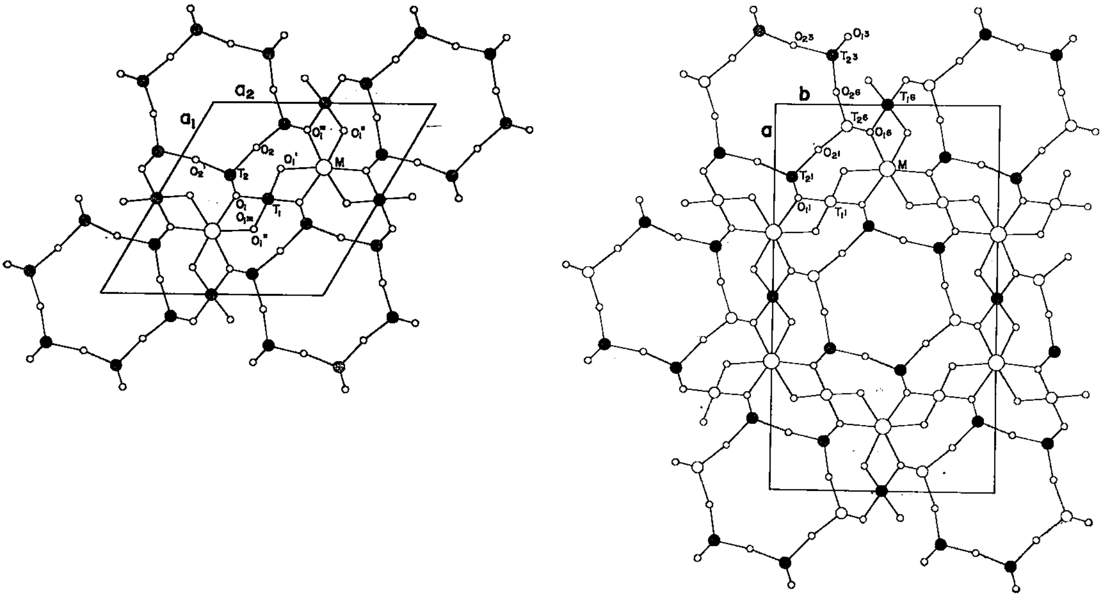

FIG. 1. The structure of indialite (left, c-axis projection) compared with an idealized drawing of the structure of low cordierite (right, c-axis projection). In the indialite structure, T_1 sites are Si-rich and T_2 are Al-rich. In the low cordierite structure, full T_1, T_2 circles are Si and open T_1, T_2 circles are Al.

Si$_5$O$_{18}$ (Fig. 1; Miyashiro & Iiyama 1954; Miyashiro *et al.* 1955). Miyashiro (1957) has since suggested an Al,Si order-disorder transformation between cordierite and indialite, analogous to that exhibited between the alkali feldspars low microcline and high sanidine. The transformation in cordierite was assumed to involve the distribution of 1 Al and 5 Si atoms in a six-membered ring; the remaining 3 Al atoms were assumed to occupy the tetrahedra outside the ring and not to be involved in the order-disorder change. Miyashiro reasoned that a disordered configuration of Al and Si in the ring would result in a hexagonal symmetry for indialite. On the other hand, a partly or completely ordered configuration would produce the symmetry of orthorhombic cordierite. However, a refinement of the structure of an orthorhombic cordierite from Guilford, Conn. (Gibbs 1966) suggests that the order-disorder transformation involves the distribution of all of the tetrahedral atoms in a continuous Al$_4$Si$_5$O$_{18}$ tetrahedral framework instead of just the arrangement of 1 Al and 5 Si atoms in a six-membered ring. Moreover, the tetrahedral atoms were found to be completely ordered within the limits of error with Al- and Si-containing tetrahedra in perfect alternation in all directions except for two Si-containing tetrahedra that share a common oxygen in the six-membered ring (Gibbs 1966; Cohen *et al.* 1977).

The present study of indialite was undertaken to clarify the polymorphism of cordierite by determining whether indialite is truly hexagonal, or dimensionally hexagonal but with a lower symmetry, and whether the order-disorder changes involve 1 Al and 5 Si atoms in a six-membered ring or 4 Al and 5 Si atoms in the Al$_4$Si$_5$O$_{18}$ tetrahedral framework (Meagher & Gibbs 1965; Meagher 1967).

EXPERIMENTAL

The specimen of indialite used in this investigation was collected from paralava formed during fusion of shale and sandstone by a burning coal seam in the Bokaro coalfield, India (specimen #23/951 of Fermor's collection–Fermor (1918). Its chemical composition was determined with an ARL-SEMQ automated electron microprobe utilizing the data correction routine of Bence & Albee (1968). The results of the analysis are listed in Table 1. A measurement of the specific gravity of indialite was not attempted because it contains minute unidentified inclusions; however, its density calculated from the unit-cell contents and cell edges is 2.59 g/cc.

Single-crystal Weissenberg and precession photographs revealed systematic absences of the type: hhl, $l=2n+1$ and $h\bar{h}l$, $l=2n+1$, consistent with space groups $P6cc$ and $P6/mcc$. The unit-cell parameters determined by least-squares

TABLE 1. CHEMICAL AND PHYSICAL DATA FOR INDIALITE

Oxide:	MgO	FeO*	Al_2O_3	SiO_2	
Weight %:	9.3	7.8	34.7	48.6	Total: 100.4%

Unit cell content
normalized to 18 oxygens: $2[Mg_{1.40}Fe_{0.66}Al_{4.11}Si_{4.89}O_{18}]$

Space group: P6/mcc Calculated density: 2.59 g/cc

Cell dimensions: $\underline{a}$ = 9.800(3)Å $\underline{c}$ = 9.345(3)Å

μ: 14.9 cm^{-1} Radiation/filter: Mo/Zr

Crystal size: 0.14 x 0.10 x 0.06 mm

Number observed reflections: 168

$R = \Sigma(|F_o| - |F_c|)/\Sigma|F_o| = 0.047$

$Rw = (\Sigma\, w(|F_o| - |F_c|)^2/\Sigma wF_o{}^2)^{1/2} = 0.061$

* Total Fe assumed ferrous

refinements of precision back-reflection Weissenberg data (*a* 9.8000(3), *c* 9.345(3)Å) compare well with those determined by Miyashiro *et al.* (1955) (*a* 9.812, *c* 9.351Å).

In order to critically analyze the diffraction symmetry of indialite, intensity data were collected on the basis of an orthorhombic cell (*Cccm*) with $a = \sqrt{3}b$. The crystal was mounted about the *c* axis and three-dimensional intensity data were collected utilizing equi-inclination geometry on a manual Weissenberg scintillation-counter diffractometer. Zr-filtered Mo radiation was used along with a pulse-height discriminator. Non-equivalent reflections for the orthorhombic cell were scanned within $\sin\theta \leq 0.43$ and traced on a strip-chart recorder. Relative intensities were then determined with an integrating planimeter and corrected for Lorentz and polarization factors. An absorption correction was not applied.

In addition to the symmetrically non-equivalent intensities collected for the orthorhombic cell, approximately thirty *hk*0 and *hkl* reflections were recorded to test their equality across all {110} and {310} planes. Hexagonal equivalent intensities measured on opposite sides of these planes were found to be statistically equal, indicating hexagonal diffraction symmetry for indialite.

The indices of the planes in the orthorhombic cell were transformed to those defined in terms of the hexagonal cell and the symmetrically equivalent intensities were averaged to give 168 non-equivalent hexagonal intensity data.

REFINEMENT

The refinement of indialite was carried out in space group *P6/mcc* because of the apparent structural relationship between indialite, beryl and low cordierite. Starting atomic positional parameters for indialite were obtained by transformation of the refined positional parameters for Haddam cordierite (Meagher 1967) to a hexagonal basis.

The structure refinement was calculated with an IBM 360/67 using a modified version of the full-matrix least-squares program ORFLS (Busing *et al.* 1964). Scattering curves from the International Tables for X-ray Crystallography (1962) were modified for half ionization and corrected for the real part of the anomalous dispersion. Unobserved reflections were not included in the refinement and the weighting scheme of Hanson (1965) was applied. Because the Al,Si distribution in the framework was initially unknown, the atoms at the T_1 and T_2 tetrahedral sites were assigned a random distribution scattering curve of 4/9 Al + 5/9 Si in the first few cycles of the refinement of a model based on isotropic temperature factor coefficients.

Upon convergence of the refinement (weighted *R*-index = 0.085) tetrahedral Al,Si contents were estimated from the $<T\text{-}O>$ bond lengths (details of this procedure are given in Discussion Section). Following an adjustment of the scattering curves for the T_1 and T_2 atoms to match that estimated from $<T\text{-}O>$, and an extension of the model to include anisotropic temperature factor coefficients, a final least-squares refinement converged to a weighted *R*-index of 0.061*. Refined positional and thermal para-

TABLE 2. POSITIONAL PARAMETERS AND EQUIVALENT ISOTROPIC TEMPERATURE FACTORS FOR INDIALITE

Atom	x	y	z	B (Å^2)*
T_1	1/2	1/2	1/4	0.9(1)
T_2	0.3727(4)	0.2668(4)	0	1.16(9)
M	1/3	2/3	1/4	1.8(2)
O_1	0.4851(6)	0.3494(5)	0.1445(6)	1.4(1)
O_2	0.2305(11)	0.3093(11)	0	2.2(3)

*Isotropic equivalent of anisotropic temperature factor (Hamilton 1959).

---

*Tables of structure factors are available, at a nominal charge, from the Depository of Unpublished Data, CISTI, National Research Council of Canada, Ottawa K1A 0S2.

TABLE 3. ANISOTROPIC TEMPERATURE FACTOR COEFFICIENTS*

Atom	β_{11}	β_{22}	β_{33}	β_{12}	β_{13}	β_{23}
T_1	212(57)	213(47)	426(44)	83	0	0
T_2	346(39)	363(40)	440(36)	204(36)	0	0
M	478(68)	478	771(59)	239	0	0
O_1	298(62)	286(69)	632(71)	99(56)	-79(58)	5(55)
O_2	793(134)	821(103)	814(112)	649(94)	0	0

* Values are reported x 10^5. The coefficients are of the form ($\beta_{11}h^2$ + ... $2\beta_{12}$ hk + ...).

TABLE 4. INTERATOMIC DISTANCES AND BOND ANGLES OF INDIALITE

T,M-O Distances (Å)		O-O Distances (Å)		Enclosed O-T-O Bond Angles (°)
T_1-O_1 [4]*	1.719(5)	O_1-O_1" [2]	2.553(10)	95.9(3)
		O_1'-O_1" [2]	2.816(9)	110.0(3)
		O_1-O_1' [2]	3.032(10)	123.7(3)
		<O-O>	2.800	
T_2-O_1 [2]	1.674(6)	O_1-O_2' [2]	2.693(9)	109.6(3)
-O_2'[1]	1.623(8)	O_1-O_2 [2]	2.688(9)	108.3(2)
-O_2 [1]	1.642(9)	O_2-O_2' [1]	2.728(10)	113.3(7)
<T_2-O>	1.653	O_1-O_1m [1]	2.701(11)	107.6(3)
		<O-O>	2.699	
M-O_1 [6]	2.108(5)	O_1-O_1" [3]	2.553(10)	
		O_1'-O_1"'[3]	2.877(10)	
		O_1'-O_1" [6]	3.227(8)	
		<O-O>	2.971	
T_1-O_1-T_1	129.1(3)	T_2-O_2-T_2	173.3(7)	
		T_2-O_1-T_1	129.1(3)	

* Number in square brackets refers to multiplicity of the bond length.

meters are given in Tables 2 and 3, respectively, and bond length and angle data are given in Table 4. Calculation of a three-dimensional difference Fourier synthesis (F_o-F_c) revealed no significant anomalies, indicating an absence of alkali atoms and H_2O in the structural channels of the indialite.

DISCUSSION

Nomenclature

Indialite (Fig. 1) is isostructural with beryl and, like low cordierite, it is a framework silicate. To facilitate discussion and comparison of the structures of the hexagonal and orthorhombic polymorphs of $Mg_2Al_4Si_5O_{18}$, we propose the nomenclature outlined in Table 5. The indialite framework consists of T_1 tetrahedra linked both laterally and vertically to six-membered rings of T_2 tetrahedra. The T_1 tetrahedron shares two opposite edges with *M* octahedra whereas the T_2 tetrahedron lacks shared edges. Thus, each O_2 atom is bonded only to two T_2 atoms and each O_1 is bonded to one T_1, one T_2 and an *M* atom. Transformation of indialite to orthorhombic cordierite results in O_1 and O_2 splitting into two sets, each consisting of three non-equivalent oxygen atoms labelled O_11, O_16 and O_13, and O_21, O_26 and O_23, respectively. In addition, the T_1 atoms split into a set of two non-equivalent tetrahedral atoms labelled T_11, T_16 and the T_2 atoms split into a set of three non-equivalent tetrahedral atoms labelled T_21, T_26 and T_23. The atoms O_1, O_2, T_1 and T_2 in indialite correspond to atoms labelled O_11, O_21, T_11 and T_21 in orthorhombic cordierite, the 1 denoting the fact that they are related by the identity map in the hexagonal polymorph. On the other hand, the atoms labelled O_16, O_26, T_16 and T_26 in cordierite are related by a sixth turn, 6, to O_1, O_2, T_1 and T_2 in indialite whereas the atoms labelled O_13, O_23 and T_23 are related by a third turn, 3, to O_1, O_2 and T_2 in the hexagonal form. Following the scheme

TABLE 5. NOMENCLATURE PROPOSED FOR ATOMS IN INDIALITE AND ORTHORHOMBIC CORDIERITE

	Indialite				Orthorhombic cordierite		
	x_h	y_h	z_h		x_o	y_o	z_o
T_1	1/2	1/2	1/4	T_11	1/4	1/4	1/4
				T_16	0	1/2	1/4
T_2	0.3727	0.2668	0	T_21	0.1864	0.0805	0
				T_26	.0530	.3200	0
				T_23	- .1334	.2393	0
O_1	0.4851	0.3494	0.1445	O_11	0.2426	0.1069	0.1445
				O_16	.0679	.4172	.1445
				O_13	- .1747	.3104	.1445
O_2	0.2305	0.3093	0	O_21	0.1153	0.1941	0
				O_26	- .0394	.2699	0
				O_23	- .1547	.0759	0
M	1/3	2/3	1/4	M	0.1667	1/2	1/4

Coordinate transformations from hexagonal to C-centered orthorhombic basis for the atoms in indialite related by indentity, 1, sixth turn, 6, and third turn, 3.

1: $x_o = x_h/2$, $y_o = y_h - x_h/2$, $z_o = z_h$

6: $x_o = x_h/2 - y_h/2$, $y_o = x_h2 + y_h/2$, $z_o = z_h$

3: $x_o = -y_h/2$, $y_o = x_h - y_h/2$, $z_o = z_h$

used for the feldspars (Kroll 1973), the Al contents of the T_1 and T_2 tetrahedra in indialite are denoted as t_1 and t_2, respectively, whereas those of the T_11, T_16, T_21, T_26 and T_23 tetrahedra in orthorhombic cordierite are denoted as t_11, t_16, t_21, t_26 and t_23, respectively.

Al, Si contents of tetrahedra

A tetrahedral site refinement using neutron diffraction data recorded for the White Well cordierite (Cohen *et al.* 1977) indicates it to be completely ordered ($t_11 = t_26 = 1.0$; $t_16 = t_21 = t_23 = 0.0$) within experimental error despite that $\langle T_1\text{–O}\rangle$ for Si- and Al-containing tetrahedra are ~0.01Å longer than those recorded for corresponding T_2 tetrahedra (Table 5). Cohen *et al.* (1977) have also asserted that the Guilford cordierite is completely ordered within experimental error because its $\langle T\text{-O}\rangle$ values are statistically identical with those of the White Well cordierite. Following analogous studies on feldspars, Al contents of the T_1 and T_2 tetrahedra in indialite were estimated assuming a linear relation betweeen $\langle T\text{-O}\rangle$ and Al content (Smith 1953; Smith & Bailey 1963). Because $\langle T_1\text{-O}\rangle$ is expected to be longer than $\langle T_2\text{-O}\rangle$ for tetrahedra with identical Al,Si contents, two separate equations were used to estimate the contents of these tetrahedra. These equations were derived utilizing the grand mean T-O bond lengths, $\langle\langle T_1\text{-O}\rangle\rangle$ and $\langle\langle T_2\text{-O}\rangle\rangle$, for the Al- and Si-containing tetrahedra of the White Well and Guilford cordierite, and show that the T_1 tetrahedra in indialite are Al-rich, $t_1 = 0.72$, and that the T_2 tetrahedra are Si rich, $t_2 = 0.30$ (Table 6). Because the Al,Si contents (1.8 Al, 4.2 Si) of the six T_2 tetrahedra in the ring differ significantly from 1 Al and 5 Si and because the contents (2.16 Al, 0.84 Si) of the 3 T_1 tetrahedra outside the ring differ significantly from 3 Al, it is apparent that the order-disorder changes in cordierite involve all the tetrahedra in the structure instead of just the ones in the ring. Hence, despite recent arguments by Strunz *et al.* (1971) that cordierite should be classified as a ring silicate, we assert that cordierite should be classified as a framework silicate because the Al,Si changes involve all nine tetrahedra in the framework.

TABLE 6. ESTIMATION OF T_1, T_2 SITE OCCUPANCIES IN INDIALITE

	Content	White Well Cordierite*	Guilford Cordierite	
		$\langle T_1O\rangle$		$\langle\langle T_1\text{-O}\rangle\rangle$
T_11	Al	1.758	1.753	1.756
T_16	Si	1.626	1.621	1.624
		$\langle T_2O\rangle$		$\langle\langle T_2\text{-O}\rangle\rangle$
T_26	Al	1.742	1.744	1.743
T_21	Si	1.614	1.611	1.614
T_23	Si	1.617	1.612	

Equations used to estimate the Al,Si contents of T_1 and T_2 tetrahedra from $\langle T_1\text{-O}\rangle$ and $\langle T_2\text{-O}\rangle$ of indialite:

$$t_1 = \frac{\langle T_1\text{-O}\rangle - 1.624}{0.132} = \frac{1.719 - 1.624}{0.132} = 0.72(4)$$

$$t_2 = \frac{\langle T_2\text{-O}\rangle - 1.614}{0.129} = \frac{1.653 - 1.614}{0.129} = 0.30(3)$$

Estimated Al content = 3(0.72) + 6(0.30) = 4.0**

Al content estimated in probe analysis = 4.1***

* data from Cohen et al. (1977)

** normalized on the basis of 9 tetrahedral atoms

*** normalized on the basis of 18 oxygen atoms .

TABLE 7. HYPOTHETICAL T_1, T_2 SITE OCCUPANCIES AND CORRESPONDING BRAGG-WILLIAMS LONG RANGE ORDER PARAMETERS FOR INDIALITE

Al,Si occupancy	Bragg-Williams parameter
t_1 = 0.44	0.00
t_2 = 0.44	
t_1 = 0.67	0.30
t_2 = 0.33	
t_1 = 0.0	0.60
t_2 = 0.67	
t_1 = 1.0	0.75
t_2 = 0.17	

Long- and short-range order

Using the estimated Al,Si contents of the tetrahedra, a Bragg-Williams (1934) long-range order parameter of 0.38 is calculated and it is now apparent that a range of intermediate Al,Si order is possible within the constraints of $P6/mcc$ symmetry just as there is a range of intermediate order within the constraints of $C2/m$ for the alkali feldspars. Ideally, one can conceive of Al, Si distributions in indialite which result in Bragg-Williams parameters ranging from 0.0 to 0.75 as illustrated in Table 7. One may consider the Al, Si distribution for the 0.60 and 0.75 degree of order to be unlikely for crystal-chemical reasons; however, the extent to which long-range order may develop in indialite has yet to be determined.

The discussion to now has dealt exclusively with long-range order; nevertheless one cannot preclude the possibility that short-range ordered domains exist in indialite. Although no conclusive evidence for their existence in indialite has been found, Langer & Schreyer (1969), in a study of synthetic cordierite, observed a discrepancy between the infrared spectra and powder diffraction data which they believe could be a result of the incipient development of short-range ordered domains followed by the development of long-range ordering with further annealing.

A simple model for the development and orientation of short-range ordered domains in indialite can be constructed based upon the preferential selection of tetrahedra, within the tetrahedral framework, by Si and Al. Ideally, in totally disordered indialite, there is a 5/9 probability of finding Si in either the T_1 or T_2 tetrahedra. In partly ordered indialite the T_1 and T_2 tetrahedra may differ in Al,Si content but there is an equal probability of finding Si in each of the three equivalent T_1 tetrahedra outside the six-membered ring (Fig. 2a). Likewise, there is an equal probability of finding Si in each of the six equivalent T_2 tetrahedra within the six-membered ring. If, however, in some localized volume, Si should preferentially occupy one of the three T_1 tetrahedra outside of the ring, and assuming the ordered distribution within the ring would follow, the orientation of the ordered unit cell within the hexagonal indialite host would be as shown in Figure 2b which coincides with the orientation of the ordered cordierite cell depicted in Figure 1b. Likewise, a preference by Si for each of the other two T_1 tetrahedra would lead to local domains with orientations as shown in Figures 2c and 2d. Based upon this model, there are three possible orientations within the hexagonal host which the short-range ordered domains may assume. Continued growth and enlargement of these ordered domains may ultimately lead to the formation of the familiar polysynthetic or trilling twin in cordierite (Meagher & Gibbs 1965; Meagher 1967) which will be discussed in terms of a possible two-step order-disorder mechanism in a forthcoming paper.

It is of interest to calculate the long-range average Al,Si contents of the T_1 and T_2 tetrahedra in a stoichiometric indialite which is composed of equal amounts of these short-range ordered domains. Such a calculation yields an occupancy of $t_1 = 0.67$ and $t_2 = 0.33$ with a long-range order parameter of 0.30 (Table 7). This agrees within experimental error with the Al, Si content determined in the refined indialite struc-

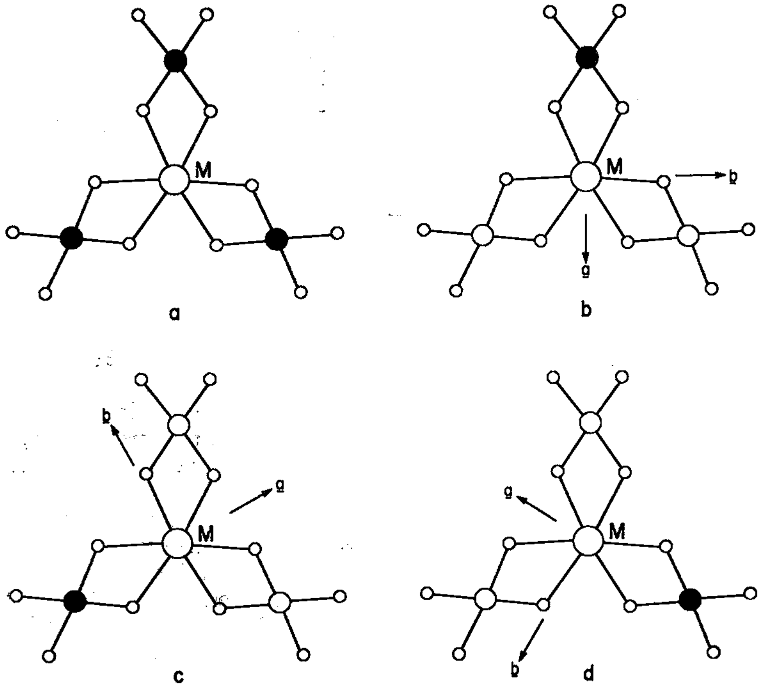

FIG. 2. C-axis projection of the arrangement of T_1 tetrahedra about the M octahedron in (a) indialite and (b), (c) and (d) three possible ordered arrangements of 1 Si (full circle) and 2 Al (open circles).

ture for which $t_1 = 0.72$, $t_2 = 0.30$ and the long-range order parameter is 0.38. This agreement may be fortuitous, however, since equal amounts of the three domains need not occur and, at the present time, we can only acknowledge the possible existence of short-range ordered domains in this indialite.

ACKNOWLEDGEMENTS

The National Research Council of Canada and the National Science Foundation (Grant DES71-00486-A03) are thanked for their generous financial support of this study. We also thank Professor A. Miyashiro of Tokyo University for donating the specimen used in our study.

REFERENCES

Bence, A. E. & Albee, A. L. (1968): Empirical correction factors for the electron microanalysis of silicates and oxides. *J. Geol.* **76**, 382-403.

Bragg, W. L. & Williams, E. J. (1934): The effect of thermal agitation on atomic arrangements in alloys. *Proc. Roy. Soc.* **A145**, 699-730.

Busing, W. R., Martin, K. O. & Levy, H. A. (1964): ORFFE, a fortran crystallographic function and error program. *Oak Ridge Nat. Lab. Rep.* ORNL-TM-306.

Byström, A. (1942): The crystal structure of cordierite. *Ark. Kemi. Min. Geol.* **15B**, 1-5.

Cohen, J. P., Ross, F. K. & Gibbs, G. V. (1977): An X-ray and neutron diffraction study of hydous low cordierite. *Amer. Mineral.* (in press).

Fermor, L. L. (1918): Preliminary note on the burning of coal seams at the outcrop. *Trans. M.G.I. Ind.* **12**, 50-63.

Folinsbee, R. E. (1941): Optical properties of cordierite in relation to alkalies in the cordierite-beryl structure. *Amer. Mineral.* **26**, 485-500.

Gibbs, G. V. (1966): The polymorphism of cordierite: I. The crystal structure of low cordierite. *Amer. Mineral.* **51**, 1068-1087.

Gossner, B. & Mussgnug, F. (1928): Vergleicherde röntgenographische untersuchung von magnesiumsilikaten. *Neues Jahrb. Mineral. Abt.* **58**, 216-227.

Hanson, A. W. (1965): The crystal structure of the agulene, S-trinitrobenzene complex. *Acta Cryst.* **19**, 19-26.

Kroll, H. (1973): Estimation of the Al,Si distribution of feldspars from the lattice translations Tr[110] and Tr[110]. *Contr. Mineral. Petrology* **39**, 141-156.

Langer, K. & Schreyer, W. (1969): Infrared and powder X-ray diffraction studies on the polymorphism of cordierite, $Mg_2(Al_4Si_5O_{18})$. *Amer. Mineral.* **54**, 1442-1459.

Meagher, E. P. (1967): *The Crystal Structure and Polymorphism of Cordierite.* Ph.D. Thesis, The Pennsylvania State University, University Park, Pennsylvania.

——— & Gibbs, G. V. (1965): Crystal structure and polymorphism of cordierite. *Program Abstr. Geol. Soc. Amer. Ann. Meet.* Kansas City, 105-106.

Miyashiro, A. (1957): Cordierite-indialite relations. *Amer. J. Sci.* **255**, 43-62.

——— & Iiyama, T. (1954): A preliminary note on a new mineral, indialite, polymorphic with cordierite. *Proc. Imp. Acad. Japan* **30**, 746-751.

———, ———, Yamasaki, M. & Miyashiro, T. (1955): The polymorphism of cordierite and indialite. *Amer. J. Sci.* **253**, 185-208.

Rankin, G. A. & Merwin, H. E. (1918): The ternary system MgO-Al_2O_3-SiO_2. *Amer. J. Sci.* **45**, 301-325.

Smith, J. V. (1954): A review of the Al-O and Si-O distances. *Acta Cryst.* **7**, 479-481.

——— & Bailey, S. W. (1963): Second review of Al-O and Si-O tetrahedral distances. *Acta Cryst.* **16**, 801-811.

Strunz, H., Tennyson, C. & Uebel, P.-J. (1971): Cordierite morphology, physical properties, structure, inclusions and oriented intergrowth. *Minerals Sci. Eng.* **3**, 3-18.

Takane, K. & Takeuchi, T. (1936): The crystal structure of cordierite. *Jap. Assoc. Mineral. Petrology Econ. Geol.* **16**, 101-127 (in Japanese).

Manuscript received September 1976, emended November 1976.