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## THE POLYMORPHISM OF CORDIERITE AND INDIALITE

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**ABSTRACT.** Three compounds of magnesium cordierite composition ( $Mg_2Al_4Si_5O_{18}$ ) have been synthesized. They are called  $\alpha$ -,  $\beta$ - and  $\mu$ -forms. The  $\alpha$ -form, and possibly the  $\beta$ -form also, have been considered to be identical with natural cordierite, but our study has shown that it is necessary to make drastic modifications of existing conceptions in this regard.

We found that the  $\alpha$ -form, and probably the  $\beta$ -form also, are truly hexagonal in symmetry, and then are different from cordierite which is pseudohexagonal. The  $\alpha$ - and  $\beta$ -forms are iso-structural with beryl. The crystal structure of natural cordierite is derived by slight deformations from those of the  $\alpha$ - and  $\beta$ -forms.

We found crystals of the  $\alpha$ -form in fused sediments from Bokaro coalfield, India, and gave it the mineralogical name of "indialite." When necessary, the  $\alpha$ - and  $\beta$ -forms should be called "high and low indialites" respectively, as there is high-low inversion relation between them.

In natural cordierite, there exist probably two polymorphic forms, here called "high and low cordierites." Low cordierite is transformed to high cordierite on heating at high temperatures. High cordierite occurs in volcanic rocks, while low cordierite occurs in other kinds of rocks, especially in metamorphic rocks.

At the end is discussed the petrological relation between cordierite and osumilite, the latter of which is a mineral discovered as a by-product of the present study. The results of our study are summarized in table 15.

### INTRODUCTION

Towards the end of the 19th century, synthetic experiments on cordierite were carried out by C. Doelter and E. Hussack, by L. Bourgeois, and by J. Morozewicz (Doelter, 1917, p. 626). Morozewicz (1899) especially made an important contribution, and called the crystals which he obtained "cordierite."

Later, G. A. Rankin and H. E. Merwin (1918) synthesized two compounds which probably have magnesium cordierite composition ( $Mg_2Al_4Si_5O_{18}$ ) in the course of their study of the ternary system  $MgO-Al_2O_3-SiO_2$ . They called the two compounds " $\alpha$ - and  $\mu$ -forms of ternary compound" or " $\alpha$ - and  $\mu$ -ternary compound," and considered that the  $\alpha$ -form is probably identical with the "cordierite" synthesized by Morozewicz and also with natural cordierite. Sometimes they called it simply "cordierite."

In the years 1929 to 1932, many investigators found that the  $\alpha$ -form has an unusually low thermal expansion coefficient and is useful in the ceramic industry.

H. S. Yoder, Jr. (1952) synthesized the  $\alpha$ -form by the hydrothermal method at temperatures above  $830^\circ C.$ , while he obtained similar crystals

having slightly higher refractive indices by the same method at temperatures below 830°C. He did not notice any significant difference in X-ray pattern between the two kinds, and considered that at least the latter is identical with cordierite.

TABLE I  
Optical Properties of Synthetic Forms of  $Mg_2Al_4Si_5O_{18}$

Authors		Rankin and Merwin (1918)	Yoder (1952)	Karkhanavala and Hummel (1953)
$\alpha$ -form	$\alpha$	1.524	1.521	1.524
	$\gamma$	1.528	1.526	1.526
	2V	0° (negative)	30° (negative)	
$\beta$ -form	$\alpha$		1.537	
	$\gamma$		1.541	
	2V		<45° (negative)	
$\mu$ -form	$\alpha$ }	1.535-1.560		1.546
	$\gamma$ }			

M. D. Karkhanavala and F. A. Hummel (1953) considered that only the  $\alpha$ -form is identical with natural cordierite. Asserting that the cordierite-like crystals synthesized below 830°C. by Yoder represent a new form of  $Mg_2Al_4Si_5O_{18}$ , they called it " $\beta$ -form." They said also that the  $\mu$ -form of Rankin and Merwin is iso-structural with  $\beta$ -spodumene, and so is decidedly different from natural cordierite.

Thus, it is generally believed that there exist three polymorphic modifications,  $\alpha$ ,  $\beta$  and  $\mu$ , of magnesium cordierite composition, and that the  $\alpha$ -form, and possibly the  $\beta$ -form also, are identical with natural cordierite. The characteristic optical properties are shown in table I.

Our attention was drawn, however, to the fact that the  $\alpha$ -form differs from natural cordierite in optical angle. According to the original description by Rankin and Merwin the former is uniaxial negative, whereas the latter is biaxial negative or positive, usually with a large optical angle. Doubting therefore the identity of the  $\alpha$ -form with natural cordierite, we examined them and came to the conclusion that the  $\alpha$ -form is truly hexagonal in symmetry. Thus the  $\alpha$ -form differs from natural cordierite which is pseudohexagonal (rhombic or possibly monoclinic).

Further, a sample of what appears to be the  $\beta$ -form was examined and found to be probably hexagonal. Hence the  $\beta$ -form also differs from natural cordierite. Thus any of the three synthetic forms of magnesium cordierite composition is not identical with natural cordierite.

Looking for the occurrence of the synthetic forms in nature, we found crystals of the  $\alpha$ -form in fused sediments from Bokaro coalfield, India. The  $\alpha$ -form is thus a mineral in the proper sense of the word. We proposed to call this new mineral "*indialite*" after India (Miyashiro and Iiyama, 1954).

We found in addition that there exist probably two polymorphic modifications in natural cordierite. They have distinctive optical properties and show characteristic modes of occurrence. Probably one or the other of the two modifications is stable at high or low temperatures respectively, while the  $\alpha$ -,  $\beta$ - and  $\mu$ -forms are perhaps metastable.

Thus we have come to know that there exist probably five polymorphs of cordierite composition. It is an important problem to avoid confusion and decrease a burden on memory by a clear nomenclature. In the original nomenclature of Rankin and Merwin, the names of " $\alpha$ - and  $\mu$ -ternary compound" were used as chemical names only to distinguish the two synthetic compounds. Some later investigators, however, called them " $\alpha$ - and  $\mu$ -cordierites" respectively. Others called the  $\alpha$ - or  $\beta$ -form simply "cordierite." Now, the  $\alpha$ -,  $\beta$ - and  $\mu$ -forms have been proved to differ from cordierite, and, as discussed below, probably they are not in high-low inversion relation to cordierite. Therefore it is undesirable to call them cordierite any longer.

We propose to use the prefixes,  $\alpha$ ,  $\beta$  and  $\mu$ , only as those for chemical names such as " $\alpha$ -,  $\beta$ - and  $\mu$ -forms of  $\text{Mg}_2\text{Al}_4\text{Si}_5\text{O}_{18}$ " or " $\alpha$ -,  $\beta$ - and  $\mu$ - $\text{Mg}_2\text{Al}_4\text{Si}_5\text{O}_{18}$ ," in harmony with the original usage by Rankin and Merwin. It is reasonable to restrict the use of the mineralogical name "*cordierite*" to the two pseudo-hexagonal forms represented by natural cordierite. When necessary, they should be distinguished from each other as "*high and low cordierites*" respectively, as they are connected by high-low inversion relation. We propose to use the mineralogical name "*indialite*" for the two hexagonal forms ( $\alpha$ - and  $\beta$ -forms.). When necessary, the  $\alpha$ -form should be called "*high indialite*," and the  $\beta$ -form "*low indialite*," as they are connected by high-low inversion relation. (The results of the present study with this nomenclature are synoptically shown in table 15.)

It is to be noted that the distinction between cordierite and indialite has been firmly established by optical as well as X-ray method, whereas those between high and low cordierites and between high and low indialites are based only on the optical data, and are not considered to have been established so firmly yet. We ourselves do not have any evidence to prove or disprove the existence of distinction between high and low indialites ( $\alpha$ - and  $\beta$ -forms).

This study was carried out in the Geological Institute, Tokyo University, intermittently from 1951 to 1954. In the winter of 1952-1953, A. Miyashiro, then temporarily at Harvard University, found "osumilite" as a by-product of the present study. Osumilite is related in crystal structure to, and has similar optical and chemical properties with, cordierite. The results are published in separate papers (Miyashiro, 1953, 1955). Some petrological considerations on the relation between osumilite and cordierite will be given in this paper.

ON  $\alpha$ - $\text{Mg}_2\text{Al}_4\text{Si}_5\text{O}_{18}$

*Hexagonal symmetry of  $\alpha$ - $\text{Mg}_2\text{Al}_4\text{Si}_5\text{O}_{18}$ .*—The  $\alpha$ -form of  $\text{Mg}_2\text{Al}_4\text{Si}_5\text{O}_{18}$  was synthesized by devitrifying glass of that composition at temperatures around 1200°C. The X-ray diffraction data obtained by means of the Philips Geiger counter X-ray spectrometer are shown in figure 1 and table 2, the latter of which shows that it can be perfectly indexed on the basis of a hexagonal unit cell with dimensions:  $a = 9.782 \text{ \AA}$  and  $c = 9.365 \text{ \AA}$ .

Rankin and Merwin (1918) described crystals of the  $\alpha$ -form as "six-sided prisms with basal termination and negative elongation." "The prism angle, within the limits of error of measurement, was 120°, and basal sections gave a nearly or quite uniaxial negative interference figure." These properties are in harmony with hexagonal symmetry.

X-ray diffraction curves for natural cordierites from Asama volcano, Japan, and from Laramie Range, Wyoming, are shown in figure 1 for comparison. The curves for the  $\alpha$ -form and natural cordierites show general similarity to one another. Every peak in the curve for the former corresponds to a peak or a group of a few closely spaced peaks in the curves for the latter. The length of the edge  $a$  of the  $\alpha$ -form is nearly equal to that of the edge  $b$  of natural cordierites (table 15). Natural cordierite is pseudo-hexagonal, so that the edge  $a$  is about  $\sqrt{3}$  times as long as the edge  $b$ . Thus it is probable that the  $\alpha$ -form and natural cordierite are very similar in crystal structure.

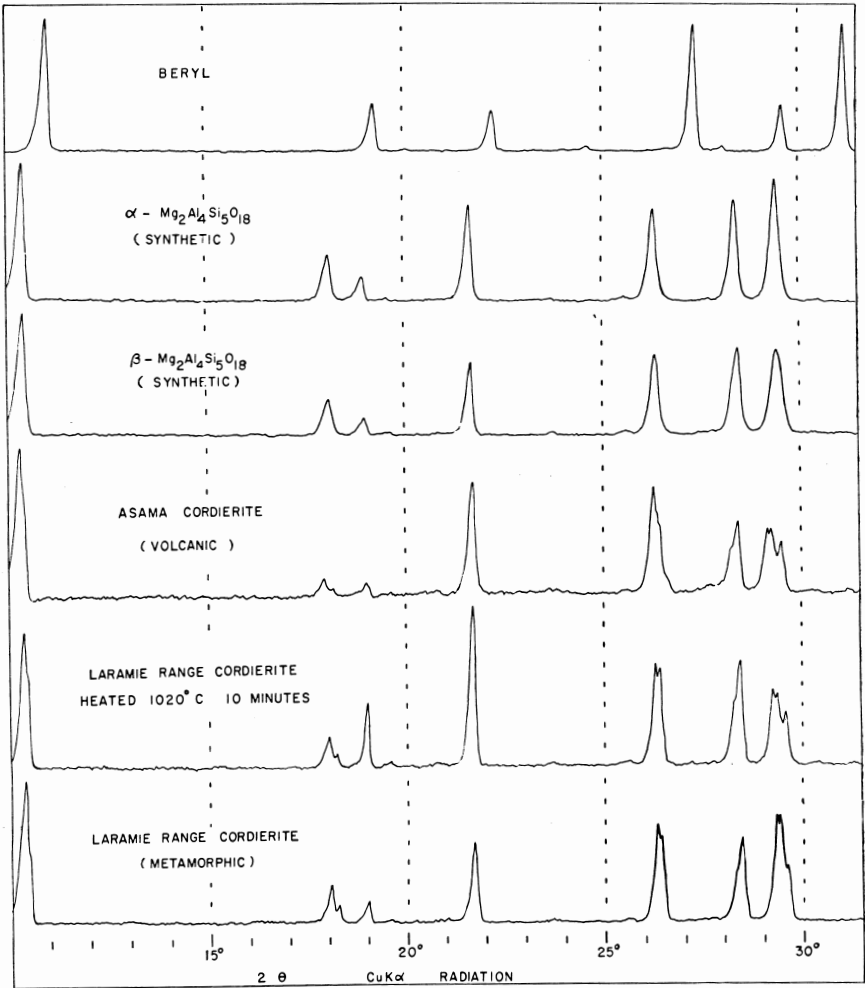


Fig. 1. X-ray spectrograms of beryl,  $\alpha$ - and  $\beta$ -Mg<sub>2</sub>Al<sub>4</sub>Si<sub>5</sub>O<sub>18</sub> (high and low indialites respectively), and cordierites.

TABLE 2  
 Powder Diffraction Data,  $\alpha$ -form of  $Mg_2Al_4Si_5O_{18}$ 

Indices $N, l$	Intensity	$2\theta^\circ$ obs	$2\theta^\circ$ calc	
1,0	100	10.42	10.44	} CuK $\alpha$
3,0	30	18.13	18.14	
0,2	15	18.96	18.95	
1,2	65	21.69	21.68	
3,2	60	26.34	26.33	
4,2	65	28.41	28.39	
7,1	75	29.44	29.45	
7,2	25	33.87	33.88	
12,0	4	36.73	36.75	
9,2	3	37.17	37.15	
0,4	10	38.47	38.46	} CuK $\alpha_1$
13,1	3	39.52	39.54	
7,3	3	40.34	40.32	
12,2	4	41.61	41.62	
13,2	8	43.02	43.03	
19,0	1	46.6	46.64	
13,3	8	48.43	48.45	
21,0	4	49.25	49.25	
21,1	4	50.30	50.28	
9,4	6	50.60	50.60	
12,4	18	54.19	54.19	

Note:  $N = h^2 + k^2 + hk$ . Filtered Cu radiation is used. The reading is taken at the mid-point of the peak width at approximately two-thirds of the peak height. ( $a = 9.782 \text{ \AA}$ ,  $c = 9.365 \text{ \AA}$ .) In tables 2, 3, 4, 5, and 12, the intensity is assumed to be proportional to the peak height.

The pattern in the range of  $2\theta$  between  $29^\circ$  and  $30^\circ$  for CuK $\alpha$  radiation is the most distinctive. There is only one peak in this range in the  $\alpha$ -form, while there are at least three peaks, (511), (421), and (131), which are closely spaced but clearly distinguishable from one another, in this range in natural cordierites.<sup>1</sup>

Supposing that natural cordierite be slightly deformed to become truly hexagonal, the three sets of indices, (511), (421), and (131), becoming orthohexagonal ones, are transformed to Bravais-Miller indices, (21 $\bar{3}$ 1), (12 $\bar{3}$ 1), and ( $\bar{1}$ 3 $\bar{2}$ 1) respectively. These planes in the hexagonal lattice have the same interplanar spacing. Thus the reflections from them appear at the same position in the powder pattern, forming a single peak. This is the reason why the three peaks in natural cordierite correspond to a single peak in the  $\alpha$ -form, whose structure is probably derived by slight deformations from that of natural cordierite.

<sup>1</sup> The Laramie Range cordierite, shown in figure 1, is very poor in iron (FeO=1.71%). Prof. H. Shibata of Tokyo University of Education kindly provided us with a specimen of an unusually iron-rich cordierite ( $\alpha=1.558$ ,  $\beta=1.570$ ,  $\gamma=1.576$ ,  $d=2.782$ ) from a protolithionite-topaz-bearing metasomatic vein in granite at Yagenyama, Japan (Shibata, 1939). The X-ray diffractogram of this mineral also shows at least three peaks in the range of  $2\theta$  between  $29^\circ$  and  $30^\circ$ .

Figure 1 shows also a diffraction pattern for a beryl from Royalstown, Massachusetts. The curves for the  $\alpha$ -form and the beryl are very similar, though the corresponding peaks are shifted systematically because of the smaller size of the unit cell of beryl. All the observed reflections of the  $\alpha$ -form are compatible with the space group of  $C6/mcc$  ( $D_{6h}^2$ ), to which beryl also belongs. Thus the  $\alpha$ -form is probably iso-structural with beryl, which is composed of hexagonal rings (Bragg and West, 1926).

In beryl the hexagonal ring is composed of six Si-O tetrahedra, while in natural cordierite the pseudohexagonal ring is composed of five Si-O tetrahedra and one Al-O tetrahedron on the average. The distribution of Si and Al atoms in the latter ring need not be random, so far as the ring does not have hexagonal symmetry. On the other hand, the true hexagonal symmetry of the  $\alpha$ -form requires random arrangement of Si and Al atoms in its ring.

The chemical composition of the  $\alpha$ -form is not fixed. The Mg atoms can be replaced by  $Fe^{+2}$  partly or wholly, as will be shown later. We also have evidence showing that AlAl atoms can be replaced by SiMg atoms to some extent, as will be shown in a later paper.

*Iron-analogue of  $\alpha$ -Mg<sub>2</sub>Al<sub>4</sub>Si<sub>5</sub>O<sub>18</sub>.*—Drs. J. F. Schairer and H. S. Yoder, Jr. of the Geophysical Laboratory, Carnegie Institution of Washington, kindly provided us with an X-ray diffraction diagram for “iron-cordierite” ( $Fe_2Al_4Si_5O_{18}$ ), synthesized by Schairer and Yagi (1952) in the course of study of the ternary system  $FeO-Al_2O_3-SiO_2$ . The curve is very similar to that for the  $\alpha$ -form of  $Mg_2Al_4Si_5O_{18}$ . There is only one peak in the range of  $2\theta$  between  $29^\circ$  and  $30^\circ$ . As shown in table 3, it also can be indexed on the basis of the hexagonal unit cell with dimensions:  $a = 9.860 \text{ \AA}$  and  $c = 9.285 \text{ \AA}$ . Thus probably the “iron-cordierite” is not true cordierite, but the iron-analogue of  $\alpha$ -Mg<sub>2</sub>Al<sub>4</sub>Si<sub>5</sub>O<sub>18</sub>—so to speak,  $\alpha$ -Fe<sub>2</sub>Al<sub>4</sub>Si<sub>5</sub>O<sub>18</sub>.

Thus the hexagonal symmetry of  $\alpha$ -Mg<sub>2</sub>Al<sub>4</sub>Si<sub>5</sub>O<sub>18</sub> is confirmed by the existence of its iron-analogue, whose unit cell also has a hexagonal shape. If the true symmetry of  $\alpha$ -Mg<sub>2</sub>Al<sub>4</sub>Si<sub>5</sub>O<sub>18</sub> were lower than hexagonal, and if the shape of the unit cell coincided accidentally with that of a hexagonal one, probably the shape of the unit cell of the iron-analogue would not coincide, since the change in composition would cause some deviation of the unit cell from the hexagonal shape.

It is of some interest that the edge  $a$  of the iron-analogue is longer than that of  $\alpha$ -Mg<sub>2</sub>Al<sub>4</sub>Si<sub>5</sub>O<sub>18</sub>, while the edge  $c$  of the former is shorter than that of the latter. The cell volume of the former is  $781.8 \text{ \AA}^3$ ; that of the latter is  $776.1 \text{ \AA}^3$ .

Schairer and Yagi (1952) described the “iron-cordierite” as fibrous aggregates radially disposed and showing undulatory extinction under the microscope. “The crystals show negative elongation and indices were approximately  $\alpha = 1.551$ ,  $\beta = 1.564$ ,  $\gamma = 1.574$ .” They could not determine the optical angle directly. If the given indices be accurate, the “iron-cordierite” must be biaxial negative, probably owing to optical anomaly, as discussed in the next section.

*Anomalous optical properties.*—According to the original description by Rankin and Merwin (1918), the  $\alpha$ -form of  $Mg_2Al_4Si_5O_{18}$  is nearly or quite

TABLE 3  
Powder Diffraction Data, Synthetic "Iron Cordierite" ( $\text{Fe}_2\text{Al}_4\text{Si}_5\text{O}_{18}$ )

Indices $N, l$	Intensity	$2\theta^\circ$ obs	$2\theta^\circ$ calc	
1,0	100	10.42	10.36	} $\text{CuK}\alpha_1$
1,2	69	21.86	21.79	
3,2	129	26.40	26.37	
4,2	57	28.40	28.37	
7,1	66	29.26	29.27	
7,2	40	33.81	33.80	} $\text{CuK}\alpha$
12,0	14	36.42	36.42	
0,4	17	38.84	38.76	
12,2	17	41.38	41.44	
13,2	17	42.75	42.83	
13,3	17	48.31	48.35	
9,4	17	50.70	50.71	
?	14	53.50	?	
25,0	14	53.65	53.62	
12,4	34	54.17	54.23	

Note:  $N = h^2 + k^2 + hk$ . Filtered Cu radiation is used. The reading is taken at the mid-point of the peak width at approximately two-thirds of the peak height. ( $a = 9.860 \text{ \AA}$ ,  $c = 9.285 \text{ \AA}$ .)

uniaxial. Several later investigators, however, reported the formation of "synthetic magnesium cordierite" with varied optical angles, such as of  $8^\circ$ ,  $30^\circ$ ,  $50^\circ$ , and even  $82^\circ$ , in their dry experiments or in industrial processes (Dittler and Köhler, 1938; Shand, 1943; Sugiura, 1951; etc.). As mentioned before, the synthetic "iron-cordierite" may also be biaxial. We paid special attention to this fact, for it might suggest that those crystals were in some structurally intermediate states between the  $\alpha$ -form and natural cordierite.

We examined by means of the Geiger counter X-ray spectrometer a "magnesium cordierite" with  $2V = 8^\circ$ , synthesized by K. Sugiura (1951) by devitrifying glass of that composition. Its unit cell was found to have a hexagonal shape within the limit of experimental error. The unit cell of the synthetic "iron-cordierite" was also proved to have a hexagonal shape. As will be shown later, cordierite-like crystals in fused sediments from India have a unit cell with a hexagonal shape, though their optical angle is highly variable.

Thus, so far as our measurements are concerned, no deviation from hexagonal lattice was noticed on these crystals with varied optical angles. They are not in any intermediate state between the  $\alpha$ -form and natural cordierite, but in the  $\alpha$ -form proper. Their biaxial character is probably due to optical anomaly. Thus the most reliable diagnostic character for the identification of the  $\alpha$ -form is not the optical angle but the Geiger counter X-ray diffractogram.

Certain writers noticed, between crossed nicols, "twinning" in what appears to be the  $\alpha$ -form. Some of the "twins" are similar in appearance to ordinary twins of cordierite, while others are much more complicated. The nature of the "twins" is not known yet. They may be only apparent twin-like

optical structures, genetically related to optical anomaly, such as was already proved, for example, in garnet by Rinne (1925), and in milarite by Ito, Morimoto and Sadanaga (1952). It is interesting in this regard that crystals of the  $\alpha$ -form with larger optical angles generally tend to show more remarkable and complicated twin-like optical structure. For example, Rankin and Merwin whose crystals were nearly or quite uniaxial, did not notice twin-like optical structure, whereas Shand (1943), whose crystals were biaxial with an optical angle of about  $82^\circ$ , observed a very complicated twin-like optical structure.

ON  $\beta$ - $\text{Mg}_2\text{Al}_4\text{Si}_5\text{O}_{18}$

Dr. F. R. Boyd of the Geophysical Laboratory kindly synthesized for us cordierite-like crystals with composition  $\text{Mg}_2\text{Al}_4\text{Si}_5\text{O}_{18}$  by the hydrothermal method at  $775^\circ\text{C}$ . and under a water vapor pressure of 2000 bars. Judging from the conditions of their formation, they probably belong to the  $\beta$ -form of Karkhanavala and Hummel (1953). The crystals were so fine grained that reliable optical measurement was not possible. The powder diffraction data are shown in table 4 and figure 1.

TABLE 4  
Powder Diffraction Data,  $\beta$ -form of  $\text{Mg}_2\text{Al}_4\text{Si}_5\text{O}_{18}$

Indices $N, l$	Intensity	$2\theta^\circ$ obs	$2\theta^\circ$ calc.	
1,0	100	10.44	10.43	} CuK $\alpha$
3,0	25	18.10	18.11	
0,2	10	19.01	19.00	
1,2	64	21.74	21.72	
3,2	70	26.39	26.35	
4,2	80	28.43	28.40	
7,1	85	29.46	29.44	
7,2	21	33.89	33.89	
12,0	4	36.74	36.70	
9,2	—	—	—	
0,4	10	38.51	38.51	} CuK $\alpha_1$
13,1	2	39.45	39.48	
7,3	2	40.34	40.35	
12,2	4	41.59	41.62	
?	2	42.49	?	
13,2	6	43.00	43.02	
19,0	—	—	—	
13,3	6	48.49	48.47	
21,0	2	49.20	49.20	
21,1	6	50.25	50.22	
9,4	6	50.63	50.65	
12,4	13	54.24	54.24	

Note:  $N = h^2 + k^2 + hk$ . Filtered Cu radiation is used. The reading is taken at the mid-point of the peak width at approximately two-thirds of the peak height. ( $a = 9.792 \text{ \AA}$ ,  $c = 9.349 \text{ \AA}$ .)

The powder pattern is very similar to that of the  $\alpha$ -form, being perfectly indexed on the basis of the hexagonal unit cell with dimensions:  $a = 9.792$

$\text{\AA}$  and  $c = 9.349 \text{ \AA}$ . Thus the  $\beta$ -form is also probably hexagonal. This fact, taken in conjunction with the optical data given by Yoder (1952), suggests that the  $\beta$ -form is probably uniaxial negative, provided that it does not show anomalous biaxial character.

The structural relation between the  $\alpha$ - and  $\beta$ -forms is not known. The difference in structure between them, even if it exists, must be very slight. The  $\beta$ -form also is iso-structural with beryl.

## INDIALITE, A NEW MINERAL

It seemed to us that the most promising material for the occurrence of the  $\alpha$ -form was the famous fused sediments (so-called para-lava) in Bokaro coalfield, India. The rocks were found in 1916 by L. L. Fermor and are believed to have been produced by the fusion of sedimentary rocks due to the burning of underlying coal seams (Fermor, 1924). According to a personal communication from Dr. A. P. Subramaniam of the Geological Survey of India, the burning took place for some natural cause. Crystals of "cordierite" in the rocks were noted by Fermor and were recently described in detail by Venkatesh (1952, 1954). Dr. M. S. Krishnan and Mr. V. Venkatesh, of the Geological Survey of India, kindly sent us two specimens of the fused sediments on our request.

TABLE 5  
Powder Diffraction Data, Indialite from Bokaro Coalfield, India

Indices $N,l$	Intensity	$2\theta^\circ$ obs	$2\theta^\circ$ calc	
1,0	100	10.42	10.41	} CuK $\alpha$
3,0	18	18.05	18.08	
1,2	54	21.68	21.69	
—	18	25.63	Impurity	
3,2	43	26.31	26.31	} CuK $\alpha_1$
—	18	27.80	Impurity	
4,2	25	28.38	28.36	
7,1	54	29.38	29.38	
7,2	21	33.81	33.83	
0,4	14	38.47	38.47	
12,4	11	54.14	54.15	

Note:  $N = h^2 + k^2 + hk$ . Filtered Cu radiation is used. ( $a = 9.812 \text{ \AA}$ ,  $c = 9.351 \text{ \AA}$ ; Specimen 23/951.)

The specimens are mainly composed of the "cordierite" and glass. X-ray diffraction diagrams of the powdered rocks clearly show the pattern of either the  $\alpha$ - or  $\beta$ -form with a few additional peaks due to admixed other minerals. There is only one peak in the range of  $2\theta$  between  $29^\circ$  and  $30^\circ$  for CuK $\alpha$  radiation. Table 5 shows that the powder data of the specimen 23/951 are indexed completely on the basis of a hexagonal unit cell with dimensions:  $a = 9.812 \text{ \AA}$  and  $c = 9.351 \text{ \AA}$ . The crystals are colorless in thin sections. The refractive indices are  $\omega = 1.539$  and  $\epsilon = 1.534$  in specimen 23/951, and  $\omega = 1.537$  and  $\epsilon = 1.532$  in specimen 23/952. Judging from the fact that the crystals were formed by dry fusion, they do not belong to the  $\beta$ -form, but to the  $\alpha$ -form. If the crystals belonged to the  $\beta$ -form, they would be trans-

formed to the  $\alpha$ -form on heating at high temperatures, e.g. at 1000°C. Heating of the specimens at 1000°C. for 40 minutes, however, did not cause any appreciable change in their refractive indices. Thus the "cordierite" is actually the  $\alpha$ -form of  $\text{Mg}_2\text{Al}_4\text{Si}_5\text{O}_{18}$ , in which a part of Mg is replaced by  $\text{Fe}^{+2}$ . If we assume that the unit cell dimensions and refractive indices of the  $\alpha$ -form vary linearly with progressive replacement of Mg by  $\text{Fe}^{+2}$ , the Mg: $\text{Fe}^{+2}$  ratio of the "cordierite" in specimen 23/951 is about 7:3. Thus the  $\alpha$ -form is a new mineral, and the name "indialite" is proposed for it, as stated before.

The optical properties of the indialite are of some interest. Specimen 23/951 contains numerous minute indialite crystals, most of which do not show twin-like optical structure between crossed nicols. According to conoscopic observations, some of them are completely or nearly uniaxial negative. In other crystals, the central portion is uniaxial negative, while the marginal portion is biaxial with optical angles about X smaller than 30°. Specimen 23/952 contains slightly larger indialite crystals, which show complicated twin-like optical structure between crossed nicols. The optical angle of the crystals is variable, ranging generally from 50° to 90° about X. Thus the degree of optical anomaly seems to be variable.

Venkatesh (1954) described the twin-like optical structure of the indialite in detail. Though he considered that it is a kind of true twinning of cordierite, he noticed differences in shape between it and ordinary twinning of cordierite. He states: "The type of twinning found in cordierite seems to bear some relation to the nature of the rock in which it occurs. . . . Simple and polysynthetic twinning are the most common types in coarse to medium grained, gneissic or granular rocks . . . . Some examples of sixlings and trillings in cordierites from gneisses, pegmatites, etc. have been recorded in the literature . . . . Cyclic twinning producing the complex radial and concentric patterns, are totally unrepresented in the rocks mentioned above and have been observed only in specimen No. 7 . . . ." Specimen No. 7 in the above-quoted sentence means the fused sediments now under consideration.

In the future, indialite may be found in some ordinary pyrometamorphic rocks such as inclusions in basalt.

#### HIGH AND LOW CORDIERITES

The probable existence of two kinds of natural cordierites is revealed by the relation between the refractive indices and the chemical compositions.

Reliable chemical and optical data of cordierites from metamorphic rocks are shown in table 6 in the order of increasing  $(\text{Fe}^{+2} + \text{Mn})/(\text{Mg} + \text{Fe}^{+2} + \text{Mn})$  ratio. They are plotted in figure 2, which shows that the  $\beta$  index of these minerals increases generally with the  $(\text{Fe}^{+2} + \text{Mn})/(\text{Mg} + \text{Fe}^{+2} + \text{Mn})$  ratio. Some of the points do not fall exactly on a line, probably because the refractive indices are slightly influenced by factors other than the above ratio.

Reliable chemical and optical data of cordierites from volcanic rocks are shown in table 7 and figure 2. The  $\beta$  index of these minerals also increases generally with the  $(\text{Fe}^{+2} + \text{Mn})/(\text{Mg} + \text{Fe}^{+2} + \text{Mn})$  ratio.

It is remarkable that the volcanic cordierites have lower refractive indices than the metamorphic cordierites with the same  $(\text{Fe}^{+2} + \text{Mn})/(\text{Mg} +$

Fe<sup>+2</sup> + Mn) ratio, as shown in figure 2. The former were probably formed at higher temperatures than the latter. As shown in the next section, the volcanic and metamorphic cordierites are reasonably considered to represent two different kinds, which are probably polymorphic modifications. The kind represented by volcanic cordierites is called "high cordierite," while that represented by metamorphic cordierites is called "low cordierite." (In this paper cordierites in inclusions in igneous rocks are treated not as metamorphic cordierites but as igneous cordierites, for such cordierites are considered to have been formed under similar physical conditions with the igneous rocks.)

The two kinds both have large optical angles.

TABLE 6  
Atomic Ratios of Cordierites from Metamorphic Rocks,  
Calculated on the Basis of O = 18 (excluding Oxygen in H<sub>2</sub>O)

Nos.	1	2	3	4	5	6	7	8	9
Si	5.02	4.93	5.04	5.02	4.99	4.95	5.17	5.07	5.00
Al	0.98	1.07	0.96	0.98	1.01	1.05	0.83	0.93	1.00
Al	2.92	2.75	2.93	2.60	3.00	3.00	3.03	2.99	2.98
Ti	0.03	0.00	—	—	0.00	0.00	—	0.00	—
Fe <sup>+3</sup>	0.11	0.08	0.02	0.17	0.05	0.04	0.06	0.03	0.05
Fe <sup>+2</sup>	0.19	0.28	0.55	0.61	0.70	0.72	0.75	0.76	0.84
Mg	1.64	1.86	1.46	1.48	1.18	1.21	1.03	1.03	1.08
Mn	0.01	0.10	0.01	0.04	0.00	0.02	—	0.01	0.01
Ca	0.03	0.00	—	0.21	0.06	0.02	—	0.06	0.01
Na	0.03	0.07	—	—	0.05	0.04	—	0.09	0.05
K	0.01	0.00	—	—	0.03	0.00	0.01	0.05	0.01
H <sub>2</sub> O +	0.46	0.96	0.36	0.64	0.64	0.23	0.49	0.21	0.67
H <sub>2</sub> O -	0.03	—			0.19	0.00		0.00	—
$\frac{Fe^{+2} + Mn}{Mg + Fe^{+2} + Mn} \times 100$	10.9	17.0	27.8	30.4	37.2	37.7	42.1	42.6	44.1
<i>d</i>	2.588	—	2.588	2.598	2.64	2.631	2.650	—	—
<i>α</i>	1.527	1.534	1.537	—	1.538	1.544	1.543	1.543	1.547
<i>β</i>	1.532	(1.538)	1.543	—	1.542	1.550	1.548	1.550	1.552
<i>γ</i>	1.538	1.543	1.548	—	1.547	1.556	1.553	1.557	1.557
2V over Z	88°	76°	99°	81°-95°	84°	85°-95°	89°	90°	94°-103°

Note: Al atoms are divided into two groups, the upper of which belongs to the pseudo-hexagonal rings. The contents of H<sub>2</sub>O + and H<sub>2</sub>O - are given in molecular ratios on the same basis.

1. From cordierite-anthophyllite gneiss at Attu. By Pehrman (1932).
2. From cordierite gneiss in Antarctica. By Tilley (1940).
3. From cordierite-anthophyllite rock at Pernio. By Eskola (1914).
4. From garnet-sillimanite-cordierite gneiss in Madura. By Krishnan (1924).
5. From garnet-spinel-cordierite-biotite-hornfels at Belhelvie. By Stewart (1942).
6. From garnet-sillimanite-spinel-cordierite gneiss in Great Slave Lake area. By Folinsbee (1941).
7. From veined gneiss at Ilmajoki. By Pehrman (1932).
8. From metamorphic cordierite deposit in Laramie Range. By Newhouse and Hagner (1949).
9. From a metasediment in Kalanti. By Hietanen (1943).

TABLE 7  
Atomic Ratios of Cordierites from Volcanic Rocks,  
Calculated on the Basis of O = 18 (excluding Oxygen in H<sub>2</sub>O)

Nos.	1	2
Si	4.95	4.98
Al	1.05	1.02
Al	2.79	2.90
Ti	—	—
Fe <sup>+3</sup>	0.11	0.24
Fe <sup>+2</sup>	0.61	0.79
Mg	1.51	1.01
Mn	—	—
Ca	0.05	—
Na	—	—
K	—	—
H <sub>2</sub> O +	0.38	—
H <sub>2</sub> O -	—	—
$\frac{Fe^{+2}+Mn}{Mg+Fe^{+2}+Mn} \times 100$	28.7	43.9
<i>d</i>	—	—
$\alpha$	1.530	—
$\beta$	1.534	1.5438
$\gamma$	1.537	—
2V over Z	99°	94°

1. From biotite-hornblende andesite at Kasyo-to. By Ichimura (1936).
2. From garnet-cordierite-mica andesite at Cabo de Gata. By Osann (1888).

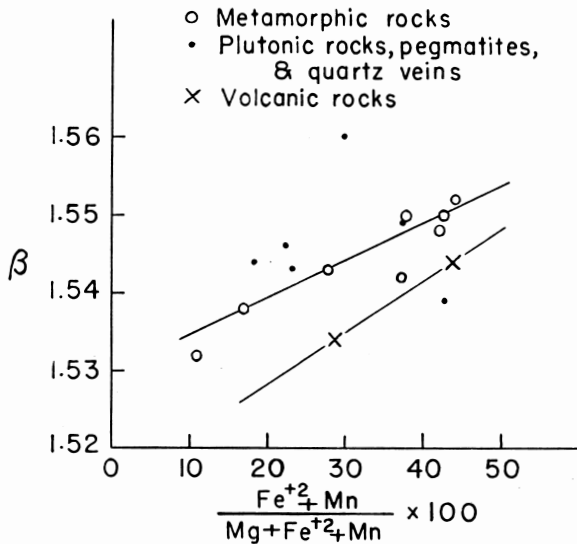


Fig. 2. The  $\beta$  index of cordierites in various modes of occurrence. The symbols indicate the host rocks.

## HEATING EXPERIMENTS OF NATURAL CORDIERITES

*Localities and modes of occurrence of the samples*

(1) Kasyo-to is a small volcanic island, about 30 km east of Formosa (Taiwan). Cordierite occurs on the island as porphyritic individuals, up to 1 cm long, in hypersthene-hornblende andesite, olivine-hornblende andesite, and olivine-hypersthene-hornblende andesite (Ichimura, 1936).

(2) Asama is an active volcano, 2542 m high, lying about 130 km north-west of Tokyo. It is mainly composed of augite-hypersthene andesite which contains numerous cordierite-bearing inclusions, probably derived from sedimentary rocks. The inclusions are composed of quartz and cordierite, together with varied amounts of andalusite, sillimanite (mullite ?), plagioclase, opaque mineral and glass. The highest temperature of lava in the crater was about 1050°C, according to T. Minakami.

(3) Cordierite deposits in the Laramie Range, Albany County, Wyoming, were formed by metasomatic replacement in metanorite (Newhouse and Hagner, 1949).

(4) The sample of cordierite from Orijarvi is a large crystal, probably formed by metasomatism (Eskola, 1914; etc.).

(5) At Nyoidake, east of Kyoto, cordierite occurs in cordierite-biotite hornfels in the inner aureole around a granite mass.

*Preliminary heating experiment at 1000°-1050°C.*—Natural cordierites were heated at 1000°-1050°C. in an electric furnace in air and taken out to cool. The refractive indices for yellow light were measured on the cooled samples by the immersion method. The results are shown in table 8.

Thus, low cordierites show notable decrease in  $\beta$  index after a short period of heating. On the other hand, high cordierites show no appreciable change in  $\beta$  index on short heating, though they show slight increase in  $\beta$

TABLE 8  
Heating Experiments of Cordierites and Osumilite at 1000°-1050°C.

Kinds	Locality	Host-rock	Refr. indices before heating	Refr. indices after heating	Time of heating (in min.)
High cordierite	Kasyo-to	Andesite	$\beta=1.533$	$\beta=1.533$	20
	Asama	Xenolith in andesite	$\beta=1.540$	$\beta=1.540$	20
	Asama	Xenolith in andesite	$\beta=1.543$	$\beta=1.545$	80
Low cordierite	Laramie Range	Metamorphic deposit	$\beta=1.548$	$\beta=1.543$	10
	Orijärvi	Metamorphic rock	$2V_z=92^\circ-103^\circ$ $\beta=1.539$	$2V_z=93^\circ-110^\circ$ $\beta=1.535$	10
	Nyoidake	Hornfels	$\alpha=1.549$	$\alpha=1.541$	60
			$\beta=1.556$	$\beta=1.545$	
$\gamma=1.561$			$\gamma=1.548$		
Osumilite	Sakkabira	Plagioliparite	$\omega=1.549$	$\epsilon=1.549$	10

index on prolonged heating, probably owing to gradual oxidation of the iron contained in the mineral.

The decrease in  $\beta$  index of low cordierites is of the same order of magnitude as the difference in refractive indices between high and low cordierites having the same  $(\text{Fe}^{+2} + \text{Mn})/(\text{Mg} + \text{Fe}^{+2} + \text{Mn})$  ratios. Hence, low cordierites acquire, on heating at  $1000^{\circ}$ - $1050^{\circ}\text{C}$ ., optical properties similar to those of high cordierites. Thus it is suggested that low cordierites are transformed to high cordierites on heating.

*Change in refractive indices of Laramie Range cordierites on heating.*—In order to elucidate the process of decrease in refractive indices of low cordierites on heating, systematic heating experiments were carried out on two specimens of cordierites from Laramie Range.

The procedure of the experiments is as follows. Many small samples were prepared from the specimens. Each sample was heated at a certain temperature, and then cooled in air. The refractive indices for yellow light were measured on the cooled samples by the immersion method.

*Specimen 1:*  $\text{FeO} = 7.06\%$ . The refractive indices before heating were  $\alpha = 1.542$ ,  $\beta = 1.548$ , and  $\gamma = 1.555$ . The results of heating are shown in table 9 and figure 3. On heating at  $700^{\circ}\text{C}$ ., or lower temperatures, the indices remained practically unchanged. On heating at  $800^{\circ}\text{C}$ ., or higher temperatures, the indices decreased within 10 minutes to the following values:  $\alpha = 1.536$ ,  $\beta = 1.543$ ,  $\gamma = 1.550$ . The optical angles were large before as well as after heating. Thus the transition took place at temperatures between  $700^{\circ}$  and  $800^{\circ}\text{C}$ ., in this case.

*Specimen 2:*  $\text{FeO} = 1.71\%$ . This is exceptionally poor in FeO among natural cordierites. Thus the transition can hardly be ascribed to any change of the state of iron in the structure. The refractive indices before heating were  $\alpha = 1.536$ ,  $\beta = 1.541$ ,  $\gamma = 1.546$ .

At first the samples were heated at varied temperatures for 10 minutes each. The results were shown in table 10 and figure 4. Thus the refractive indices decreased, not abruptly at a definite temperature, but gradually through the temperature range from  $930^{\circ}$  to  $1000^{\circ}\text{C}$ ..

TABLE 9  
Changes in Refractive Indices of the Laramie  
Range Cordierite 1 on Heating

Temp. in degrees C.	Time of heating in minutes			
		10	20	
	$\alpha$	$\beta$	$\gamma$	$\beta$
Room temp.	1.542	1.548	1.555	1.548
350		1.548		
400		1.548		
500				1.547
600		1.548		1.548
700	1.542	1.548	1.555	1.547
800	1.536	1.544	1.550	1.544
900		1.543		1.544
1000	1.536	1.543	1.550	

TABLE 10  
Changes in Refractive Indices of the Laramie  
Range Cordierite 2 on Heating

Temp. in degrees C.	Time of heating in minutes			
	$\alpha$	$\beta$	$\gamma$	$\beta$
Room temp.	1.536	1.541	1.546	1.541
590				1.541
690				1.537
800	1.536	1.541	1.546	1.532
900	1.536	1.541	1.545	1.530
930	1.536	1.540	1.545	
950	1.533	1.536	1.543	
960	1.532	1.535	1.539	
970	1.530	1.533	1.538	
980	1.528	1.533	1.538	
1000	1.525	1.530	1.535	
1020	1.525	1.530	1.535	

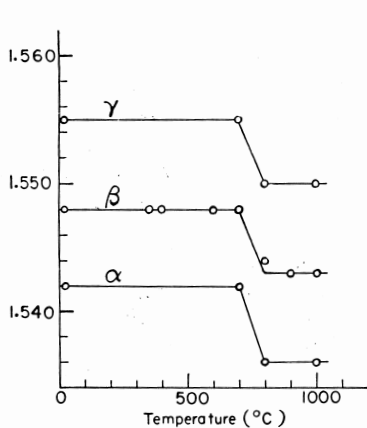


Fig. 3. Decrease in refractive indices of Laramie Range cordierite no. 1 on heating at varied temperatures for 10 minutes each.

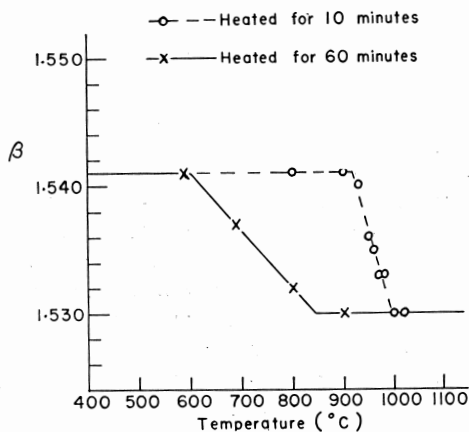


Fig. 4. Decrease in  $\beta$  index of Laramie Range cordierite no. 2 on heating at varied temperatures for 10 minutes each and for 60 minutes each.

Then the samples were heated at varied temperatures for 60 minutes each. The results are also shown in table 10 and figure 4. The transition took place through the temperature range from 590° to 900°C. in this case. Thus prolonged heating caused transition at lower temperatures.

A definite temperature range for the equilibrium transition is very difficult to establish, because longer heating at lower temperatures may cause transition as well. The gradual decrease in refractive indices suggests that the transition is of the second order. The transition from high to low cordierite does not take place in any case, so far as we have tried. The preservation of high cordierite in volcanic rocks is in harmony with this fact.

*Change in optical angle of Nyoidake cordierites on heating.*—In order to elucidate the process of change in optical angle of low cordierite on heat-

ing, a systematic heating experiment of cordierites from cordierite-biotite hornfels at Nyoidake was carried out. The crystals showed ordinary pseudo-hexagonal twinning. The two sectors in opposite positions of a twinned crystal have the same optical angle, though different pairs of sectors have generally different optical angles. Thus, for example, the optical angle 2V over Z in the three pairs of twinning sectors of an unheated crystal were  $98^\circ$ ,  $104^\circ$ , and  $114^\circ$ . In another unheated crystal, they were  $96^\circ$ ,  $100^\circ$ , and  $114^\circ$ .

TABLE 11  
Changes in Optical Angle of Nyoidake Cordierites  
on Heating around  $1000^\circ\text{C}$ .

Time of heating in hours	Number of twinning crystals measured	2V over Z
Before heating	3	$96^\circ$ — $114^\circ$
1	2	$80^\circ$ — $104^\circ$
4	2	$84^\circ$ — $102^\circ$
33	4	$85^\circ$ — $94^\circ$

Such cordierites were heated around  $1000^\circ\text{C}$ . for various lengths of time. The results are shown in table 11. Thus 2V over Z became slightly smaller on heating within the first hour, and did not change any more even with a very long period of further heating. In all cases before and after heating the optical angles were large. (During the heating, borders of the cordierite grains became gradually reddish brown, probably through oxidation, while the central portions were colorless even after 50 hours of heating.)

Probably the change in optical angle took place within the first 10 minutes, simultaneously with the decrease in refractive indices.

*Emission of water from Laramie Range cordierite on heating.*—Natural cordierites have variable contents of water. The water is unessential to the mineral, being probably stuffed in the pseudo-hexagonal tunnels of the structure as  $\text{H}_2\text{O}$  molecules.

The water content of a high cordierite shown in table 7 is of the same order as those of low cordierites in table 6. Hence the systematic difference in refractive indices between high and low cordierites can not be ascribed to a difference in water content, so far as these chemical analyses are reliable.

This view has been confirmed by the results of the following heating experiments. The specimen used in this case is the same as specimen 1 of Laramie Range cordierite, previously used in the measurement of change in refractive indices on heating. The specimen before heating contained 7.06% FeO, 1.63%  $\text{H}_2\text{O}+$ , and 0.00%  $\text{H}_2\text{O}-$ . Heating at  $650^\circ\text{C}$ . for 10 minutes did not cause any decrease in refractive indices, while heating at  $800^\circ\text{C}$ . for 10 minutes caused a decrease as shown before. The two samples, heated at  $650^\circ$  and  $800^\circ\text{C}$ ., were again heated strongly on the Mecker burner to determine their ignition losses. The ignition loss of the sample previously heated at  $650^\circ\text{C}$ . was 0.86%, while that of the sample previously heated at  $800^\circ\text{C}$ . was 0.69%. Thus, the difference in ignition loss is only 0.17%, that is, only one-tenth of the original water content, in spite of taking place of

the transition between 650° and 800°C. The transition has probably no serious connection with the loss of water on heating.

*X-ray data.*—Diffraction diagrams of an iron-poor cordierite from Laramie Range (specimen 2 in heating experiment) were taken before and after heating that caused transition, as shown in table 12 and figure 1. The most remarkable change is an increase in intensity of a peak with  $2\theta$  at about 21.7° for  $\text{CuK}\alpha$  radiation. The change in unit cell dimensions is almost imperceptible.

TABLE 12  
Powder Diffraction Data of Cordierites

High cordierite from volcano Asama (unheated)		Cordierite from Laramie Range, heated around 1000°C. for 10 minutes		Low cordierite from Laramie Range (unheated)	
$2\theta$	I	$2\theta$	I	$2\theta$	I
10.30	100	10.36	100	10.36	100
17.92	13	18.00	23	18.04	27
18.16	7	18.20	9	18.23	12
18.96	10	18.96	46	18.98	15
21.70	73	21.68	115	21.70	55
26.29	70	26.28	75	26.30	67
26.36	54	26.39	72	26.40	60
28.24	31	28.28	47	28.30	35
28.40	47	28.44	75	28.47	57
29.18	42	29.26	55	29.31	71
29.26	43	29.35	52	29.40	73
29.51	34	29.58	40	29.59	38
33.68	22	33.79	20	33.82	28
33.94	10	34.01	8	34.05	12

Note: Filtered Cu radiation is used. The reading is taken at the top of the peak. Thus the values are practically those for  $\text{CuK}\alpha_1$ .

Diagrams for a few other low cordierites were also compared with those of the same samples after heating that caused transition. Some of them showed changes in intensities of peaks in a similar way but to a smaller extent, and others showed little change. Such a change in intensity may be interpreted in various ways. It may be due to slight structural changes, though it is possibly due to some change in preferred orientation of powder grains.

The crystal structure of cordierite was studied in detail by K. Takane and T. Takeuchi (1936) and also by A. Byström (1942). The former investigators used a specimen from a quartz vein at Hitati, which probably belongs to low cordierite (table 14). The latter used a specimen from Risör, a part of which we obtained from Mrs. A. Byström through arrangements made by Prof. C. Frondel of Harvard University. It showed notable decrease in refractive indices on heating as follows:

$$\alpha = 1.530, \beta = 1.535, \gamma = 1.539 \text{ before heating, and}$$

$\alpha = 1.522, \beta = 1.527, \gamma = 1.532$  after heating around 1000°C. for 10 minutes. Therefore it also belongs to low cordierite.

Though all these investigators adopted the point group  $mmm$  ( $D_{2h}$ ) and the space group  $Cccm$  ( $D_{2h}^{20}$ ) as a basis of their structure studies, Takane and Takeuchi state that they observed, under a favorable condition, very weak reflections incompatible with this space group—such as (401), (601), (801), and (10,0,1). If such reflections are taken into consideration, and if the point group  $mmm$  is adopted, the space group of low cordierite is probably  $Ccmm$  ( $D_{2h}^{17}$ ). We think, however, that the point group of cordierite is not certain. Hence the space group of high and low cordierites should be regarded as uncertain.

Thus, though we have not reached any definite conclusion on this problem, it is conceivable that the structural difference between high and low cordierites lies in the manner of distribution of Al, Mg, and  $Fe^{+2}$  atoms between four- and six-coordinated positions linking the pseudo-hexagonal rings.

TABLE 13

Atomic Ratios of Cordierites from Plutonic Rock, Pegmatites, and Quartz-Veins, Calculated on the Basis of O=18 (excluding Oxygen in  $H_2O$ )

Nos.	1	2	3	4	5	6	7	8
Si	4.97	5.02	5.05	4.94	4.94	4.95	4.83	4.83
Al	1.03	0.98	0.95	1.06	1.06	1.05	1.17	1.17
Al	2.92	2.96	2.95	2.88	2.95	3.05	2.84	2.81
Ti	—	—	—	—	0.00	—	—	—
$Fe^{+3}$	0.06	0.03	0.10	0.09	0.01	0.03	0.13	0.27
$Fe^{+2}$	0.37	0.43	0.42	0.57	0.74	0.80	1.22	1.43
Mg	1.67	1.57	1.46	1.35	1.25	1.09	0.45	0.25
Mn	—	0.02	0.02	—	0.01	0.01	0.27	0.11
Ca	—	—	—	—	0.00	—	0.02	0.01
Na	—	—	—	0.28	0.11	—	0.28	0.76
K	—	—	—	0.07	0.06	—	0.05	0.13
$H_2O^+$	} 0.56 {	} 0.56 {	1.15	0.38	0.69	} 0.43	1.27	0.99
$H_2O^-$				0.04	0.07		—	—
$\frac{Fe^{+2}+Mn}{Mg+Fe^{+2}+Mn} \times 100$	18.2	22.3	23.2	29.7	37.5	42.7	76.7	86.1
$d$	2.593	2.607	2.632	2.660	2.63	—	—	—
$\alpha$	1.5392	1.5403	1.5364	1.5520	1.543	1.536	1.555	1.558
$\beta$	1.5443	1.5460	1.5431	1.5599	1.549	1.539	1.566	1.568
$\gamma$	1.5475	1.5483	1.5462	1.5610	1.555	1.543	1.571	1.573
$2V$ over $Z$	$110^\circ$	$115^\circ$	$112^\circ$	$137^\circ$	$80^\circ$	$80^\circ$	$114^\circ$	$111^\circ$

1. From quartz vein cutting mica-schist at Mont Bity. By Duparc, Wunder, and Sabot (1911).
2. From pegmatite cutting gneiss at Guilford. By Farrington (1892) and Oppenheimer (1914).
3. From andalusite-bearing muscovite-biotite-cordierite quartz vein cutting schistose metamorphic rocks at Hitati. By Takeuchi (1935).
4. From pegmatite at Haddam. By Oppenheimer (1914).
5. From quartz monzonite in New Hampshire. By Heald (1950).
6. From cordierite-biotite-microcline-andesine pegmatite at Degero. By Pehrman (1932).
7. From garnet-andalusite-bearing biotite-microcline-plagioclase pegmatite at Dosi. By Shibata (1936).
8. From cordierite-biotite-microcline pegmatite at Sasago. By Shibata (1936).

CORDIERITES FROM HYPABYSSAL ROCKS, PLUTONIC ROCKS,  
PEGMATITES, AND QUARTZ VEINS

The natural cordierites hitherto treated in this paper were from volcanic and metamorphic rocks. It is interesting to examine cordierites of other modes of occurrence.

Chemical and optical data of cordierites from a plutonic rock, pegmatites and quartz veins are shown in table 13 and figure 2. Most of these cordierites show still higher refractive indices than metamorphic cordierites. This is probably due, at least partly, to a systematic difference in alkali content between these and metamorphic cordierites. If so, heating experiment is necessary to decide which modification a particular cordierite belongs to.

Four cordierites from pegmatites and quartz vein were heated around 1000°C. as shown in table 14. They showed notable decreases in refractive indices. Hence they are low cordierites.

A big cordierite xenocryst in biotite-granite porphyry from Seto, Ariimura, Minami-muro-gun, Mie prefecture, was heated around 1000°C. for 20 minutes. The  $\beta$  index was 1.542 before heating, and became 1.537 after heating. Thus it also belongs to low cordierite. (The biotite-granite porphyry occurs in genetical association with rhyolite. All the volcanic cordierites that we examined came from basaltic and andesitic rocks. It will be of interest to determine in the future which modification cordierites from rhyolite belong to.)

TABLE 14  
Heating Experiments of Cordierites from Pegmatite and  
Quartz-Vein around 1000°C.

Locality	Host-rock	Refr. index before heating	Refr. index after heating	Time of heating in minutes
Hitati, Japan	Quartz-vein	$\beta=1.546$	$\beta=1.535$	15
Haddam, Conn.	Pegmatite	$\beta=1.561$	$\beta=1.543$	10
Guilford, Conn.	Pegmatite	$\left\{ \begin{array}{l} \alpha=1.539 \\ \beta=1.546 \\ \gamma=1.550 \end{array} \right.$	$\left\{ \begin{array}{l} \alpha=1.532 \\ \beta=1.536 \\ \gamma=1.539 \end{array} \right.$	10
Micanite, Colorado	Pegmatite	$\left\{ \begin{array}{l} \alpha=1.560 \\ \beta=1.568 \\ \gamma=1.572 \end{array} \right.$	$\left\{ \begin{array}{l} \alpha=1.544 \\ \beta=1.549 \\ \gamma=1.555 \end{array} \right.$	10

## STABILITY RELATION BETWEEN INDIALITE AND CORDIERITE

There are high-low inversion relations between high and low indialites and between high and low cordierites, as stated above. However, the stability relation between indialites and cordierites is not certainly known yet. So far as we have tried, indialite and cordierite can not be transformed to each other.

Cordierite occurs in various kinds of rocks, which were formed at widely different temperatures.<sup>2</sup> For example, cordierite in low grade metamorphic

<sup>2</sup> Prof. F. Stella Starrabba of the University of Catania kindly provided us with a specimen of an inclusion containing cordierite from the Etna lava of the year 1669. The X-ray spectrogram confirmed that the mineral is truly cordierite. (The 1669 lava is composed of olivine-augite andesine-basalt.)

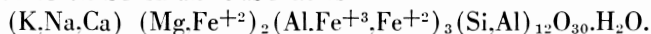
rocks was formed probably at low temperatures (presumably at 300°C. or so), while cordierite in volcanic rocks was formed probably at high temperatures (presumably at 700°-1200°C.). On the other hand, indialite is very rare in nature. Probably high and low cordierites are stable at high and low temperatures respectively below the decomposition point, while high and low indialites are metastable in all cases.

The existence of crystals in some structurally intermediate state between indialites and cordierites is not impossible, as the crystal structures of cordierites are derived by slight deformations from those of indialites. However, we have not found such an example.

The results of the present study are synoptically shown in table 15.

#### RELATION OF OSUMILITE TO CORDIERITE

At the beginning of our study of cordierite, A. Miyashiro was interested in the fact that nearly or completely uniaxial positive "cordierite" was reported to occur characteristically in volcanic rocks. He thought that such "cordierite" might represent a high temperature form of the mineral. He made a detailed study of one such uniaxial positive "cordierite." The sample examined is from biotite-bearing hypersthene plagioliparite in Sakkabira, Tarumizu-mati, Kagosima prefecture, Kyusyu, Japan. He found that it is not cordierite, but a new hexagonal mineral. It was named "osumilite" after Osumi, the province to which Sakkabira belongs (Miyashiro, 1953, 1955). The theoretical formula is as follows:



The quantities of (K,Na,Ca) and H<sub>2</sub>O are probably variable to some extent (table 15).

So far as we have observed, all osumilite crystals are optically uniaxial positive. But certain persons have stated that some crystals are optically biaxial with variable optical angles. Indeed, some osumilite may show anomalous biaxial character. Osumilite, being composed of hexagonal double rings (Si,Al)<sub>12</sub>O<sub>30</sub>, is iso-structural with milarite, which is optically biaxial in spite of exhibiting hexagonal symmetry for X-ray as well as for morphological study.

Osumilite from Sakkabira occurs in the groundmass and in cavities of the host rock. As discussed in another paper (Miyashiro, 1955), descriptions are found in the literature of minerals which were identified as cordierite but we think to be probably osumilite, judging from their optical and chemical properties. They all occur in the groundmass or cavities or in inclusions in volcanic rocks. Recently Aoki (1954) reported another example of what appears to be osumilite in the groundmass of hypersthene-bearing hornblende-biotite plagioliparite (or biotite plagioliparite) in the vicinity of Haneyama, Oita prefecture, Kyusyu. Thus osumilite occurs only in volcanic rocks as a constituent of the groundmass or in cavities or as a constituent of inclusions, but not as porphyritic crystals, so far as we are aware at present.

On the other hand, high cordierite occurs only in volcanic rocks as porphyritic crystals or as a constituent of inclusions. (It is not known whether

TABLE 15  
Polymorphs of  $Mg_2Al_4Si_5O_{18}$ , together with Osumilite

	System	Space group	Struct. type	Unit cell (A)	Optical angle	Refr. indices	Modes of occurrence or formation
High indialite ( $\alpha$ - $Mg_2Al_4Si_5O_{18}$ )	Hex.	$C \frac{6}{m}$	Single rings	$a=9.782$ $c=9.365$	Uniaxial negative	$\epsilon=1.524$ $\omega=1.528$	In fused sediments; synthesized above 830°C.
Low indialite ( $\beta$ - $Mg_2Al_4Si_5O_{18}$ )	Hex.	$C \frac{6}{m}$ (?)	Single rings	$a=9.792$ $c=9.349$	Uniax. (?) negative	$\epsilon=1.537$ $\omega=1.541$	Synthesized below 830°C.
High cordierite	Pseudo-hex. (Rhombic?)	?	Single rings	$a=17.1$ $b=9.7$ $c=9.3$	$2V_z=80^\circ$ $110^\circ$	$\alpha=1.53$ $1.54$ $\gamma=1.54$ $1.55$	In volcanic rocks
Low cordierite	Pseudo-hex. (Rhombic?)	?	Single rings	$a=17.1$ $b=9.7$ $c=9.3$	$2V_z=75^\circ$ $140^\circ$	$\alpha=1.52$ $1.56$ $\gamma=1.53$ $1.57$	In metamor. rocks, plutonic rocks, etc.
$\mu$ - $Mg_2Al_4Si_5O_{18}$	?	?	$\beta$ -spod. (?)	?	?	1.53- 1.56	Synthesized at 850°- 900°C.
Osumilite	Hex.	$C \frac{6}{m}$	Double rings	$a=10.17$ $c=14.34$	Uniaxial positive	$\omega=1.545$ $1.547$ $\epsilon=1.549$ $1.551$	In volcanic rocks

Note: We ourselves have no evidence to prove or disprove the existence of the  $\beta$ -form (low indialite), distinct from the  $\alpha$ -form and also from cordierites. It is not certain whether the so-called  $\mu$ -form has composition  $Mg_2Al_4Si_5O_{18}$  or not.

the porphyritic crystals are true phenocrysts crystallized from magma, or xenocrysts incorporated from outside.)

As is well known, minerals which are stable at high temperatures or low pressures or both, such as tridymite, cristobalite, and pigeonite, occur in the groundmass of volcanic rocks, but very rarely as porphyritic crystals. Since osumilite occurs in the groundmass, but not as porphyritic crystals, it may be stable at high temperatures, low pressures, or both.

The chemical composition of osumilite differs slightly from that of cordierite. Hence, chemical environment also will play a role in controlling the formation of one or other of them. For example, as osumilite is higher in alkali content than cordierite, high concentration of alkalis in acid residuum during crystallization of volcanic magma may favor the formation of osumilite in the groundmass and in cavities. As osumilite can accommodate iron perhaps more easily than cordierite, high concentration of iron in acid residuum also may play a role in this regard.

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