OSUMILITE, A NEW SILICATE MINERAL, AND ITS CRYSTAL STRUCTURE*

AKIHO MIYASHIRO, *Geological Institute, Faculty of Science,*
Tokyo University, Tokyo, Japan.

ABSTRACT

Osumilite (K, Na, Ca)(Mg, Fe^{+2})$_2$(Al, Fe^{+3}, Fe^{+2})$_3$(Si, Al)$_{12}O_{30}\cdot H_2O$ is dihexagonal-dipyramidal ($6/m\ 2/m\ 2/m = D_{6h}$). Space group $P\ 6/mcc = D_{6h}{}^2$. Unit cell: $a_0 = 10.17$ Å, $c_0 = 14.34$ Å; $a_0 : c_0 = 1 : 1.410$; containing two molecules. The crystals are black, prismatic or tabular. G = 2.64. Uniaxial positive. Indices of refraction (for Na): $\omega = 1.545 - 1.547$, $\epsilon = 1.549 - 1.551$; ϵ-$\omega = 0.004$. Dichroic with ω = light blue and ϵ = colorless.

Structurally, osumilite is composed of hexagonal double rings (Si, Al)$_{12}O_{30}$, connected by metal atoms (Al, Fe^{+3}, Fe^{+2}) in 4-coordination, (Mg, Fe^{+2}) in 6-coordination, and (K, Na, Ca) in 12-coordination. It is iso-structural with milarite.

Osmulite was found in a volcanic rock at Sakkabira, Kyusyu, Japan, and is named after the province Ôsumi. Probably osumilite is a relatively common mineral, but always has been mistaken for cordierite. It appears to occur characteristically in volcanic rocks and inclusions in them.

INTRODUCTION

Many mineralogists have tried unsuccessfully to correlate the optical properties of cordierites with their chemical compositions. The writer thought that the difficulty in correlation might be due, at least partly, to the existence of polymorphism in cordierite. This thought prompted him to begin a study of cordierite. It was soon found that there is a close relationship between the modes of occurrence and the optical properties of the mineral. Of particular interest is the fact that nearly or completely uniaxial positive cordierite is reported to occur characteristically in volcanic rocks. The writer considered that such material might represent a high-temperature form of cordierite. Then, he examined a uniaxial positive "cordierite" from a volcanic rock in Sakkabira, Hayasaki, Tarumizumati, Kimotuki-gun, Kagosima Prefecture, Kyusyu (Kyushu), Japan, and found that it is not cordierite, but actually is a new *hexagonal* mineral, here named *osumilite* after Ôsumi, the name of an old province in Sakkabira.

The locality is near the southeastern foot of the famous volcano Sakurazima (Sakurajima). Morimoto (1948) and Morimoto and Minato (1949) described the mode of occurrence, physical properties, and chemical composition of the mineral in detail, though they considered it to be cordierite. The writer owes very much to their descriptions.

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MODE OF OCCURRENCE

According to Morimoto and Minato (1949), the osumilite occurs in biotite-bearing hypersthene plagioliparite (rhyodacite) which contains andesine, quartz, hypersthene, biotite, and magnetite as phenocrysts scattered in the groundmass. Yamaguchi (1938) gives three chemical anlayses of the rock in this neighborhood, the ranges of each component being given in Table 1.

Osumilite is present in the brittle porous part of the plagioliparite and not in the hard compact portion. Euhedral crystals occur in association with tridymite and quartz in drusy cavities in the brittle porous part, whereas anhedral crystals occur in the groundmass. The groundmass is comparatively highly crystalline, and is composed mainly of tridymite, oligoclase and alkali feldspar, with small quantities of glass ($n=1.487-1.492$), quartz, fayalite ($\alpha=1.800$, $2V(-)=56°$), hypersthene ($\gamma=1.720$), biotite ($\gamma=1.608$), osumilite, and iron oxide dust.

TABLE 1. COMPOSITION RANGE OF BIOTITE-BEARING HYPERSTHENE PLAGIOLIPARITE NEAR SAKKABIRA, KYUSYU

SiO_2	72.75–74.65
Al_2O_3	13.67–14.54
Fe_2O_3	0.40– 0.64
FeO	1.43– 1.92
MgO	0.61– 0.67
MnO	0.04
CaO	2.66– 2.77
Na_2O	3.05– 4.23
K_2O	2.16– 2.50
TiO_2	0.13– 0.17
P_2O_5	0.07– 0.26
H_2O+	0.24– 0.47
H_2O-	0.02– 0.39

Osumilite crystallized from the plagioliparite magma at the final stage of consolidation.

PHYSICAL PROPERTIES

Osumilite occurs in crystals, a few millimeters or less in size, black to the unaided eye, and prismatic or tabular in habit. Neither cleavage nor twinning was observed. The specific gravity is 2.64 as determined by suspending the mineral in a mixture of bromoform and acetone.

Under the microscope, osumilite is nearly or completely uniaxial positive. The refractive indices are (for Na):

$$\omega=1.545\text{–}1.547$$
$$\epsilon=1.549\text{–}1.551$$
$$\epsilon-\omega=0.004$$

It shows remarkable dichroism as follows:

$$\epsilon(\|c) = \text{colorless}$$
$$\omega(\perp c) = \text{light blue}$$

UNIT CELL AND SPACE GROUP

Laue, rotation, Weissenberg and powder photographs of osumilite were taken, using CuKα radiation throughout the experiments. The powder data are shown in Table 2.

TABLE 2. *X*-RAY POWDER DATA OF OSUMILITE

Intensities	d(Å)	Indices
VW	8.8	$10\bar{1}0$
S	7.17	0002
W	5.55	$10\bar{1}2$
S	5.08	$11\bar{2}0$
M	4.41	$20\bar{2}0$
S	4.15	$11\bar{2}2$
M	3.74	$20\bar{2}2$
W	3.58	0004
W	3.34	$12\bar{3}0$, $10\bar{1}4$
VS	3.24	$12\bar{3}1$
W	3.02	$12\bar{3}2$
S	2.930	$11\bar{2}4$, $30\bar{3}0$
M	2.776	$20\bar{2}4$
W	2.543	$4\bar{2}\bar{2}0$
W	2.390	0006
W	2.170	
W	2.016	
M	1.855	
W	1.737	
VW	1.462	
W	1.437	
W	1.406	

Laue and Weissenberg photographs show that it is hexagonal in symmetry. The unit cell has the dimensions:

$$a_0 = 10.17\ \text{Å}$$
$$c_0 = 14.34\ \text{Å}$$
$$a_0 : c_0 = 1 : 1.410$$

The reflections ($hh\overline{2h}l$) and ($h0\bar{h}l$) were observed only when l = even. Thus, the space group is $P\ 6/mcc = D_{6h}^2$ or $P\ 6cc = C_{6v}^2$. As will be shown in the next section, the writer succeeded in determining the crystal structure of osumilite by assuming the existence of a center of symmetry. We may then consider that the space group is probably $P\ 6/mcc$.

Chemical Formula and Crystal Structure

A chemical analysis of prismatic crystals of osumilite by Minato is given in Table 3. A simple calculation from the analysis, unit cell dimensions, and specific gravity reveals that the unit cell contains 60 oxygen atoms (except those constituting H_2O molecules). Then, in the third column of the Table, the content of atoms in half a unit cell is shown on the basis of 30 oxygen atoms.

Table 3. Chemical Analysis of Osumilite

	Wt. %	Mol. prop.	Atomic ratios (O=30)				
SiO_2	50.78	8455		9.01		12.00	17.78
Al_2O_3	24.85	2438		5.20	2.99 2.21		
TiO_2	0.01	1		0.00		3.39	
Fe_2O_3	1.95	122		0.26			
FeO	9.97	1388		1.48	0.92 0.56		
MgO	6.68	1657		1.77		2.39	
MnO	0.40	56		0.06			
CaO	0.61	109		0.12		0.79	
Na_2O	1.00	161		0.35			
K_2O	1.42	151		0.32			
H_2O+	1.73	962	H_2O	1.03			
H_2O-	0.27						
	99.67						

Note. BeO is absent according to a spectrographic analysis by H. C. Harrison.

Osumilite has unit cell dimensions similar to milarite in which $a_0 = 10.5$ Å and $c_0 = 13.9$ Å (Belov and Tarkhova, 1949; Ito, Morimoto and Sadanaga, 1952). Osumilite has the same space group and the same number of oxygen atoms in the unit cell as milarite. The chemical formula of osumilite can be written in the form corresponding to that of milarite as follows:

$$\textit{osumilite:}\ (K, Na, Ca)(Mg, Fe^{+2})_2(Al, Fe^{+3}, Fe^{+2})_3(Si, Al)_{12}O_{30}\cdot H_2O$$
$$\textit{milarite:}\ K \quad Ca_2 \quad (Be, Al, Si)_3 \quad (Si, Be)_{12}O_{30}\cdot \tfrac{1}{2}H_2O.$$

Moreover, the rotation and Weissenberg photographs of the two minerals show remarkable similarity, especially in reflections at smaller angles. Therefore, it is considered that osumilite is iso-structural with milarite.

The parameters of atoms in osumilite were determined by the method of trial and error. Intensities of reflections were estimated visually, and converted into numerical values on an arbitrary scale. They are cor-

TABLE 4. COORDINATES OF ATOMS IN OSUMILITE

No. of the equivalent atoms of the unit cell	Atoms	x	y	z
2	K, Na, Ca	0	0	$\frac{1}{4}$
2	H_2O	0	0	0
4	Mg, Fe	$\frac{1}{3}$	$\frac{2}{3}$	$\frac{1}{4}$
6	Al, Fe	$\frac{1}{2}$	0	$\frac{1}{4}$
24	Si, Al	0.10	0.35	0.11
12	O_I	0.13	0.42	0
24	O_{II}	0.21	0.29	0.12
24	O_{III}	0.13	0.47	0.17

rected for the Lorentz and polarization factors, but not for extinction nor absorption. At the final stage of structure analysis, these values were rendered comparable with corresponding calculated values by multiplying by an appropriate number which minimizes the differences between observed and calculated values.

The agreement between observed and calculated structure amplitudes is good with the parameter values shown in Table 4. The atomic scattering factors used are shown in Table 5, and observed and calculated structure amplitudes are shown in Table 6.

The structure of osumilite is based on hexagonal double rings $(Si, Al)_{12}O_{30}$, each of which is composed of 12 (Si,Al)−O tetrahedra, as shown in Fig. 1. The two horizontal reflection planes of the unit cell (0.00 and 0.50 high) pass the central position of each double ring. The plan of the lowest quarter of the unit cell is shown in Fig. 2, which reveals how those double rings are linked by (Al,Fe^{+3},Fe^{+2}) atoms in 4-coordination, by (Mg,Fe^{+2}) atoms in 6-coordination and (K,Na,Ca) atoms in 12-coordination. H_2O molecules are inside the double rings. Probably the water content is not essential to this mineral, and the quantity of (K,Na,Ca) is variable to some extent.

TABLE 5. ATOMIC SCATTERING FACTORS

$\frac{\sin\theta}{\lambda}\times10^{-8}$	0	0.2	0.4	0.6	0.8	1.0
K, Na, Ca	14.0	11.0	7.1	5.1	4.1	3.3
Mg, Fe	13.5	11.2	7.8	5.4	3.9	3.1
Al, Fe	14.6	12.2	8.9	6.4	4.7	3.6
Si, Al	10.0	9.1	7.0	4.9	3.3	2.5
O and H_2O	10.0	5.5	2.7	1.8	1.5	1.3

TABLE 6. COMPARISON OF OBSERVED AND CALCULATED STRUCTURE AMPLITUDES

$hk \cdot l$	F_0	$\lvert F_c \rvert$	$hk \cdot l$	F_0	$\lvert F_c \rvert$
00.2	112	98	22.0	153	185
00.4	195	204	32.0	68	49
00.6	39	78	42.0	96	127
00.8	340	358	52.0	201	164
00.10	80	82	33.0	123	120
00.12	45	67	43.0	41	27
00.14	317	248	53.0	14	5
00.16	169	142			
			10.2	38	64
10.0	9	4	10.4	72	68
20.0	43	40	10.6	9	26
30.0	55	75	10.8	25	35
40.0	9	21	10.10	13	22
50.0	168	168	10.12	32	56
60.0	50	56	20.2	66	112
70.0	129	78	20.4	176	197
80.0	164	124	20.6	0	4
90.0	76	122	20.8	36	29
10,0.0	12	15	20.10	60	116
			30.2	8	11
11.0	64	124	30.4	9	13
21.0	15	58	30.6	67	76
31.0	43	70	30.8	76	79

As is shown in Table 3, the total number of metal atoms, Si, Al, Fe, Mg, and Mn, calculated from the actual analysis on the basis of $O=30$, is 17.78. It is a little larger than that expected from the theoretical formula (17.00). This may be due to the following reasons:

(1) The sample for chemical analysis was not pure, or the analysis was not sufficiently accurate.

(2) Some of the above mentioned metal atoms may occupy some other positions.

(3) Some of the hydrogen atoms which are so far provisionally calculated as forming H_2O molecules, may actually be present as (OH) ions connected with Si atoms. (In milarite only one half of the total available space for water molecules is statistically filled.)

MORPHOLOGY

The crystallographic axes a_1 and a_2 are determined so as to be parallel to horizontal edges of the unit cell. Goniometric data are given in Table 7. Crystals of osumilite are generally a few millimeters or less in size, but rarely reach 5 millimeters in longest dimension. Commonly they are short

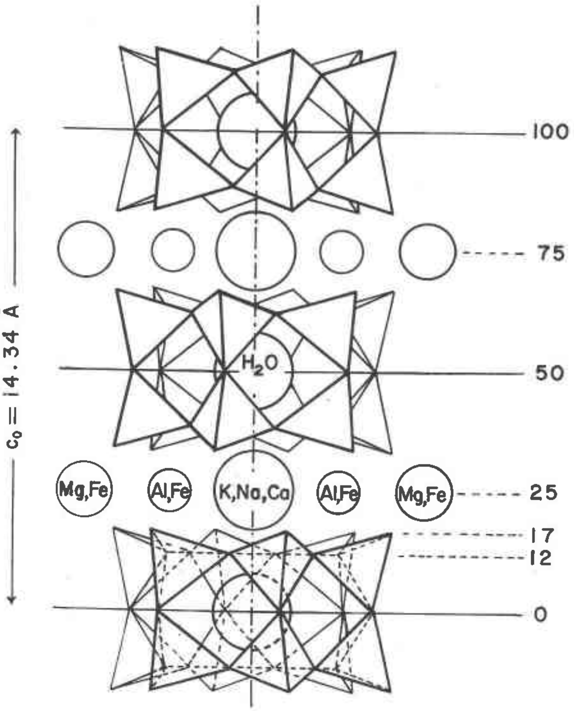

FIG. 1. Hexagonal double rings, projected on $(01\bar{1}0)$. Figures give heights by a percentage of the *c*-translation.

prismatic, being elongated parallel to the *c*-axis, or tabular on $\{0001\}$. Usually they have well-developed faces of forms: $c\{0001\}$, $m\{10\bar{1}0\}$, and $a\{11\bar{2}0\}$. Generally the faces of $a\{11\bar{2}0\}$ are much larger than those of $m\{10\bar{1}0\}$. The faces of $j\{21\bar{3}0\}$ are common, though they are usually very small.

TABLE 7. TWO CIRCLE GONIOMETRIC MEASUREMENTS OF OSUMILITE

Form		Measured		Calculated ($p_0 = 1.634$)		No. of faces	No. of crys-tals	Size	Quality
		ϕ	ρ	ϕ	ρ				
c	0001	—	0° 00′	—	0° 00′	2	2	large	good
m	$10\bar{1}0$	29° 45′–30° 00′	89° 52′–90° 08′	30° 00′	90° 00′	12	2	medium	good
a	$11\bar{2}0$	0° 00′	89° 35′–90° 16′	0° 00′	90° 00′	12	2	large	good
j	$21\bar{3}0$	10° 49′–11° 01′	89° 40′–90° 03′	10° 54′	90° 00′	16	2	very small	fair
p	$10\bar{1}1$	29° 52′–29° 55′	57°–60°	30° 00′	58° 32′	4	1	very small	bad
q	$10\bar{1}2$	29° 52′–30° 00′	38°–39° 15′	30° 00′	39° 15′	7	2	small	poor
e	$11\bar{2}2$	0° 00′	54°	0° 00′	54° 45′	2	1	very small	bad

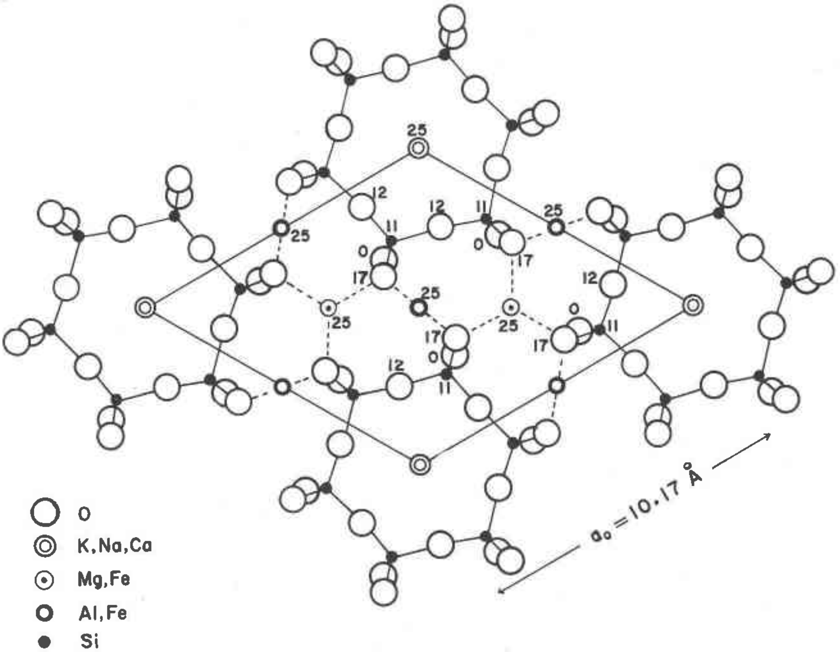

FIG. 2. The lowest quarter of a unit cell of osumilite, projected on (0001). The height of each atom is given by a percentage of the c-translation. (Si, Al) bonds to oxygen are shown as solid lines, and other bonds are shown as broken lines.

Dipyramidal faces $p\{10\bar{1}1\}$ and $e\{11\bar{2}2\}$ are not rare, but frequently very small in size and bad in quality of reflection. Dipyramidal faces $q\{10\bar{1}2\}$ are rare in prismatic crystals but not rare in tabular ones, in which sometimes they are medium in size, but their quality of reflection is not good. Certain faces are frequently composed of a few to many vicinal faces.

As the quality of reflection of the dipyramidal faces is not good, a precise determination of the axial ratio from the goniometric data is difficult. Fortunately a dipyramidal face $(10\bar{1}2)$ showing relatively good reflection was found, and gives the values:

$$\phi = 30°00', \ \rho = 39°15'; \text{ and then } a{:}c = 1{:}1.415.$$

This is in good agreement with the ratio $a_0{:}c_0 = 1{:}1.410$ as determined by an x-ray method.

COMPARISON WITH CORDIERITE

Osumilite is composed of hexagonal double rings $(Si,Al)_{12}O_{30}$, while cordierite is composed of pseudohexagonal single rings $(Si,Al)_6O_{18}$. Therefore, a_0 of osumilite is similar to b_0 of cordierite, while c_0 of osumilite is much longer than that of cordierite (Table 8). Thus, the most reliable method of distinction between osumilite and cordierite is in the measurement of unit cell dimensions. Their *x*-ray powder patterns are also entirely different.

Osumilite and cordierite have similar refractive indices, but the birefringence of osumilite is lower. Osumilite is nearly or completely uniaxial positive, while cordierite is biaxial positive or negative with a large optical angle. Thus the measurement of optical angles is an important way of differentiation.

TABLE 8. COMPARISON OF CORDIERITE WITH OSUMILITE

	Cordierite	*Osumilite*
a_0	17.1 Å	10.17 Å
b_0	9.7 Å	—
c_0	9.3 Å	14.34 Å
Refr. indices	$\begin{cases}\alpha=1.52\text{–}1.56\\ \gamma=1.53\text{–}1.57\end{cases}$	$\omega=1.545\text{–}1.547$ $\epsilon=1.549\text{–}1.551$
Birefringence	0.008–0.016	0.004
2V over γ	Usually 75°–140°	Usually 0°

Osumilite is more deeply colored than, at least, most cordierites. Osumilite is black to the unaided eye and light blue in ordinary thin sections. Even when cordierite is rich in iron, it is usually colorless in ordinary thin sections, as is exemplified by iron-cordierites from Sasago and Dosi (Shibata, 1936), or by artificial iron-cordierites (Richardson and Rigby, 1949; Schairer and Yagi, 1952).

Osumilite and cordierite have different chemical formulas, but when the weight percentages are compared in analytical tables, the difference is very small and might be overlooked. In Table 9 are shown analyses of osumilite and of two cordierites. If we erroneously calculate an analysis of osumilite to the formula of cordierite (that is, on the basis of 18 oxygen atoms), the following discrepancy appears:

cordierite: $\{(Mg, Fe)_2Al_3\}(Si_5Al)O_{18}\cdot n\,H_2O$;
osumilite: $(K, Na, Ca)_{0.6}(Mg, Fe)_{1.2}(Al, Fe, Fe)_{1.8}(Si, Al)_{7.2}O_{18}\cdot 0.6\,H_2O$
$= (K, Na, Ca)_{0.6}\{(Mg, Fe)_{1.2}(Al, Fe, Fe)_{1.8}Al_{1.2}\}(Si, Al)_6O_{18}\cdot 0.6\,H_2O$.

Thus, an analysis of osumilite should give a total number of Al, Fe, Mg, and Mn atoms which do not belong to hexagonal rings, equal to 4.2, that is smaller than 5 in true cordierite. The writer calculated this value from many available actual analyses of cordierites and found it to range from 4.87 to 5.11, while the value from the actual analysis of

TABLE 9. CHEMICAL ANALYSES OF CORDIERITE AND OSUMILITE

	I	II	III	IV	V
SiO_2	49.50	48.19	50.78	48.32	49.15
Al_2O_3	32.45	33.45	24.85	22.13	31.84
TiO_2	0.06	0.01	0.01	n.d.	n.d.
Fe_2O_3	0.40	0.55	1.95	3.88	2.88
FeO	8.90	8.40	9.97	9.58	11.49
MgO	6.73	7.95	6.68	9.10	0.55
MnO	0.11	0.18	0.40	0.68	n.d.
CaO	0.57	0.17	0.61	4.95	4.30
Na_2O	0.46	0.22	1.00	n.d.	n.d.
K_2O	0.39	0.02	1.42	n.d.	n.d.
H_2O+	0.62	0.67	1.73	1.54	0.06
H_2O-	—	0.01	0.27		
	100.19	99.82	99.67	100.18	100.27

I. Cordierite from Laramie Range, Albany County, Wyoming (Newhouse and Hagner, 1949).
II. Cordierite from Great Slave Lake, Canada (Folinsbee, 1941).
III. Osumilite from Sakkabira, Japan.
IV. Ca-rich "cordierite" (or Ca-rich osumilite) from Laacher See, Germany (Gossner and Reindl, 1932).
V. Ca-rich "cordierite" (or Ca-rich osumilite) from Celebes (Bücking, 1900).

osumilite is 4.68. The discrepancy between the theoretical value 4.2 and the actual value 4.68, is due to a slightly too large total number of metal atoms Si, Al, Fe, Mg, and Mn, as stated and discussed previously. Cordierite is variable in alkali and H_2O contents, and that may also be true of osumilite.

OCCURRENCE OF OSUMILITE IN GENERAL

Probably osumilite is a relatively common mineral, but has always been mistaken for cordierite. So far as the writer is aware, osumilite occurs characteristically in volcanic rocks and inclusions in them.

For example, Taneda (1950) described a "cordierite" with small positive optical angles in the groundmass of a hypersthene-rhyodacite from Hamanoiti, Kagosima Prefecture, Kyusyu. It is in anhedral crystals, less than 0.5 millimeter in diameter, pleochroic from light blue to colorless. This is probably osumilite, showing anomalous biaxial character, if the description is accurate. Recently M. Yamasaki and the writer found a uniaxial positive mineral which is probably osumilite from a volcanic rock in Isigami-yama, Kumamoto Prefecture, Kyusyu.

In the literature the writer found two analyses of "cordierites" which may actually represent osumilite. They are "cordierite" from the Laacher See district described by Brauns (1911) and later analyzed by Gossner and Reindl (1932), and "cordierite" from Celebes described by Bücking (1900). They both occur in ejecta from volcanoes.

Braun's described the former as being newly formed in an ejecta, and being nearly uniaxial positive. Bücking described the latter as occurring in six-sided prismatic crystals, pleochroic from colorless ($\|c$) to light bluish purple ($\perp c$). Their compositions, which are shown in Table 9, are abnormal for cordierite as indicated by the describers themselves. They both contain a very large amount of CaO. No other cordierites in the literature show such a large content of CaO. When we calculate the compositions to the formula of cordierite, the total numbers of Al, Fe, Mg, and Mn atoms which do not belong to hexagonal rings, are too small (4.67 and 4.35 respectively) when compared with that for cordierite (4.87–5.11). If we calculate them to the formula of osumilite, however, the Laacher See "cordierite" shows fairly good agreement with the formula, and the Celebes "cordierite" is very close, as shown in Table 10.

TABLE 10. COMPOSITIONS OF CA-RICH OSUMILITES

Locality	I			II		
	Laacher See			Celebes		
O	30.00			30.00		
Si	8.65		12.00	8.55		12.00
Al	4.67	3.35 1.32		6.51	3.45 3.06	3.06
Fe^{+3}	0.52		3.27	0.38		2.20
Fe^{+2}	1.43			1.67		
Mg	2.42		2.52	0.15		
Mn	0.10					
Ca	0.95		0.95	0.80		0.80
H_2O	0.92		0.92	0.03		0.03

These comparisons suggest that actually they may represent Ca-rich osumilites, which have the formula:

$$Ca(Mg, Fe^{+2}, Fe^{+3})_2(Al, Fe^{+3}, Fe^{+2})_3(Si, Al)_{12}O_{30}\cdot nH_2O,$$

where $n=0-1$. Bücking observed a twin-like structure in the Celebes mineral under the microscope. But it may be only apparent, like the similar structure in milarite (Ito, Morimoto, and Sadanaga, 1952).

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POSTSCRIPT

This paper was written in the spring of 1953, when the writer was temporarily at Harvard University. Owing to the loss of the manuscript in transit, its publication has been delayed. (The drawings of osumilite crystals were lost as a result of the accident.) In 1954, the writer and his coworkers in Tokyo University found that so-called "artificial cordierite" formed in synthetic experiments and industrial processes is not true cordierite, but actually a hexagonal polymorph of cordierite composition. As they found this polymorph to occur in a rock in India, they gave it the name "indialite." The "artificial iron-cordierite" synthesized by Schairer and Yagi (1952), which is referred to in this paper, is actually iron-indialite. Refer to "A preliminary note on a new mineral, indialite, polymorphic with cordierite" by A. Miyashiro and T. Iiyama (*Proc. Japan Acad.*, **30,** 746–751, 1954), and also to "The polymorphism of cordierite and indialite" by A. Miyashiro, T. Iiyama, M. Yamasaki, and T. Miyashiro (*Am. Jour. Sci.*, 253, 185–208, 1955).

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