

OCCURRENCE OF CORDIERITE AND ALUMINOUS ORTHOENSTATITE IN THE ALLENDE METEORITE¹

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ABSTRACT

The first meteoritic occurrence of cordierite, in a nodule in the Allende Type III carbonaceous chondrite, is reported. X-ray powder patterns indicate that its structure is very close to that of hexagonal, or high cordierite. Associated minerals are aluminous orthoenstatite, anorthite, spinel, olivine and sodalite. A discussion of the mineral assemblage is presented but it cannot be established whether the inclusion formed as a highly fractionated system within the meteorite or as part of a chemical-pressure environment foreign to that indicated for the meteorite.

INTRODUCTION

The first meteoritic occurrence of cordierite has been observed in a light colored inclusion in the Allende chondrite. This meteorite, which fell near Parral, Chihuahua, Mexico on February 8, 1969, is a Type III carbonaceous chondrite. A preliminary account of its fall and composition was published by King *et al.* (1969) and three papers discussing its mineralogy and chemical composition were presented before the 50th Annual Meeting of the American Geophysical Union (April, 1969) by Marvin *et al.*, Clarke *et al.*, and King. Because of its abundance and unusual features, studies of this meteorite will undoubtedly attract the attention of investigators for many years. Although it is estimated that over one ton of the meteorite may eventually be found, it should be noted that the present work results from the examination of 14 g of material made available to the writer. The meteorite is composed of chondrules embedded in a black fine-grained matrix but of great interest is the presence of white crystalline aggregates randomly distributed in the dark matrix. These white patches are commonly irregular in shape but occasionally are spherical and then resemble chondrules. Those observed in the present study were irregularly shaped and contained mineral associations which are quite different than those normally encountered in chondrules. In this sense, the non-spherical aggregates may be termed inclusions which implies that they represent achondritic material incorporated in a mass which is largely chondritic. An objection arises here in that we know of no achondrites with these mineral associations. For lack of a better term these will be referred to as inclusions. Preliminary examination of four of these showed that they contain minerals high in Ca and Al but the specific minerals and their relative abundances varies from one

¹ Based on work performed under the auspices of the U. S. Atomic Energy Commission.

inclusion to another. One was found to contain coarsely crystalline gehlenite as the main constituent with minor amounts of a new titaniferous pyroxene (average TiO_2 16.0 percent and Al_2O_3 21.0 percent) together with minute (1–10 micron) crystals of perovskite. Another contained spinel, grossular, anorthite, sodalite and trace amounts of unidentified phases. Still another was fine-grained (5–15 micron) and contained spinel, grossular and an unidentified oxide with exceptionally high alumina content up to about 80 percent. It appears that these inclusions are made up of a variety of compositions and each warrants a detailed examination. The common meteoritic silicates, olivine, orthopyroxene, and diopside are conspicuously absent in the above inclusions.

The present paper is a report on one inclusion, 1.0×0.7 mm, rimmed by pentlandite, containing a nodule of pink cordierite about 0.5 mm across, resembling a porphyroblast in metamorphic rocks, but differing in that the surrounding material within the inclusion is not depleted in those elements which are concentrated in the porphyroblast-like interior. The core is predominantly cordierite with inclusions of spinel, anorthite, and aluminous enstatite and the surrounding material also contains finely divided sodalite and a trace of olivine. Hence, this inclusion is relatively low in Ca as compared to those previously mentioned. Grains were pried out of the inclusion, and sorted out under the microscope for X-ray and microprobe studies. All identifications were confirmed by the characteristic powder patterns (film) which in most cases were essentially single-phased but occasionally showed trace amounts of contaminants.

CORDIERITE

Optical properties. Anhedral grains are transparent and colorless in transmitted light but are a light pink when viewed under the binocular microscope. Extinctions are wavy and the negative interference figure shows broad isogyres with some separation indicating a $2V$ of about 20° . Refractive indices (white light) are $\omega = 1.528$, $\epsilon = 1.520, \pm 0.002$. The slight biaxiality and wavy extinction may indicate a strained, normally hexagonal, cordierite.

X-ray data. Powder patterns were taken using a Norelco powder camera, diameter 11.45 cm with Ni-filtered $\text{CuK}\alpha_1\alpha_2$ and V-filtered $\text{CrK}\alpha_1\alpha_2$ radiation. The pattern was indexed on a hexagonal cell with $a_0 = 9.825 \pm 0.009$ Å and $c_0 = 9.325 \pm 0.008$ Å. The data (Table 1) is compared with that for synthetic $\text{Mg}_2\text{Al}_4\text{Si}_5\text{O}_{18}$ (high cordierite) as given on ASTM card 13-293 and by Swanson *et al.* (1962).

The differences between the cell dimensions for Allende cordierite and those of the synthetic high cordierite probably are related to differences

TABLE 1. X-RAY DIFFRACTION DATA FOR CORDIERITE

<i>hkl</i>	Allende meteorite ^a			Synthetic high-cordierite ^b	
	<i>d</i> (calc.) Å	<i>d</i> (obs.) Å	<i>I</i>	<i>d</i> (obs.) Å	<i>I</i>
100	8.51	8.52	100	8.48	100
110	4.91	4.92	24	4.89	32
002	4.662	4.658	17	4.679	15
102	4.089	4.088	50	4.094	51
112	3.382	3.382	47	3.379	57
202	3.143	3.140	62	3.138	66
211	3.040	3.040	69	3.027	86
212	2.647	2.648	27	2.640	26
220	2.456	2.456	9	2.441	6
302	2.423	2.426B	5	2.414	4
004	2.331	2.330	11	2.338	12
311	2.288	2.291	5	2.276	5
213	2.235	2.235	5	2.231	5
222	2.173	2.174	5	2.165	6
114}	2.106	2.106	20	2.108	8
312}				2.098	12
204	2.044	2.041	2	2.046	3
320	1.952}	1.946B	16	1.941	8
402				1.927	6
321	1.911	1.912	4	1.901	3
313	1.880	1.880	16	1.875	15
410	1.857	1.858	5	1.846	6
411	1.821	1.824	6	1.811	7
304	1.801	1.801	11	1.800	9
412	1.725	1.726	1	1.718	3
500	1.702	1.705	7		
224	1.691	1.691	27	1.688	28
314	1.658}	1.657B	6	1.656	3
323				1.647	4
330	1.638	1.638	4	1.629	4
215	1.613}	1.612	7	1.615	3
420				1.599	6
413	1.594	1.594	8	1.589	9
404	1.571	1.569	3	1.569	2
006	1.554}	1.552B	8	1.558	3
332				1.538	3
324	1.497	1.498	7	1.494	8
116	1.482	1.481	9	1.485	5
315	1.463}	1.460B	7	1.463	5
206				1.460	
	$a(\text{Å}) = 9.825 \pm 0.009$			$a(\text{Å}) = 9.770$	
	$c(\text{Å}) = 9.325 \pm 0.008$			$c(\text{Å}) = 9.352$	

^a Norelco powder camera (diam. 11.45 cm), CuK α , α_2 , Ni filter. Diamond used as an internal standard. Abbreviation *B* = broad.

^b Swanson *et al.* (1962).

in composition. Otherwise, line positions and relative intensities are in good agreement. There are, however, a few minor discrepancies in the pattern of the meteoritic mineral. The (402) reflection which should exist at 1.935 Å is only faintly visible but not measurable. The line broadening

of the (302) reflection cannot be accounted for other than by an impurity. The line at 1.705 Å is not reported for the synthetic material but it can be indexed as (500) which is allowed by the space group $P6/mcc$. Except for these provisos, this meteoritic cordierite can be considered to have a structural state close to or equal to high cordierite and in this respect is similar to relatively few terrestrial cordierites from buchites and from a fused shale from the Bokaro coal field in India, which is the only known natural occurrence for the high, or hexagonal, form.

Miyashiro (1957) showed that X-ray powder diffractometer patterns can be used to distinguish the hexagonal (high) from the orthorhombic (low) forms. The main difference is the presence of a single peak for the hexagonal form in distinction to at least 3 or possibly 4 or 5 peaks for the orthorhombic in the region 2θ between 29 and 30°, CuK α radiation. The structural state of cordierite can be characterized by a distortion index based on the separation of three reflections in this region. The index being zero for hexagonal and a maximum of 0.3° for orthorhombic, intermediate values indicate transitional states. The test is less sensitive when applied to film patterns taken on a 11.45 cm powder camera, but in the present study, this was necessary because of the small amount of material available. No line separation could be detected for Cu radiation in the hexagonal (211) reflection at 3.04 Å; a densitometer tracing across the line showed it to be a single sharp peak with a symmetrical profile. For Cr radiation, a slight shadowing of the line appeared on the high-angle side but no line separation could be measured. The sensitivity of the method for Cr radiation is estimated to be about 0.1° in 2θ , hence the structural state of this cordierite is very close to the high form with a distortion index less than 0.1°. The shadow of a doubt prevents this from being identified positively as the second natural occurrence of high cordierite.

Composition. Analyses were performed on an ARL-EMX electron microprobe using a chemically analyzed cordierite of unknown origin, Amelia albite, Asbestos K-feldspar and synthetic diopside as standards. Grains of cordierite, 0.1–0.3 mm, were removed from the inclusion, embedded in epoxy resin, polished and carbon coated; results are included in Table 2. The average structural formula is:



It can be seen that the ratio Al:Si in the rings of (Si,Al)O₄ tetrahedra is close to 1:5 as it is in terrestrial cordierites; furthermore the total number of XY ions in the structure is greater than the ideal 5 per formula unit which is characteristic of those terrestrial cordierites which contain large

TABLE 2. ELECTRON MICROPROBE ANALYSES OF CORDIERITE

Grain Number	SiO ₂	Al ₂ O ₃	MgO	FeO	Na ₂ O	K ₂ O	Total ^a
1	50.3	34.1	10.5	0.5	4.0	0.6	100.2
2	50.4	33.7	11.2	0.4	4.3	0.6	100.8
3	49.8	33.4	10.5	0.4	4.4	0.6	99.3
4	50.9	34.2	10.7	0.5	5.8	0.8	103.1
5	51.1	34.8	12.4	0.1	2.7	0.4	101.7
6	51.0	33.3	9.8	0.6	5.2	0.8	100.9
7	49.8	33.9	9.4	0.7	4.5	0.8	99.3
8	50.9	35.0	12.2	0.1	2.4	0.3	101.1
9	50.7	33.9	10.7	0.3	4.1	0.6	100.5
10	50.1	34.2	9.2	0.6	6.5	0.9	101.7
Average	50.5	34.1	10.7	0.4	4.4	0.6	100.9
Cations per 18 oxygens	4.98	3.97	1.57	0.03	0.84	0.08	11.49 ^b

^a Includes 0.1% CaO and 0.1% Cr₂O₃.

^b Includes 0.01 Ca and 0.01 Cr ions.

Compositions were corrected for deadtime, background, absorption, fluorescence, and atomic number according to Smith (1965).

amounts of alkali ions. The sum of the alkalis exceeds that of any of the analyzed cordierites listed by Deer, Howie and Zussman (1962). This may be a consequence of associated sodalite in the inclusion. It is noteworthy that the alkalis enter the sodalite and cordierite structures in preference to the associated plagioclase, which has a low Na₂O content (1.2 wt.%).

The grain to grain compositions show fairly uniform amounts of Al and Si, but wide fluctuations in Na, K, Mg and Fe. Although there is considerable scatter in the results, plots of these variations reveal some general trends. No correlations are present between either Al or Si with the other cations even though the normal substitution of Mg+Si=2 Al might be expected. Fe replaces Mg, not 1:1 but in a 1:9-10 ratio, about 16 Na ions accompany each Fe, and one K accompanies 10-11 Na ions. Thus each Mg is replaced by a sum of Na+K+Fe roughly equal to 2. Charge balance is thereby maintained but an excess of one alkali ion must be stuffed into open channel positions for each Mg ion replaced. These open positions in terrestrial cordierites have been suggested as sites for H₂O and gas, in addition to alkali ions. Here the effect is shown for grains which formed in the same environment and which have approximately the same bulk compositions.

MINERALS ASSOCIATED WITH CORDIERITE

Aluminous orthopyroxene. The average composition for three orthoenstatite grains in contact with cordierite is in weight percent: SiO₂ 53.4, MgO 35.0, FeO 0.35, CaO 1.04, Al₂O₃ 7.5, Cr₂O₃ 0.71, TiO₂ 1.55, sum = 99.6. Standards used were Bowen enstatite, Hess-1 clinopyroxene, Amelia albite, synthetic rutile and an analyzed chromite. Data were corrected by the same method stated in the footnote to Table 2. The X-ray powder pattern of the analyzed material was that of an ordered orthopyroxene. Alumina contents of 7.9 and 8.2 weight percent were indicated by the separation of two pairs of reflections according to the data relating these variables as reported by Boyd and England (1960). The good agreement with the analyzed Al₂O₃ may not be fortuitous if the line separations in the X-ray patterns are a function of the alumina content and independent of pressure and temperature. This orthoenstatite is unique because most of the pyroxene elsewhere in the meteorite is of the clinoenstatite—clinohypersthene variety. The effect of alumina in stabilizing the enstatite structure relative to protoenstatite was shown by O'Hara and Schairer (1963) for the system diopside—pyrope at one atmosphere pressure. They considered the aluminous orthoenstatite formed below about 1260°C to be stable.

Trace minerals. Minor amounts of spinel, anorthite, olivine and sodalite accompany cordierite and aluminous orthopyroxene. The spinel is essentially the Mg-Al end member but contains 1.1 percent FeO and 2.3 percent Cr₂O₃ (probe), anorthite is An₉₀ (probe) and the olivine is Fo₉₆ (refractive indices). No attempt was made to determine the composition of sodalite because it is too finely divided. It forms brownish aggregates interstitial to cordierite grains.

DISCUSSION

It was previously pointed out that the structural state of the Allende cordierite is close to, but might not be equal to, that of high cordierite. In their study of the system MgO-Al₂O₃-SiO₂, Schreyer and Schairer (1961) conclude that high cordierite is first formed at any temperature and transforms to the low form with further heating, the rate of transformation being related to the temperature and bulk composition or chemical environment. In comparison, the Allende mineral may be a metastable high cordierite that has not been heated to a temperature high enough, or if heated to a sufficiently high temperature was not heated for a time long enough, to convert it to the stable low form. As a rough guide to temperatures and times involved, Schreyer and Schairer

found that high cordierite in the Bokaro fused shale is stable when a specimen of the shale was heated to 1000°C for 19 days, but that transformation to low cordierite occurred after 7 days at 1250°C, all at one atmosphere. Thus it would appear that relatively little information pertaining to the thermal history of the cordierite-bearing inclusion in the meteorite can be gained from the questionable assignment of either state. Further experimental studies involving the composition of the meteoritic mineral may be more meaningful.

The alumina content of the enstatite is several times that of other meteoritic enstatites but only sketchy comparisons can be made because of lack of data. Keil (1968) reported a maximum amount of 0.7 percent Al_2O_3 in either clino or orthoenstatites from 13 enstatite chondrites examined. This amount may also be an upper limit for other chondrites which may have formed in low pressure, low alumina environments. Yet low aluminous pyroxenes are also characteristic of the high alumina-containing pyroxene-plagioclase achondrites for which there is data. Duke and Silver (1967) report about 0.2 percent Al_2O_3 in the pigeonites and orthopyroxenes in Nuevo Laredo, Sioux County, and Juvinas which have about 12 percent total alumina. Thus the high alumina content of the orthoenstatite associated with cordierite in this particular inclusion in Allende could be a consequence of conditions other than the presence of a high aluminous environment.

Aluminous enstatites may indicate high pressures and temperatures as shown by Boyd and England (1964) for the system $\text{MgSiO}_3\text{-Mg}_3\text{Al}_2(\text{SiO}_4)_3$ (pyrope). Their experimental data however refers to pressures and temperatures greater than 20 kilobars and 1100°C. An enstatite with 7.5 weight percent Al_2O_3 for example could form below the boundary of the miscibility gap which at 1100°C occurs at 25 kilobars where pyrope appears. On the other hand, a general comparison with terrestrial aluminous orthoenstatites is of interest; the pertinent information is summarized by Boyd and England (1960). Volcanic and plutonic igneous rocks formed at moderate depths contain enstatites with from 0.5–3.5 weight percent Al_2O_3 . Those from ultramafic inclusions in basalt range from 2.0–5.5 weight percent, and those from granulites may contain up to 9.5 weight percent Al_2O_3 . Evidently some granulites formed at pressures where cordierite breaks down to enstatite, sillimanite and quartz. Hence, the association of cordierite with aluminous enstatite in Allende suggests formation at pressures comparable to those under which some of the lower grade granulites formed. Pressures in this range however would appear to be too high for this meteorite as revealed by the presence of voids and even hollow chondrules. (The presence of the latter is especially

interesting in view of present beliefs that chondrules were accumulated in the meteorite body through violent collision processes). If the inclusion did not form in a higher pressure environment than that indicated for the whole meteorite, then it must be postulated that the minerals in the inclusion formed in a relatively lower pressure environment. Some support for this possibility is found in the system, diopside—pyrope studied at one atmosphere pressure by O'Hara and Schairer (1963). They found that mixtures containing 70–84 weight percent pyrope below 1260°C yielded a stable assemblage of anorthite, forsterite, orthoenstatite, and cordierite. The orthoenstatite formed in this region contained up to 6 weight percent Al_2O_3 . The adjoining field from 84–100 weight percent pyrope contained the assemblage spinel, forsterite, anorthite and cordierite. The assemblage in the inclusion (excluding the small amount of sodalite present), under equilibrium conditions, is represented at the boundary composition 84 percent pyrope which separates the two fields. There are a few inconsistencies in the above picture. It is unlikely that the enstatite, when accompanied by spinel, could contain the maximum amount of alumina as some should go into spinel. Furthermore, the composition 84 percent pyrope would contain about 30 volume percent forsterite which is inconsistent with the trace amounts observed.

It is apparent that it is difficult to decide whether the mineral assemblage in this inclusion formed at pressures higher than those consistent for the whole meteorite or at lower pressures. Some of the difficulty is related to the possibility that the assemblage formed under nonequilibrium conditions as evidenced by the absence of crystal faces and the variation in the composition of the individual cordierite grains. The system is further complicated by the presence of alkalis which have not been considered in relating the assemblage to experimental systems.

Meteorites contain small amounts of several osumilite-type minerals but until now, closely related cordierite has not been found. Merrihueite was discovered by Dodd, Van Schmus, and Marvin (1965) in the Mezo-Madaras chondrite. Roedderite, the Na-Mg analogue of merrihueite, was discovered in the Indarch enstatite chondrite by Fuchs, Frondel and Klein (1966) and was subsequently reported in the Wichita County iron by Olsen (1967) and in the Canyon Diablo iron by Olsen and Fuchs (1968). Yagiite, the Na-Mg analogue of osumilite, was discovered in the Colomera iron by Bunch and Fuchs (1969). It is not clear why cordierite should form in Allende instead of the closely related alumina-containing minerals osumilite or yagiite. Since both osumilite and yagiite are more siliceous than cordierite and also occur with quartz or tridymite, perhaps the formation of cordierite reflects the higher alumina and lower silica environment in this inclusion.

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