

Variation of refractive index of synthetic Mg-cordierite with H₂O content

OLAF MEDENBACH, WALTER V. MARESCH, PETER W. MIRWALD AND WERNER SCHREYER

*Institut für Mineralogie, Ruhr-Universität
4630 Bochum, F.R. Germany*

Abstract

The mean refractive index of grain mounts has been measured for 50 synthetic end-member Mg-cordierites with water contents varying from 0.83 to 2.90 weight percent. A linear relationship appears adequate to describe the variation of the mean refractive index with water content and can be formulated as

$$W = [n(m) - n(m_0)]/0.00762 \text{ (esd} = 0.13 \text{ weight percent H}_2\text{O)}$$

where W = H₂O content in weight percent; $n(m)$ = mean refractive index of hydrated Mg-cordierite; and $n(m_0)$ = mean refractive index of the anhydrous starting material. The calibration curve is useful to determine water contents of Mg-cordierites in multiphase reaction products of the appropriate experimental systems. No change in the distortion index upon hydration of anhydrous Mg-cordierite has been observed, so the Stout model of cordierite hydration cannot be applicable to end-member Mg-cordierite.

Introduction

Pure Mg-cordierite, Mg₂Al₄Si₅O₁₈ · nH₂O, is an essential and common phase in the synthetic system MgO–Al₂O₃–SiO₂–H₂O, which is an important model, or critical subsystem of models, for most pelitic rocks (e.g. Schreyer, 1976). As a result, Mg-cordierite plays an important part in many experimental and theoretical studies directed toward the investigation of progressive metamorphism in pelitic sediments, so that a detailed knowledge of the stability relations of this mineral is critical.

From studies of natural cordierites (Leake, 1960; Iiyama, 1960; Schreyer, 1965), as well as experimental investigations of the system Mg-cordierite–H₂O (Schreyer and Yoder, 1964; Mirwald and Schreyer, 1977), cordierite is known to incorporate significant and varying amounts of water. Most investigators concur that this water is held in the molecular state in the open channels of this framework structure (Smith and Schreyer, 1962; Schreyer and Yoder, 1964; Farrell and Newnham, 1967; Goldman *et al.*, 1977), but the question of whether or not hydrogen-bonding to the silicate framework occurs is still debated (Stout, 1975, 1976; Langer and Schreyer, 1976; Schreyer *et al.*, 1979).

Variable water content has a significant effect on

the stability relations of cordierite (Newton, 1972; Mirwald and Schreyer, 1977). In order to assess these relations both experimentally and thermodynamically, the equilibrium water content should be known for a given temperature and pressure. As discussed by Schreyer and Yoder (1964) and Newton (1972), however, the direct determination of equilibrium H₂O in quenched Mg-cordierite samples meets with serious problems, because of the extremely rapid exchange of water between the cordierite channels and the aqueous vapor phase during quenching.

During our investigations on the hydration behavior of Mg-cordierite (Mirwald *et al.*, 1979), we have found that with a quench of 10 seconds or less it is possible to conserve the equilibrium water content of Mg-cordierite for experiments below 600°–900°C, depending upon the pressure involved (Fig. 1). Such quench rates are possible in cold-seal autoclaves with rapid-quench attachments (Wellmann, 1970) and in piston-cylinder-type apparatus, both of which are in common use. Our purpose is to refine the calibration of mean refractive index *vs.* water content given by Schreyer and Yoder (1964), and thus provide a basis for routinely measuring equilibrium water contents of Mg-cordierite even in multiphase reaction products. The above relationship also forms a basic part

of the scheme developed by Lepezin *et al.* (1976) for determining the H_2O contents of natural iron-bearing cordierites from their optical properties.

In this study, we restrict ourselves to cordierites with a maximum water content of 3 weight percent. The resulting $P(H_2O)$ - T range in which such a calibration curve may be meaningfully applied is given in Figure 1.

Experimental method

Starting material

Cordierite from two separate batches, synthesized in different ways, was used in the experiments reported here (Table 1). Cordierite I was prepared from a 10 g oxide mix containing ground, purified Brazilian quartz, reagent grade MgO fired at 1200°C prior to weighing, and γ - Al_2O_3 prepared from reagent grade Al-metal. The amount of γ - Al_2O_3 was corrected for a 5.43 wt% H_2O content determined gravimetrically prior to weighing. The oxide mix was repeatedly melted at 1560°C in a Pt-resistance furnace and intermittently ground in several cycles until a homogeneous glass was obtained. The glass was then tempered at 950° (1 hour), 1000° (72 hours), and 1400°C (168 hours). Cordierite II is equivalent to "Cordierite A" of Johannes and Schreyer (1979) and has been described in detail by them. It was ob-

Table 1. Chemical, optical, and structural characteristics of starting material

	Cord. I	Cord. II	Theoretical
Comp. (wt%):			
SiO ₂	51.6	52.1	51.36
Al ₂ O ₃	34.1	34.2	34.86
MgO	14.4	13.5	13.78
CaO	.05		
Na ₂ O	.055	.05	
K ₂ O	.0055		
BeO		n.d. [†]	
Total	100.21	99.85	100.00
Numbers of Cations on the Basis of 18(O)			
Si	5.02	5.07	5
Al	3.91	3.93	4
Mg	2.09	1.96	2
Ca	.005		
Na	.01	.009	
Total	11.04	10.97	11
$n(m_o)$	1.5238 ^{††}	1.5250 ^{††}	1.5226 ^{†††}
	.20°	.19°	
[†] not detected ^{††} estimated error ± .0005 ^{†††} Gladstone-Dale relationship after Larsen and Berman (1934)			

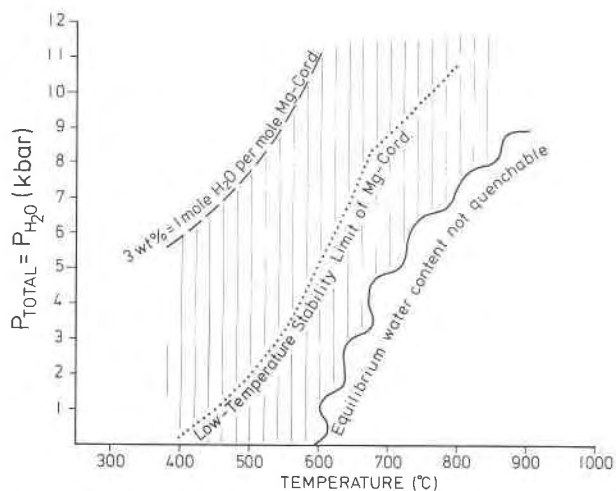


Fig. 1. Approximate range of $P(H_2O)$ - T conditions where the calibration curve developed in this study may be applied to determine equilibrium water contents, provided that the quench interval does not exceed 10 seconds. Dotted line: low-temperature stability limit of Mg-cordierite after Seifert and Schreyer (1970), Schreyer (1970), and Mirwald and Schreyer (1977). Other data from Mirwald *et al.* (1979).

tained by crystallization of a gel prepared according to the method of Hamilton and Henderson (1968).

Both cordierite starting materials have been chemically analyzed (Table 1), using microscale solution photometry for Si and atomic absorption spectrophotometry for all other elements. Slight differences in composition between cordierite I and II as well as minor deviations of both from the theoretical composition are within the limits of analytical error and attainable accuracy with the synthesis method employed. Trace mullite (?) was observed in cordierite II, while cordierite I was free from impurities.

The material subjected to hydration was in the form of irregular fragments not exceeding 50 μm in longest dimension. Only fragments less than 20–25 μm were, in general, "single-crystal" grains, although undulose wavy extinction was characteristic of even the smallest particles. No significant changes in these characteristics were observed during the hydration runs.

Experimental apparatus and techniques

For runs at and below 5 kbar, 25 cm Tuttle-type, cold-seal autoclaves (38 mm outer diameter; 8 mm bore) with rapid-quench attachments (Wellmann, 1970) were employed with water as the pressure medium. According to Rudert *et al.* (1976), the temperature gradients in this type of apparatus are potentially quite large and can lead to inhomogeneous hydration of the cordierite sample. However, we used short capsules of 20 mm length and crimped them so that the sample was restricted to the lower one-third of the capsule. Multiple water analyses were carried out on several samples, but no detectable inhomogeneity in H₂O content was found. Runs above 5 kbar were carried out in a piston-cylinder apparatus employing the salt-cell assembly of Mirwald *et al.* (1975). Temperature gradients are less than 10–15°C in the sample region.

Starting material of 70 to 100 mg of anhydrous cordierite was sealed into gold capsules along with excess doubly-distilled H₂O, usually 10–15 percent by weight. The seal of each capsule was checked by drying and weighing before and after each run. Run products were routinely examined both microscopically in grain mounts and *via* X-ray powder diffraction analysis in order to monitor any growth of contaminating phases. The distortion index, Δ , was measured on diffractometer charts obtained using CuK α radiation, 1/8°/min scanning speed, 1 cm/min chart speed, and a 1°/0.1 mm/1° slit system.

Experiments varying in run duration from 2 to 24 hours and yielding identical results indicate that equilibrium hydration for most pressures and temperatures is reached in less than 2 hours in the cold-seal apparatus. However, in order to ensure that thermal equilibrium is reached in these rather large inhomogeneous aggregates, run durations of 24 hours were chosen. In the piston-cylinder apparatus, run times varied between 0.5 and 4 hours, depending upon the *P-T* conditions, because of the necessity of ensuring equilibrium hydration without incurring the growth of breakdown assemblages outside the Mg-cordierite stability field (Fig. 1).

H₂O analysis

In general, 40–60 mg of hydrated cordierite were used for the analytical determination of the water content. Water analyses were carried out using a coulometric method modified after Lindner and Rudert (1969). This procedure has been detailed by Johannes and Schreyer (1979), who report a relative standard deviation of ± 3 percent for each analysis.

Determination of the mean index of refraction

The problem was to find an accurate and rapid method of determining the refractive index, suitable for processing large numbers of fine-grained powdered samples. As indicated above, "single-crystal" fragments of hydrated cordierite did not exceed 20–25 μ m in size. In fact, samples which had equilibrated outside the stability field of Mg-cordierite (Fig. 1) often had to be reground, because the presence of slight surface alteration of the cordierite fragments (to breakdown assemblages such as kyanite-chlorite-quartz or kyanite-talc-quartz) proved to be normally unavoidable. In our samples this zone of surface breakdown was generally so thin that it was evident only in phase contrast and is of no significance as far as the bulk water analysis is concerned. As a result of the grain size, grain mounts rather than single-crystal, spindle-stage methods were necessary.

The λ -variation method with a 450 W xenon source and prism monochromator, along with phase contrast (*e.g.* Piller, 1952) was selected. Phase contrast increases the accuracy of finding a match between cordierite and immersion medium by an order of magnitude relative to the Becke line approach.

For the immersion medium a large optical dispersion is desirable. We employed mixtures of PALATINOL M (BASF, Ludwigshafen, F. R. G., $n_D^{20} = 1.5146$), which is a fluid, and SANTOLITE MHP (Monsanto Chemical Co., St. Louis, Mo., $n_D^{20} = 1.5849$), which is a resin. Homogenization was effected through gentle warming. The advantages of these dispersion media, beyond their large optical dispersion, are the thermoplastic properties for mixtures with refractive indices close to 1.6, allowing semi-permanent grain mounts to be made, and their very low vapor pressure. PALATINOL/SANTOLITE mixtures were remeasured after 3 weeks of storage in open containers, and after 10 months in closed bottles. No deviation in the refractive index could be discerned in either case. The refractive index of these mixtures was determined by the method of minimum deviation, using a prism constructed according to Larsen and Berman (1934), and a Hg-Cd lamp. The results for PALATINOL M and SANTOLITE MHP are given in Table 2.

PALATINOL/SANTOLITE mixtures were so chosen that at least three "matches" between cordierite and immersion medium could be obtained in the visible region. An accuracy of ± 3 nm at 550 nm was attained. n_D was interpolated from the cordierite dispersion curve constructed on a Hartmann dispersion

Table 2. Dispersion of natural cordierite and immersion medium components. Wavelengths used are the strongest lines of the Hg-Cd spectrum

nm	1 n	2 n	3 n_Y
643.8	1.5109	1.5805	1.5443
576.9	1.5156	1.5859	1.5472
546	1.5185	1.5894	1.5490
508.6	1.5230	1.5945	1.5515
480.0	1.5272	1.5993	1.5540
467.8	1.5293	1.6016	1.5550
435.8	1.5359	1.6092	1.5585
405	1.5444	1.6191	1.5631

1. Palatinol M
2. Santolite MHP
3. Gem quality natural Fe-bearing cordierite

net. Bloss (1978) has pointed out that such empirically determined dispersion curves may actually be steeper than the true curves, due to the selective sensitivity of the human eye. We have therefore measured the dispersion curve for n_Y , using a prism of natural iron-bearing cordierite (Table 2) and the method of minimum deviation noted above. A number of comparisons have shown that the dispersion curves of synthetic Mg-cordierite and the Fe-bearing natural cordierite are exactly parallel. This slope constraint was a useful additional constraint during interpolation of n_D .

All measurements were carried out at an ambient temperature of 20–21°C. Under these conditions and with the procedure outlined above, absolute accuracies of ± 0.0005 are attainable. The uncertainty includes errors in optical matching, graphical construction, and the slight fluctuations of temperature between 20 and 21°C.

Because the cordierite under investigation is anisotropic, we originally made a survey of 50–100 randomly oriented grains for each cordierite product in each of several mounts with a different immersion medium, in order to determine the absolute minimum and maximum index of refraction of the cordierite. Thus an $n(\text{average})$ may be calculated from $[n(\text{min.}) + n(\text{max.})]/2$. During these measurements, especially in fine-grained dense mounts, a marked "contrast reversal" in phase contrast was noted at a specific, critical wavelength. The index of refraction corresponding to this critical wavelength was always found to be identical with $n(\text{average})$. Subsequently, we employed this wavelength of critical contrast re-

versal as a convenient and rapidly obtainable measure of the mean index of refraction, as defined by $\sqrt[3]{n_\alpha \cdot n_\beta \cdot n_\gamma}$.

The calibration curve

The results of optical and chemical determination are plotted in Figure 2. Here the reason for the necessity of using two different cordierites, apart from the fact that only a limited number of analyses is possible from a given cordierite batch, may be explained. Hydration experiments were originally carried out using cordierite I and cold-seal, rapid-quench autoclaves within the stability field of hydrous Mg-cordierite. The work of Mirwald and Schreyer (1977) on the metastable extension of the reaction $\text{Mg-cordierite} \cdot n\text{H}_2\text{O} = \text{talc} + \text{kyanite} + \text{quartz} \pm \text{H}_2\text{O}$ showed that the breakdown of cordierite I outside its stability field is so rapid that water analyses of the multiphase product are meaningless. Since the maximum water content attainable within the hydrous Mg-cordierite stability field is approximately 2.4 wt%, the extension of the calibration curve to 3.0wt% was possible only with cordierite II, which, for reasons not yet satisfactorily explained, proved to be far less reactive.

A linear least-squares fit has been calculated for the two data populations of Figure 2, which corre-

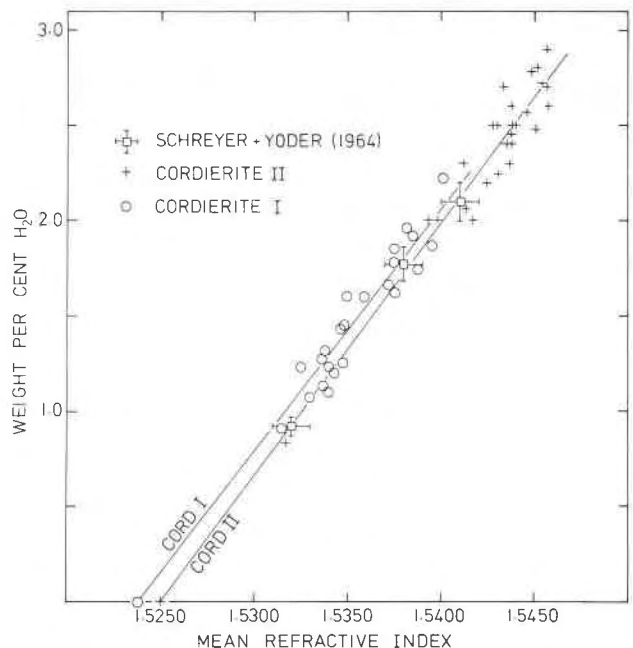


Fig. 2. Results of optical and chemical analysis of 50 hydrated Mg-cordierites. Straight lines are linear regression analyses on the cordierite I and II data populations.

spond to cordierite I and II. The two fits are essentially parallel, indicating that the effect of water content on the mean refractive index is independent of the slight differences in starting material. Three pairs of values reported by Schreyer and Yoder (1964) are also shown and are in good agreement with our present data.

Because of the basically similar response to hydration of both cordierites, as far as the mean index of refraction is concerned, we have combined the two data sets by correcting for the difference in $n(m_0)$ of the two starting materials. Cordierite I was chosen as the basis, because $n(m_0)$ of cordierite I corresponds more closely to the theoretical value calculated from the Gladstone–Dale relationship (1.5226). The result is shown in Figure 3. For the population of 50 data points, the following regression equation may be calculated:

$$\text{wt\% H}_2\text{O} = (131.234) [n(m)] - (199.975)$$

(correlation coefficient $r = +0.932$; $esd = 0.13$ weight percent H_2O).

We have also attempted to fit the data with a third-degree polynomial (dashed line in Fig. 3). Statistically speaking, the quality of the fit is improved only slightly, and in consideration of the analytical errors discussed in the previous section, we feel that a linear fit is adequate.

From the above equations, we may derive the following general expression:

$$W = [n(m) - n(m_0)]/0.00762$$

where $W = \text{H}_2\text{O}$ content in weight percent, $n(m) =$ mean refractive index of hydrated Mg-cordierite, and $n(m_0) =$ mean refractive index of the anhydrous starting material.

Discussion

The calibration curve is meant to be an aid in determining the water content of Mg-cordierite in multiphase reaction products. Ideally, $n(m_0)$ should be known. If cordierite is newly formed in a reaction, this value may be obtained by dehydrating the product cordierite through heating in air. Figure 1 shows those P – T conditions where our experiments indicate that a quench of 10 seconds or less will make a water determination meaningful, because the equilibrium water content during the experiment will be conserved.

The factors that appear to be the main reason for differences in $n(m_0)$ of the analyzed Mg-cordierites are small differences in composition (e.g. Table 1)

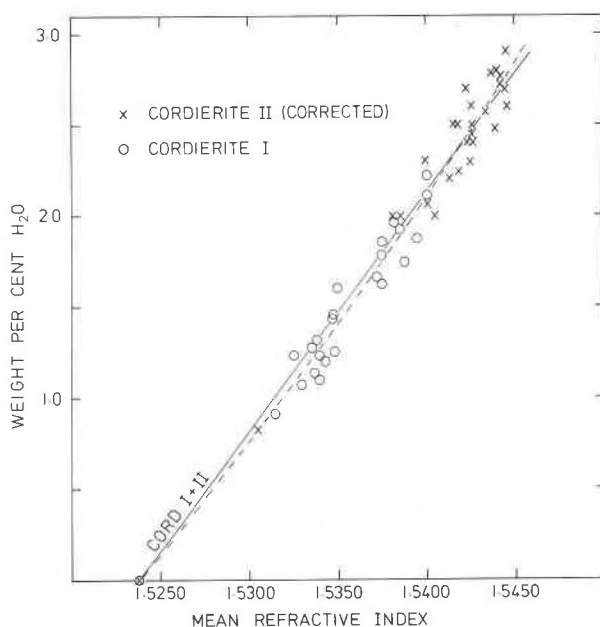


Fig. 3. Combined results for cordierite I and II as discussed in text. Straight line: linear regression analysis and proposed calibration curve for combined data; dashed curve: regression analysis using a third-order polynomial.

and the structural state, as indicated by Δ , the distortion index (Miyashiro, 1957; Langer and Schreyer, 1969). Schreyer and Schairer (1961) have reported that the difference in mean refractive index between “low” ($\Delta > 0.20^\circ$) and high ($\Delta = 0.0^\circ$) Mg-cordierite is 0.003. Using the procedure outlined in this paper, we have obtained the following refractive indices of a synthetic, anhydrous Mg-cordierite in two different structural states:

Δ	$n(m_0)$
0.0°	1.5265 ± 0.0005
0.21°	1.5244 ± 0.0005

This result confirms the finding of Schreyer and Schairer (1961), also shown graphically by Lepezin *et al.* (1976, Fig. 5), and suggests that $n(m_0)$ is decreased by 0.0001 for each increase in Δ of 0.01° .

In order to ensure that $n(m_0)$ does not change appreciably during the run, we have monitored both $n(m_0)$ and Δ before and after the experiment for a number of runs. The results are summarized in Table 3 and confirm that no significant variation in either parameter has occurred. Evidently the change in mean refractive index upon hydration of synthetic Mg-cordierite is due solely to the additional “refractive energy” of molecular H_2O in the cordierite channels. These observations are significant, because

Table 3. Comparison of the mean index of refraction, $n(m_0)$, for anhydrous Mg-cordierite and Δ , the distortion index, before and after the run for several cordierites with varying water contents

Run No.	Starting Material	wt% H ₂ O	Difference [†]	
			$n(m_0)^{††}$	Δ
Initial values	Cord. I	.00	1.5238 ^{†††}	.20 ^{0††††}
	Cord. II	.00	1.5250 ^{†††}	.19 ^{0††††}
PWM 1	Cord. II	.83	—	-.03 ⁰
RQV 327	Cord. I	1.23	—	-.01 ⁰
RQV 282	Cord. I	1.25	—	-.01 ⁰
RQV 263	Cord. I	1.60	+ .0003	+ .02 ⁰
RQV 261	Cord. I	1.85	—	+ .02 ⁰
PWM 9	Cord. II	2.06	—	-.02 ⁰
RQV 235	Cord. I	2.22	—	.00 ⁰
PWM 15	Cord. II	2.40	+ .0005	—
PWM 6	Cord. II	2.60	—	-.01 ⁰
PWM 43	Cord. II	2.86	—	-.02 ⁰

† Starting Material minus Product

†† determined on material used in water analysis

††† estimated error $\pm .0005$

†††† estimated error $\pm .03^0$

Stout (1975, 1976) has formulated a theoretical crystal-chemical model, contested by Langer and Schreyer (1976), which suggests that bonding between H₂O in the cordierite channels and the Si/Al tetrahedra of the surrounding rings results in a distortion of these rings such that the distortion index Δ is decreased significantly. The present results indicate that the effects indicated by the Stout model must be negligible, as far as pure end-member Mg-cordierite is concerned. It is possible, nonetheless, that the Stout model may be applicable when further solid solution components are introduced into end-member Mg-cordierite (Schreyer *et al.*, 1979).

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