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A note on the miscibility of magnesia-cordierite and beryl

IN a study of the highly beryllian cordierite from Vežňá, Czechoslovakia, Černý and Povondra (1966) pointed out that the beryllium content of pegmatitic cordierites might be due to a substitution $\text{Na}^+ + \text{Be}^{2+} \rightleftharpoons \text{Al}^{3+}$ in the cordierite structure. On the other hand, formation of cordierite mixed crystals in the system magnesia-cordierite-beryl, i.e. a substitution $2\text{Mg}^{2+} + 2\text{Al}^{3+} \rightleftharpoons 3\text{Be}^{2+} + \text{Si}^{4+}$, was invoked for an explanation of the beryllium content in pegmatitic cordierites from Haddam, Bjordan (probably a misspelling of Bjørdammen), and Micanite in a 1966 paper by Newton. This author succeeded in synthesizing a homogeneous high-cordierite mixed crystal at 1300 °C and 1 atm containing 15 wt % beryl. On the basis of his experiments he concluded that high-cordierite takes in 21.0 mole % beryl at this same temperature. Similarly he deduced a cordierite miscibility of about 6 to 7 mole % beryl at 700 °C and 1 kb. For a proper evaluation of the results of an experimental investigation of the substitution $\text{Na}^+ + \text{Be}^{2+} \rightleftharpoons \text{Al}^{3+}$ in magnesia-cordierite by Povondra and Langer (1971) it seemed necessary to reinvestigate some compositions in the sodium-free system along the join $\text{Mg}_2\text{Al}_4\text{Si}_5\text{O}_{18}$ - $\text{Al}_2\text{Be}_3\text{Si}_6\text{O}_{18}$.

Finely powdered, homogeneous glasses with 3, 6, and 10 mole % of the beryl component (SG-3, SG-2, and SG-1 resp.) and a fourth with 10 wt % beryl (11 mole %; glass no. 1) served as starting materials. The first three glasses were prepared at 1540 °C from gels, the theoretical and analysed composition of which (excluding BeO) are compiled in table I, while the fourth had been prepared from the oxides at the same temperature.

Table II gives the results of hydrothermal crystallization of glasses SG-3, SG-2, and SG-1 and the lattice constants of the hexagonal cordierites formed. The phases obtained were identified from their X-ray powder diagrams and optical properties. Lattice constants were determined from powder patterns using NaCl as an internal standard, processing the data by a least-squares program (Burnham, 1962). While at 3 mole % beryl homogenous cordierite is produced, at 6 mole % beryl appears as an additional phase in the X-ray pattern. Furthermore, when proceeding towards higher beryl contents no significant changes of lattice constants of the cordierite phase could be detected. From these observations we conclude that the range of solid solubility on the join magnesia-cordierite-beryl does not exceed 4 mole % beryl at 3 kb and 750 °C, i.e. a composition $(\text{Mg}_{1.92}\text{Al}_{0.08})^{[6]}(\text{Al}_{3.84}\text{Be}_{0.12}\text{Si}_{5.04})^{[4]}\text{O}_{18}$. On this basis it seems improbable that the beryllium content of the Micanite cordierite can be explained in terms of a cordierite-beryl solid solution alone, since the observed BeO-content of the mineral would yield a concentration of 5.7 mole % beryl (Newton 1966). Unfortunately no analytical data including Na_2O are published so far on the

Micanite cordierite, and therefore it is impossible to decide as to whether or not a substitution $\text{Na}^+ + \text{Be}^{2+} \rightleftharpoons \text{Al}^{3+}$ has to be taken into account for explaining the BeO contents of this mineral. On the other hand, the Haddam cordierite, the beryllium content (0.52 wt % BeO) of which would yield a concentration of 4.3 mole % beryl (Newton, 1966), contains also an appreciable amount of sodium (0.85 % Na_2O ;

TABLE I. *Composition of gels on the join magnesia-cordierite-beryl (analytical data have been recalculated for H_2O -free substance)*

Gel No.	Mole % beryl	MgO (wt %)		Al_2O_3 (wt %)		SiO_2 (wt %)		H_2O content (wt %)
		calc.	detd.	calc.	detd.	calc.	detd.	
G-3	3	13.40	13.85	34.40	34.46	51.79	51.82	2.50
G-2	6	13.02	13.24	33.97	33.50	52.24	52.54	2.23
G-1	10	12.60	12.81	33.38	32.90	52.82	52.89	2.50

TABLE II. *Products of hydrothermal crystallization of glasses fused from gels compiled in table I; run conditions 3 kb, 750 °C, 220 hrs*

No. of starting material	Mole % beryl	Phases obtained		Lattice constants of cordierite	
		cordierite	beryl	<i>a</i>	<i>c</i>
SG-3	3	vst	—	$9.784 \pm 0.003 \text{ \AA}$	$9.341 \pm 0.003 \text{ \AA}$
SG-2	6	vst	vw	9.777 ± 0.004	9.337 ± 0.004
SG-1	10	st	w	9.780 ± 0.004	9.338 ± 0.004

Iiyama, 1956, no. 12). Recalculation of the structural formula of this cordierite including beryllium leads to almost identical values for Be^{2+} and Na^+ thus suggesting the substitution $\text{Na}^+ + \text{Be}^{2+} \rightleftharpoons \text{Al}^{3+}$ rather than a miscibility of cordierite towards beryl as an explanation of the BeO content in the Haddam cordierite.

In table III the results of some experiments with glass no. 1 (10 wt % beryl) are compiled. The phases obtained and their estimated proportions in the products are given along with temperatures of annealing, run durations, and angles 2θ Cu- $K\alpha$ of the 22 $\bar{4}$ 4 X-ray peak of hexagonal cordierite, which had been found to be very sensitive to composition changes during the study mentioned above (Povondra and Langer, 1971). These values were measured using the 222 interference of NaCl at 56.53° 2θ Cu- $K\alpha$ as internal standard. The first crystalline phase formed by devitrification of the 10 wt %-beryl glass is a stuffed high-quartz derivative similar to that forming through crystallization of pure magnesia-cordierite glass (Schreyer and Schairer, 1961a; Langer and Schreyer, 1969). This stuffed high-quartz transforms into a homogeneous cordierite containing 10 wt % beryl. However, this cordierite unmixes after continued heating at higher temperatures to yield beryl and a beryl-poorer cordierite. The cordierite with 10 wt % beryl is, therefore, a metastable product. This process of unmixing is also indicated by changes in the position of the critical cordierite peak.

Judging from these changes the equilibrium composition of the cordierite mix-crystal is attained approximately after 144 hrs/1200 °C. This cordierite_{ss} apparently has a composition very close to that obtained by hydrothermal crystallization at 3 kb, 750 °C, 72 hrs. If we assume a linear relationship between peak position and composition of cordierite_{ss} and use the peak positions of the homogeneous but metastable 10 wt % sample (54.56°) and that of pure high-cordierite (54.29°; Schreyer and Schairer, 1961b), again a concentration of about 4 mole % beryl is estimated for the stable cordierite_{ss} in question.

TABLE III. *Development of crystalline phases during annealing a glass with 10 wt % beryl at 1 atm (glass no. 1; 11 mole %). Heating at a higher temperature (cf. first column) followed the run of maximum duration at the next lower temperature. For comparison the result of hydrothermal crystallization at 3 kb, 750 °C, 72 hrs is also given*

Temp.	Duration	Phases obtained			Position of critical cordierite peak (Cu-K α)
		stuffed high-quartz	cordierite	beryl	
950 °C	0.5 hr	st	m	—	—
	1	m	st	—	—
	2	vw	vst	—	—
	24	—	vst	—	54.56°
980	24	—	vst	—	—
1160	24	—	vst	vw	54.45
1200	24	—	vst	vw	54.49
	72	—	vst	vw	54.41
	144	—	vst	vw	54.41
1230	104	—	st	w	54.39
1260	88	—	st	w	54.39
1350	120	—	st	w	54.40
Hydrothermal crystallization	—	—	st	w	54.39

Despite the fact that the conclusions given have yet to be confirmed by a detailed study it is quite evident that the extent of the stable solid solubility on the join magnesia-cordierite-beryl does not exceed 4 mole % beryl at 3 kb and 750 °C as well as at 1 atm and temperatures up to 1300 °C. Therefore, the homogeneous cordierite with 15 wt % beryl, prepared by Newton (1966) by devitrifying glass at 1300°, 1 atm, 216 hrs must be a metastable product.

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Dumortierite from Iran: a first record

DUMORTIERITE, not previously recorded from Iran, has recently been found by the writer at an old quarry, Maaden-e-Tala, one km south of Kohrud village in the Kashan district, north-central Iran. The approximate co-ordinates for the locality are: lat. $34^{\circ} 20' N$, long. $51^{\circ} 25' E$. In this area the main rock formation is a large dioritic to granodioritic intrusion cutting a volcano-sedimentary sequence of Eocene age (H. Huber, priv. comm.). The intrusion is cut by porphyritic dykes of similar composition to the main mass, and a later stage of activity is defined by pegmatitic quartzo-feldspathic veins. The dumortierite appears as an apparently pneumatolytic mineral giving conspicuous deep-blue spots on the broken surface of the pegmatitic rock.

Under the microscope the dumortierite is almost invariably in irregular fibrous and felted aggregates, up to one cm across, of minute acicular crystals only a few microns long. Many of the aggregates, and individual crystals where these are observable, are colourless in thin section; in thick sections, however, the aggregates are coloured and are markedly pleochroic from deep-blue to light rusty- or pinkish-brown. The needle-like crystals are elongated fast and have straight extinction. The mean refractive index of the aggregates is close to 1.686; $\gamma-\alpha$ is about 0.012. Identification was confirmed from a powder photograph of hand-picked dumortierite-rich aggregates containing some quartz and sericite.

The main minerals present in the rock are quartz, acid plagioclase, dumortierite, and sericite, with small amounts of epidote, opaque grains (probably ilmenite), sphene, and zircon. In addition, there are rare grains of a mineral with medium refractive index, low to medium birefringence, and rather poorly developed wedge-shaped outline; this is tentatively identified as axinite. From the textural relations the sericite present appears to have partly replaced both feldspar and dumortierite.