Abrupt high/low-transition in flux-grown Mg-cordierite single crystals with hour-glass structure

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Abstract: Mg-cordierite single crystals with a composition Mg$_{1.98}$Al$_{4.08}$Si$_{4.98}$O$_{18}$ were grown from a PbO/V$_2$O$_5$-flux according to the method of Lee & Pentecost (1976). They exhibit an optically visible hour-glass structure with a distinctly higher face-specific Pb concentration up to 0.83 weight % (= 0.02 Pb per formula unit) on the (0001) basal faces. Determination of the distortion index by Weissenberg photographs shows the crystals to be hexagonal ($\Delta = 0.0$) without any deviation from the space group P6/mcc. Nevertheless, the optic axial angle $2V_x$ varies between 0° and 20° in different parts of one single crystal, and features reminiscent of twinning perpendicular to (0001), combined with undulatory extinction can be observed.

Heating of the crystals at 1290°C for 10 hours leads to the disappearance of the hour-glass structure and of the undulatory extinction in the now orthorhombic crystals, while $2V_x$ rises to a homogeneous value of nearly 90°. During heating at 1290°C, the flux-grown single crystals remained hexagonal up to durations of 3.5 hours; after 4 hours a rather abrupt change to the stable orthorhombic form ($\Delta \approx 0.20$) was noted. Thus, at this temperature, the total heating time necessary to transform the metastable hexagonal single crystal into twinned orthorhombic ones is much less, by a factor of 5, than for intergrown polycrystalline aggregates of Mg-cordierite. In fact, the high/low transition itself occurs within less than 30 minutes, whereas polycrystalline aggregates tend to produce intermediate structural states over considerable periods of time.

Key-words : cordierite, single crystals, flux-growth, phase-transition, hour-glass structure.

1. Introduction

Mg-cordierite, Mg$_2$Al$_4$Si$_2$O$_{18}$, occurs in three polymorphic forms. The so-called "µ-cordierite" (Rankin & Merwin, 1918) is a highly metastable hexagonal phase, which appears after short annealing of a glass with cordierite composition between 800° and 1050°C. Schreyer & Schairer (1961b) found that this phase exhibits a stuffed high-quartz structure of the β-eucryptite (LiAlSiO$_4$) type. A detailed structure determination is presently undertaken by Daniels (pers. comm.).

Another hexagonal polymorph, now with the essential features of the cordierite structure, has space group P6/mcc, and is only stable over a small temperature interval between 1450°C and 1463°C at atmospheric pressure (Schreyer & Schairer, 1961a). Above 1463°C it melts incongruently to mullite and a liquid richer in Mg and Si. Below 1450°C the orthorhombic polymorph, space group Cccm, is the stable phase. However, hexagonal high-cordierite generally forms metastably as the first cordierite phase to crystallize also within the stability field of low-cordierite (Schreyer & Schairer, 1961a).

The reduction of symmetry from hexagonal to orthorhombic is caused by the ordering of Si and Al in the tetrahedral framework of