Cordierite under hydrostatic compression: Anomalous elastic behavior as a precursor for a pressure-induced phase transition

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ABSTRACT

The high-pressure behavior of cordierite was investigated by means of in situ experiments using piston-cylinder press and diamond-anvil cell. Static compression in diamond-anvil cells was conducted with various penetrating and non-penetrating pressure media (H2O up to 2 GPa, argon and 4:1-methanol-ethanol up to 7 GPa). The measurement of lattice parameters revealed neither a significant influence on the elasticity nor any indication for effects in analogy to over-hydration within the experimental pressure ranges. Volumetric compression experiments at constant rates up to 1.2 GPa in a piston-cylinder apparatus insinuate subtle irregularities in the low-pressure range at around ~0.35 and ~0.85 GPa. The ΔV/V contribution related to the anomalous compression behavior in that pressure range is of the order of 5 × 10⁻⁴. The results obtained from single-crystal X-ray diffraction between 10⁻⁴ and 7 GPa revealed an unexpected and anomalous linear volume decrease, corresponding to K' = 131±1 GPa for the bulk modulus and K' = −0.4±0.3 for its pressure derivative for a third-order Birch-Murnaghan equation of state. The compressional behavior of the main axis directions is anisotropic with β_a⁻¹ ≈ β_e⁻¹ for an initial pressure regime up to ~3 GPa. At pressures above ~4 GPa, the compression of the a- and b-axis starts to differ significantly, with the b-axis showing elastic softening as indicated by negative values for ∂(β_e⁻¹)/∂P. The diversification between the a- and b-axis is also expressed by the pressure-dependent increase of the distortion parameter Δ. The pronounced elastic softening in both the b-axis and c-axis directions ∂(β_e⁻¹)/∂P = −4.3±0.9, ∂(β_c⁻¹)/∂P = −1.2±0.8 are responsible for the apparent linear bulk compression, which indicates the structural instability and precedes a so far not reported ferroelastic phase transition to a triclinic polymorph, following a primitive lattice above the critical transition at ~6.9 GPa.

Keywords: Cordierite, high pressure, compressibility, elastic softening, phase transition

INTRODUCTION

The mineral physics and structure-property relationships of the silicate mineral cordierite, corresponding to (Mg,Fe)2AlSi2O5 in a simplified form, has received considerable attention for various outstanding physical properties. The eminent properties of cordierite include the pronounced stability at high temperatures and the remarkable robust thermal shock resistance. The ability to sustain large temperature gradients and temperature changes on a short timescale is the consequence of relatively low thermal expansion (Hochella et al. 1979; Mirwald 1981; Ikawa et al. 1986; Camerucci et al. 2001). Another prominent aspect of the thermodynamic properties is related to the polymorphism at high temperatures (Miyashiro 1957; Langer and Schreyer 1969). The associated order/disorder phase transition, which originates from the Al,Si distribution within the aluminosilicate (Al2Si2O5) subunits, is accompanied by a symmetry change from orthorhombic (C2mma) to hexagonal (P6/mmc) symmetry above the critical temperature of ~1750 K (Meagher and Gibbs 1977; Putnis 1980a; Armbruster 1985a; Redfern et al. 1989; Daniels et al. 1994; Malcherek et al. 2001). Apart from the Al,Si ordering on tetrahedral sites and the mechanism of the transformation, numerous investigations focused on the structural origins of optical absorption properties, in particular on the remarkable macroscopic pleochroism in crystals. This includes the Fe2⁺:Fe3⁺ distribution (Faye et al. 1968; Goldman and Rossmann 1977; Abs-Wurmbach et al. 1989; Geiger et al. 2000a, 2000b; Kohnen et al. 2001) and, most recently, the radiocoloration and changes of the optical properties induced by irradiation (Vance and Price 1984; Nasdala et al. 2006; Weikusat et al. 2008, 2010; Miletich et al. 2010). In addition to the complex crystal chemistry, the microporous nature of the framework structure reveals the possibility for structural storage of a variable amount of extra-framework atoms (Na⁺, K⁺, Ca²⁺) and molecules, such as of H2O,

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