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Spiral growth of grossular under hydrothermal conditions

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ABSTRACT

Grossular ($Grs_{99}Adr_{01}$) crystals were synthesized in hydrothermal experiments by the reaction wollastonite + calcite + anorthite = grossular + CO_2 (400 MPa; 695–750 °C; X_{CO_2} = 0.1; solid:fluid = 2:1) using natural minerals as starting materials. Growth spirals are visible due to silica decorations. The crystals are bounded by {110} and {211}. If crystallized at $T \leq 730$ °C, they are birefringent and show sector twinning; grown at $T \geq 730$ °C, they are optically isotropic. The crystal growth rates in the experiments ranged from about 1 to >100 μm/d. The edges of the growth terraces moved by 5–200 nm/s.

The morphology of the growth steps is interpreted against the background of supersaturation with respect to grossular in the fluid. The highest supersaturation occurs during the initial stage of the reaction (~1 h), ultimately at the wollastonite surfaces. In this initial stage, grossular crystallizes with only {111} faces present covered by round growth spirals. When a steady state with respect to dissolution and precipitation rates is reached (after ~24 h), all grossular crystals show {110} and {211} with polygonal spirals on {110} and parallel growth steps on {211}. All straight growth-step edges parallel $[1\bar{1}1]$. When the solid phases assume equilibrium with the fluid (after calcite in calcite-deficient experiments is used up completely), round growth spirals appear on {211}. The spirals are elongated parallel to $[1\bar{1}1]$ and compete against parallel steps in that same direction. The surface morphology of the crystals synthesized under steady state conditions mimicks that of natural grandite crystals, and their growth rates equal the estimated growth rates of grandite crystals from natural hydrothermal systems.