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Stishovite single-crystal growth and application to silicon self-diffusion measurements

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Abstract

Large single crystals of stishovite were successfully synthesized at 11 GPa from a silica solution in water. The potential of both slow cooling and thermal gradient methods were examined. The thermal gradient method provided crystals of $0.8 \times 0.8 \times 1.3$ mm in size grown at 1350 °C and a thermal gradient of 50 °C/mm using stishovite as a silica source. The use of quartz as a source resulted in the appearance of numerous stishovite crystals in the solution interior resulting in diminished space for the growth of large crystals. This can be explained by a significant difference in the solubility of metastable quartz and stishovite in water, estimated to be 85.3 and 5.6 wt% SiO_2 at 1000 °C and 11 GPa, respectively. Crystals up to $0.8 \times 1.3 \times 1.5$ mm were grown by the slow cooling method in the system SiO_2 + 14.7 wt% H_2O as temperature was decreased from 1600 to 1000 °C with a cooling rate of 2 °C/min. The size of single crystals obtained was large enough to carry out silicon self-diffusion experiments, which were performed at a pressure of 14 GPa and temperatures from 1400 to 1800 °C. The lattice diffusion coefficients along the [110] and [001] directions can be expressed as $D_{[110]}$ (m^2/s) = 4.10×10^{-12} exp [–322 (kJ/mol)/RT] and $D_{[001]}$ (m^2/s) = 5.62×10^{-12} exp [–334 (kJ/mol)/RT], respectively, where R is the gas constant and T is the absolute temperature.

Keywords: Stishovite, single-crystal growth, high pressure, aqueous fluid, silica solubility, diffusion