American Mineralogist, Volume 100, pages 1483–1492, 2015

Synthesis of large and homogeneous single crystals of water-bearing minerals by slow cooling at deep-mantle pressures

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Abstract

The presence of water in the Earth's deep mantle is an issue of increasing interest in the field of high-pressure mineralogy. An important task for further advancing research in the field is to create homogeneous single crystals of candidate deep-mantle water-bearing minerals of 1 mm or larger in size, which is required for applying them for the time-of-flight (TOF) single-crystal Laue diffraction method with a third-generation neutron instrument. In this study, we perform several experiments to demonstrate an improved methodology for growing hydrous crystals of such large sizes at relevant transition zone and lower-mantle conditions via very slow cooling over a maximum period of 1 day. Successfully synthesized crystals using this methodology include dense hydrous magnesium silicate (DHMS) phase E, hydrous wadsleyite, hydrous ringwoodite, and bridgmanite (silicate perovskite). It is also demonstrated that these hydrous crystals can be grown from deuterium enriched starting materials in addition to those having a natural hydrogen isotope ratio.

Magnitudes of chemical and crystallographic heterogeneities of the product crystals were characterized by comprehensive analysis of X-ray precession photography, single-crystal X-ray diffraction (SCXRD), field-emission scanning electron microscope (FE-SEM), electron probe microanalyzer (EPMA), secondary ion mass spectroscopy (SIMS), powder X-ray diffraction (PXRD), and TOF neutron powder diffraction (TOF-NPD). The product crystals were confirmed to be inclusion free and crystallographically homogeneous. Compositional and isotopic differences of major elements and hydrogen isotope abundances were lower than 1 and 3%, respectively, among intracrystals and intercrystals within each recovered sample capsule. Phase E crystals up to 600 μm in the largest dimension were grown at a constant temperature of 1100 °C kept for 3 h. Using a lattice parameter-to-temperature relation of phase E, the thermal gradient in the sample capsules for the phase E synthesis has been evaluated to be 20 °C/mm. Hydrous wadsleyite crystals up to 1100 μm in the largest dimension were grown at 1390 °C with a temperature reduction of 70 °C during heating for 10 h. Hydrous ringwoodite crystals up to 1000 μm in the largest dimension were grown at around 1400 °C with a temperature reduction of 110 °C during heating for 12 h. Bridgmanite crystals up to 600 μm in the largest dimension were grown at 1700 °C with a temperature reduction of 30 °C during heating for 12 h. A TOF single-crystal diffraction instrument has been successfully used for analyzing one of the hydrous wadsleyite crystals, which demonstrated that single crystals appropriate for their expected usage are created using the method proposed in the present study.

Keywords: Hydrous minerals, single-crystal growth, phase E, wadsleyite, ringwoodite, bridgmanite, slow-cooling method