

Hydrogen incorporation mechanism in the lower-mantle bridgmanite

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ABSTRACT

Bridgmanite, the most abundant mineral in the lower mantle, can play an essential role in deep-Earth hydrogen storage and circulation processes. To better evaluate the hydrogen storage capacity and its substitution mechanism in bridgmanite occurring in nature, we have synthesized high-quality single-crystal bridgmanite with a composition of $(\text{Mg}_{0.88}\text{Fe}_{0.05}^{2+}\text{Fe}_{0.05}^{3+}\text{Al}_{0.03})(\text{Si}_{0.88}\text{Al}_{0.11}\text{H}_{0.01})\text{O}_3$ at nearly water-saturated environments relevant to topmost lower mantle pressure and temperature conditions. The crystallographic site position of hydrogen in the synthetic (Fe,Al)-bearing bridgmanite is evaluated by a time-of-flight single-crystal neutron diffraction scheme, together with supporting evidence from polarized infrared spectroscopy. Analysis of the results shows that the primary hydrogen site has an OH bond direction nearly parallel to the crystallographic *b* axis of the orthorhombic bridgmanite lattice, where hydrogen is located along the line between two oxygen anions to form a straight geometry of covalent and hydrogen bonds. Our modeled results show that hydrogen is incorporated into the crystal structure via coupled substitution of Al^{3+} and H^+ simultaneously exchanging for Si^{4+} , which does not require any cation vacancy. The concentration of hydrogen evaluated by secondary-ion mass spectrometry and neutron diffraction is ~0.1 wt% H_2O and consistent with each other, showing that neutron diffraction can be an alternative quantitative means for the characterization of trace amounts of hydrogen and its site occupancy in nominally anhydrous minerals.

Keywords: Bridgmanite, lower mantle, hydrogen substitution, neutron diffraction