

H-D inter-diffusion in olivine: implications for hydrogen migration mechanism in the upper mantle

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Karato [2013] has proposed a Nernst-Einstein relation model connecting proton conductivities with hydrogen self-diffusivities at high temperatures when considering multiple hydrogen species in olivine and predicted transformation of dominant hydrogen migration mechanism at 900-1000 K. Although recent conductivity measurements on hydrous olivine single crystals seemed to observe this transformation [Dai and Karato, 2014], comparisons between conductivities and diffusivities at >1000 K has not been yet clear owing to poor knowledge of hydrogen self-diffusion in olivine. On the other hand, the observed transformation has been argued as a result of dehydration at high temperature.. To determine hydrogen self-diffusivities and its anisotropy, isotope inter-diffusion method has advantage to distinguish between incorporation and self-diffusion, and provides more accurate hydrogen diffusion coefficients contributing to proton conduction than traditional incorporation method [Kohlstedt and Mackwell, 1998, Demouchy and Mackwell, 2003]. However, there has been only one research conducted by Du Frane and Tyburczy [2012] through hydrogen (H) and deuterium (D) exchange, which only determined hydrogen diffusivities along a-axis, and might be largely affected by grain-boundary diffusion since they used polycrystals as a source of D. In this study, we employed diffusion couples composed of H and D-doped olivine single crystal pairs to perform H-D inter-diffusion experiments. After determination of crystallographic orientation, a pair of olivine crystals aligned to the same axis was placed together into gold capsule. The polished surfaces were tightly contacted each other. The inter-diffusion experiments were conducted at different temperatures (1000-1300K) and 8 GPa in a Kawai-type multi-anvil apparatus. The H-D inter-diffusion profiles were obtained by SIMS in Hokkaido University. Hydrogen diffusivity along a-axis in olivine determined from this study is about 0.5 order of magnitude lower than that obtained from Du Frane and Tyburczy [2012] at 2 GPa and the activation enthalpies are similar. Comparison with conductivity measurements from Dai and Karato [2014], the activation enthalpies and anisotropy of three typical orientations in olivine are consistent, characterized by the highest mobility and enthalpy along a-axis and the lowest mobility and enthalpy along c-axis. Though enthalpies and anisotropy are same, the expected electrical conductivities from present diffusivities along three typical orientations based on Nernst-Einstein equation are about 1 order lower than Dai's results, respectively.