American Mineralogist, Volume 104, pages 878–889, 2019

Re-configuration and interaction of hydrogen sites in olivine at high temperature and high pressure

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Abstract

Fingerprinting hydrogen storage sites in olivine at high temperature and high pressure is fundamental to understand water distribution and its impact on the upper mantle. We carried out in situ high-temperature and high-pressure IR spectroscopic investigations on hydrogen storage sites in the natural olivine and synthetic Fe-free forsterite. Based on in situ observations of hydrogen in both the natural olivine and synthetic Fe-free forsterite at high temperatures and high pressures, we find that hydrogen does not transfer between storage sites with increasing temperature, but displays disordering at temperatures over 600 °C. In contrast, pressure can induce re-configuration of hydrogen storage sites corresponding to the 3610 and 3579 cm^{-1} bands. Hydrogen storage sites also exhibit disordering at high pressure. In addition, the dehydrogenation experiments of the natural olivine indicate interacts of hydrogen storage sites. Protons released from titanium-clinohumite defects move to pure Si vacancies, and also to Mg vacancies coupling with trivalent cations. This study is the first attempt to fingerprint hydrogen storage sites in olivine at high temperature and high pressure using in situ IR spectroscopy. The implications of the temperature- and pressure-induced disordering and re-configuration of hydrogen storage sites are discussed. The disordering and re-configuration of hydrogen storage sites at high temperature and high pressure favor better understanding of the water effects on physical properties of olivine. The interactions of hydrogen storage sites during dehydrogenation warn that some hydrogen if observed in dehydrated mantle-derived samples may not be original and also make hydrogen diffusivity complex.

Keywords: Hydrogen sites, olivine, high temperature, high pressure, in situ IR spectroscopy, water, upper mantle