

Hydrogen in olivine – how it influences properties and where it resides in the crystalline structure

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As charged point defects, hydrogen ions profoundly influence the kinetic properties of nominally anhydrous minerals, NAMS. Since the first observation of water weakening in quartz in the mid-1960s, experimentalists have investigated the dependence of not only viscosity but also ionic diffusivity and conductivity on hydrogen concentration or water fugacity. For olivine, Mg/Fe diffusivity and Si diffusivity depend on water fugacity to the 1st and 1/3rd powers, respectively, while O diffusivity is, at most, weakly dependent on water fugacity. In both the diffusion and dislocation creep regimes, viscosity decreases approximately linearly with increasing water fugacity. Conductivity varies as water concentration to the 1/2 to 3/4 power. In addition, a number of experimental studies have investigated hydrogen solubility or storage capacity in NAMS with the general conclusion that, for olivine, solubility increases linearly with increasing water fugacity. At the same time, researchers have explored possible mechanisms of incorporation (i.e., speciation) of water-derived point defects in silicate minerals through comparison of FTIR spectra with results from first-principles calculations. This approach predicts that four hydrogen ions associated with a silicon vacancy and one hydrogen ion associated with an oxygen interstitial are the most important incorporation mechanisms with the former dominating. In this case, Si diffusivity and hydrogen solubility are predicted to vary as water fugacity squared, contrary to experimental observations. Here we present an overview of the experimentally determined dependencies of hydrogen solubility and kinetic properties on water fugacity. We then examine the theoretical predictions and experimental constraints on mechanisms of hydrogen incorporation in silicate minerals, exploring areas in which good agreement exists as well as those in which disagreement persists, with an emphasis on provoking further research in both areas.