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Pressure-induced enhancement of proton migration in brucite

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Proton migration plays a key role in the macroscopic properties of hydrous minerals. Knowledge about proton migration in hydrous minerals is critical for understanding point defects, defects-dominated processes and electrical conductivity in the hydrous minerals. Brucite, which is a stable phase in a wide pressure range, has the simple crystal structure, and therefore it is the best material to understand proton migration process in hydrous minerals with layered structure. Pressure effect on the crystal structure of brucite has widely been investigated by high pressure experiments. Regarding the large compressibility anisotropy of brucite between *c* and *a*-axis, brucite is expected to show different conductive behavior with compression. However, there have been no experimental studies about the pressure effect on the hydrogen diffusivity in the hydrous minerals. Here we report the mechanism for proton migration in brucite.

To understand the proton migration process in brucite as a function of temperature and pressure, two different experimental approaches were used: (1) Hydrogen-deuterium (H-D) exchange diffusion experiments and (2) electrical conductivity measurements.

Raman spectroscopy was used to determine the hydrogen concentration. H-D exchange diffusion experiments at pressures from 3 to 15 GPa and 950 K show that the hydrogen diffusivity in brucite is enhanced about one order by compression. The relationship between Log D (m^2/s) and P (GPa) can be described by an asymptotic model: $LogD=-11.73-2.38*0.78^P$. Arrhenius parameters for proton diffusion in the direction of perpendicular and parallel to *c*-axis of brucite at 3 GPa and 750-1050 K yielded a pre-exponential factor of $1.53*10^{-9}$ and $6.86*10^{-11}$ m^2/s and an activation enthalpy of 0.69 and 0.59 eV, respectively. Brucite single crystal shows strong anisotropy of electrical conductivity. Measurements of the electrical conductivity of brucite polycrystals at 2.2- 14.7 GPa and 600 to 750 K show that the conductivity of $Mg(OH)_2$ increased about one order of magnitude with increasing pressure. The proton migration in brucite within its stability is likely to be an n-type mechanism. The enhanced proton migration might correspond to the reduction of $O...O'$ distance with increasing pressure. The mobile proton concentration over the total proton concentration in brucite [x (%)= mobile proton/total proton $*100$] was calculated from the Nernst-Einstein equation. At 3 GPa in a temperature range from 550 to 750 K, x is less than 2.5%, whereas, it reaches to about 10% at 14.7 GPa and 950 K.

Keywords: proton migration, hydrous minerals, brucite, electrical conductivity, diffusion, Raman