

## Fast diffusion of water in silica glass

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Water diffusion in silicate glass is a fundamental process controlling physical and chemical aspects of magmatism, but its diffusion mechanism is not fully understood. In this study, we report one-order of magnitude faster diffusion of water in SiO<sub>2</sub> glass than that reported previously<sup>[1]</sup>, observed in diffusion experiments of deuterated water (<sup>2</sup>H<sub>2</sub>O) in SiO<sub>2</sub> glass at 900-750°C with water vapor pressure of 50 bar.

The <sup>2</sup>H concentration profile, measured by SIMS (Cameca ims-6f at Hokkaido Univ.), rapidly decreases from rim to core of the sample. This profile can be explained by a water diffusion model in SiO<sub>2</sub> glass<sup>[1]</sup>, where molecular water diffuses through the diffusion pathways formed by breaking Si-O-Si bonds via a Si-OH formation reaction. We found that the tail of <sup>2</sup>H diffusion profile extends further into the deep region of the sample that cannot be explained by the previous diffusion model<sup>[1]</sup>. This suggests that a small fraction of <sup>2</sup>H-bearing species migrates with a faster diffusion rate.

The fast diffusion profile can be fitted by a diffusion model with a constant diffusivity. The obtained diffusivity is about one order of magnitude larger than that of water in SiO<sub>2</sub> glass<sup>[1]</sup>, and satisfy the Arrhenius relation. The obtained activation energy ( $75 \pm 31$  kJ/mol) suggests that the diffusion species is molecular water, and that fast diffusion pathways are present for molecular water in a SiO<sub>2</sub> glass.

The fast diffusion coefficient of water at 900-750°C and its activation energy resemble the relation between diffusivities of noble gases in silica glass and their size<sup>[2]</sup>. Noble gas diffuses through “free volume sites” in the SiO<sub>2</sub> glass structure, and the activation energy for diffusion depends on the atomic radius. We therefore conclude that molecular water can diffuse through free volume sites, which provide faster diffusion pathways than those formed by Si-OH. The concentration of free volume sites, estimated from the solubility of noble gases<sup>[3]</sup> is much higher than that of <sup>2</sup>H observed in the fast diffusion in this study. This implies that free volume sites are not fully occupied by molecular water under the present experimental conditions. The contribution of fast diffusion may increase at higher water vapor pressures, where the concentration of molecular water dissolved into free volume sites is likely to increase as noble gases.

[1] Kuroda M. *et al* (2018) *Am. Min.*, **103**, 414-417. [2] Behrens H. (2010) *Rev. Min. Geochem.*, **72**, 227-267. [3] Shackelford J.F. (1999) *J. Non-Cryst. Solids*, **253**, 231-241.