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Ab initio study of diffusion mechanism of water in silicate melt

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Water diffusion in silicate melt is a fundamental process crucial for comprehending transport properties and dehydration kinetics, pivotal in understanding magmatic processes and volcanic evolution [1-3]. Despite its significance, deciphering water diffusion has posed challenges in both experimental and theoretical studies, owing to the rapid liquid dynamics and interconversion of hydrous species [3]. Existing diffusion models primarily focus on two water species: molecular water and hydroxyl groups. However, emerging evidence suggests the involvement of complex species and protons in water diffusion. In response, we developed a bonding analysis method to unambiguously identify stable and transition-state hydrous species, and obtained their characteristic diffusion coefficients by using restrained ab initio molecular dynamics simulations at high temperatures and high pressures. We find that the stable species include hydroxyl group, molecular water and a trace amount of hydronium. Proton is only a transition-state species, but proton hopping dominates the overall water diffusion with a contribution over 60%, primarily through triggering the rotational motion of the O-H bond. Thus, the synergy between proton hopping and O-H bond rotation emerges as the key driver for rapid water diffusion in silicate melt. This mechanistic understanding facilitates the development of a robust water diffusion model grounded in thermodynamics. Furthermore, extrapolating this model to temperatures below 2000 K yields results consistent with experimental observations.

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