

Understanding Molecular Structure and Diffusion Dynamics of Nanoconfined Water in Imogolite Nanotubes

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Confined water plays a pivotal role in subsurface geochemistry, influencing processes such as mineral dissolution, ion exchange, and solute transport. In this study, we investigate the molecular configurations and diffusion dynamics of water confined within nanotubes, using imogolite, a naturally occurring aluminosilicate nanotube material, as a model system. By employing a suite of advanced nuclear magnetic resonance (NMR) techniques, including both solid-state and solution-state approaches, such as magic angle spinning (MAS), correlation spectroscopy (2D-HETCOR), diffusometry, and T_1/T_2 relaxometry, we characterize and differentiate water configurations and mobility inside and outside the nanotubes. We have identified distinct T_2 values of water and imogolite hydroxy groups, and different coupling strengths of water ^1H with Si-OH / Al-OH in the partially and fully hydrated imogolite samples. This indicates different confined structures and diffusion dynamics of the inner and outer-tube bound water. We believe that this combined approach enables us to identify distinct hydration states and provide detailed insights into the interaction of confined water with the nanotube surfaces, with the help of molecular dynamic simulations, and our findings could lead to direct, quantitative measurements of water structure and diffusion kinetics in confined environments, ubiquitous in the natural mineral systems.