Surface Hydrophobicity and Properties of Interfacial Water

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The structure, energetics and kinetics of various reactions occurring at the phyllosilicate-water interfaces are relevant to numerous processes such as heterogeneous nucleation and subsurface reactive transport properties. Phyllosilicates are of particular interest as their intrinsic chemical heterogeneity can give rise to diverse crystal structures and surface physicochemical properties such as hydrophobicity, which in turn determines the nature of interactions.

We investigated the effects of fluorine substitution on the hydrophobicity of the basal surface of phlogopite in the presence of various counterions. Water adsorption processes were monitored *in situ* using near-ambient pressure X-ray photoelectron spectroscopy and interfacial water properties were probed by X-ray reflectivity, which was further combined with classical molecular dynamics simulations to obtain energetic properties. Results suggest that the surface hydrophobicity of phlogopite stems from a competitive contribution from surface oxygens and counterions for water molecules. Our study overall provides molecular descriptions of how surface hydrophobicity is manifested.