

Atomistic simulation of the structure and reactivity of hydroxylated mineral surfaces in aqueous conditions

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We present our recent work modelling hydroxyl terminated mineral surfaces and nanoparticles in contact with water. Our approach is to combine electronic structure methods based on DFT with potential-based simulation techniques. Where possible simulations are supported with experimental data, such as Raman spectroscopy and atomic force microscopy (AFM). We will briefly describe our procedure for generating the surfaces, which begins with a potential based approach to screen many surface terminations thereby identifying the most stable. These are then modelled using DFT including van der Waals corrections to provide a check on the relative stability and to allow us to explore the surface composition. Finally, the surfaces of either bulk crystals or nanoparticles are immersed in water to explore the effect of the solution on structure and transport at the mineral-water interface, which is achieved using primarily potential-based molecular dynamics simulations.

We will describe how the surface composition affects both the surface structure and transport properties in solution by comparing the results from different mineral systems. These will include a comparison of the alkaline earth hydroxides of portlandite and brucite with the alumina and alumino-silicate mineral surfaces. In each case on forming a layered structure the basal plane is calculated to be more stable and hence less reactive. The reactivity is investigated by studying the interaction of the surfaces with both water and carbonate species. We find that interaction with carbonate is far stronger at the edge surfaces, but as yet have been studied much less. Indeed, we find that in some cases there are large reconstructions. Finally, in addition to describing the structure and transport at these interfaces, we show how simulation approaches can give useful insight into the atom-level mechanisms involved, not least as the hydrogen content of the surfaces are modified.