

Computational modelling of the role of water and CO₂ on adsorption at oxide mineral surfaces

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We present our recent work using atomistic simulation techniques to model the surface and stability of hydroxylated mineral surfaces in contact with CO₂ and water, and how the resulting surface alteration affects adsorption processes. Our approach is to combine potential-based simulation techniques with standard electronic structure methods based on DFT. In each case we calculate the vibrational properties, which helps in identifying dynamically unstable structures, evaluate vibrational free energies and predict spectroscopic data. The latter can be used to compare with experimental IR and Raman spectroscopic data, thereby validating the approaches. We will begin by briefly describing our procedure for identifying thermodynamically likely structures and compositions and the resulting structures.

We will then describe how the surface composition affects both the surface structure properties in solution, focussing particularly on alkaline earth hydroxides, portlandite and brucite. We find that much of the differences are the result of the relative reactivities of the surfaces. For example, we find that interaction with carbonate is far stronger at the edge surfaces, but as yet have been studied much less. Finally, we show how simulation approaches can give useful insight into the atom-level mechanisms involved, not least how the surface composition modifies adsorption processes.