

## Molecular simulations of CO<sub>2</sub> in aqueous solutions at calcite surfaces

A. SILVESTRI<sup>1</sup>, A. BUDI<sup>1</sup>, E. ATAMAN<sup>1</sup>,  
M.P. ANDERSSON<sup>1</sup>, S.L.S. STIPP<sup>1</sup>,  
J.D. GALE<sup>2</sup> AND P. RAITERI<sup>2</sup>

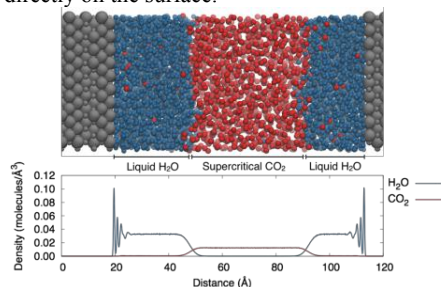
<sup>1</sup>Nano-Science Center, Department of Chemistry, University of Copenhagen, Universitetsparken 5, 2100 København Ø, Denmark (\*alessandro.silvestri@nano.ku.dk)

<sup>2</sup>Curtin Institute of Computation, The institute for Geoscience Research (TIGeR), Department of Chemistry, Curtin University, GPO Box U1987, Perth, WA 6845, Australia

Mineral carbonation, the reaction of CO<sub>2</sub> with minerals to form stable carbonates, is a promising option to store CO<sub>2</sub> and mitigate the induced climate change and ocean acidification [1]. To gain a fundamental understanding of the sequestration process, it is essential to study the reactions and interactions of CO<sub>2</sub> with mineral surfaces.

Previous simulations [2] have shown that CO<sub>2</sub> adsorbs in preference to water on calcite but this was a direct result of a poor description of CO<sub>2</sub>-calcite intermolecular interactions. We used density functional theory to calculate the adsorption energy of a CO<sub>2</sub> molecule on the calcite {10.4} surface. The reproduced adsorption energy landscape was then used to parametrize a classical force field.

The new set of force field parameters was used along with a thermodynamically consistent force field for calcite [3] to run molecular dynamics simulations. For all pressures and temperatures in the studied range, CO<sub>2</sub> was not able to penetrate the structured water layers and therefore could not adsorb directly on the surface.



**Figure 1.** Snapshot from an MD simulation of CO<sub>2</sub> (red) and water (blue) on calcite (gray) at 25 MPa and 323 K and the corresponding density profile.

[1] IPCC (2013), *Climate Change: The Physical Science Basis*. Cambridge University Press

[2] Van Cuong *et al.* (2012) *Mol. Phys.* **110**, 1097–1106

[3] Raiteri *et al.* (2015) *J. Phys. Chem. C* **119**, 24447–24458