Atomistic Simulations of CO_2 Sequestration by Magnesium Containing Minerals

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We describe recent work using atomistic simulation methods based on the Born model of solids to study the sequestration of CO_2 on mineral surfaces and nanoparitcles. The aim of our work is to identify the atomic processes which control the incorporation of CO_2 into minerals, and ultimately identify which surfaces of which minerals under what conditions will be optimised for CO_2 incorporation.

We have begun this study by considering a number of magnesium containing minerals: periclase, MgO; brucite, $Mg(OH)_2$; forsterite, $MgSiO_4$; and serpentines, $Mg_3Si_2O_5(OH)_4$. In each case we have used atomistic simulation techniques to model the most stable surfaces and energetics of incorporating CO_2 as carbonate ions both in the absence and presence of water. As a further check on the viability of the quality of the models describing the interatomic forces, we have have performed some density-functional theory calculations.

Finally, we recognise that fine, ground mineral particles, with high surface areas are more reactive than flat surfaces. Therefore we have calculated the energetics of the process on small nanoparticles.

The results of the simulations show that the lowest energy carbonated surfaces of MgO that we have found so far are the {n10} stepped surfaces with terraces of the more stable, 'perfect' {100} surface. For example, the {310} carbonated surface energy is reduced to 0.56 J m^{-2} from 1.91 J m^{-2} for the stochiometric surface. Moreover, when CO_2 binds to the surface oxygen, the magnesium ions relax out of the surface, encouraging the formation a $MgCO_3$ monolayer.

Another general feature is that water adsorption appears to enhance the carbonation by causing a further reduction in surface energy. However, hydroxylated surfaces are more resistant, this has been found for not only brucite but also for hydroxylated {110}, {111} and {310} MgO surfaces.

Initial results suggest that the carbonation of forsterite surfaces is relatively unfavourable. We are now beginning to investigate methods for activating the surfaces, including small particle sizes.

In summary, atomistic simulation methods, thanks to increased computer power and to reliable descriptions of interatomic forces, can now begin to provide useful predictions for complementary experiments.