Wollastonite hydration, dissolution, and calcite precipitation for targeted mineral carbonation

FLORENCE T. LING[1*], ZHIYUAN TAO[2], DAN PLATTENBERGER[2], JEFFREY P. FITTS[1], CATHERINE A. PETERS[1], ANDRES CLARENS[2]

[1]Princeton University, Civil and Environmental Engineering, Princeton, NJ 08540, ftling@princeton.edu,
[2]University of Virginia, Civil and Environmental Engineering, Charlottesville, VA 22903

The storage of CO_2 in geologic reservoirs can help mitigate climate change, but CO_2 leaks from reservoirs may potentially damage potable water aquifers and underground natural gas reserves. To explore engineered approaches to seal leakage pathways, we investigated targeted mineral precipitation by delivery of temperature-sensitive coated wollastonite ($CaSiO_3$) particles. The reactivity of the system was studied in sintered glass bead column (GBC) laboratory experiments. $CaSiO_3$ particles were delivered into the GBC pore space, comprising ~20% of the total pore volume. GBCs were then placed in a reactor with 200 mL H_2O, 150 bar CO_2 or N_2, and reacted at 120°C for 48 hrs. Permeability was measured with customized air permeametry before and after reaction. Reacted GBCs were imaged with X-ray computed tomography and sampled for scanning electron microscopy/electron-dispersive X-ray spectroscopy (SEM/EDS), Raman, and X-ray diffraction (XRD) analysis.

The permeability of the GBCs with $CaSiO_3$ decreased by an order of magnitude after 48 hrs. Raman, SEM/EDS, and XRD identified $CaCO_3$ and $SiO_{2(am)}$ post-reaction. Interestingly, calcium silicate hydrate was also identified, distinct from the original $CaSiO_3$. Prior studies recognize $CaSiO_3$ dissolution, $CaCO_3$ precipitation, and $SiO_{2(am)}$ precipitation under similar conditions to our system. We now add $CaSiO_3$ hydration and subsequent dissolution of calcium silicate hydrate to produce Ca^{2+} ions for carbonation. This reaction product is expected to play an important role in controlling permeability evolution. A one-dimensional reactive transport model was developed to simulate the geochemical reactions and predict permeability changes. This model may be used in practical applications of predicting leakage mitigation of CO_2 storage reservoirs.