Rock/cement fracture in geological CO_2 storage

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CO_2 injection wellbores for Geological Carbon Storage (GCS) are vulnerable to gas or fluid leakage. During and after the injection, the resulting CO_2-rich acid water may penetrate, through fractures, the interface between the cement well and the surrounding sedimentary rocks. In this study we aim to assess the reactivity of cement and rocks and to understand the chemical processes occurring at this interface under GCS conditions.

Artificially fractured cores of 5 cm in length and 2.5 cm in diameter were made of half cylinders of cement and rock (sandstone, limestone and marl). They were reacted with a synthetic sulfate-rich saline water under (a) atmospheric conditions ($10^{-3.4}$ bar CO_2 and 25 ºC, pH 6.2) and (b) GCS conditions (130 bar CO_2 and 60 ºC, pH $\approx$ 3) in flow-through reactors. The evolution of solution chemistry at the outlet (Na, K, Ca, Mg, Si, Al, SO_4) was monitored over time.

Under atmospheric conditions, the pH of the output solution increased up to 12 in the first 24 h (due to portlandite dissolution), leading to a deficit in aqueous Mg caused by precipitation of a brucite layer over the fracture walls (observed through SEM-EDS and XRD analyses). After this first stage pH was about 8 as calcite dissolved. Under GCS conditions, the input pH of the CO_2-rich injected solution was about 3, promoting a strong dissolution of calcite (rock wall) and portlandite (cement wall). These reactions led to an increase in both the fracture aperture at the very inlet and initial porosity. After portlandite exhaustion, the pH of the output solution was around 6 and was buffered by calcite dissolution.

2D reactive transport modeling was used to reproduce the aqueous chemistry of the outflow. The model was calibrated by adjusting the mineral reactive surface areas and using experimentally-determined effective diffusion coefficients.