

Self-sealing of faults by CO₂-rich fluids

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Long-term geologic storage of anthropogenic "greenhouse gases" such as CO₂ is a proposed approach for addressing human-induced climate change. Deep subsurface brine reservoirs in geological sedimentary basins are possible sites for sequestration, given their ubiquitous nature. Leakage through pre-existing or injection-induced faults is a primary concern of risk assessment. We examined some natural CO₂ reservoirs in central Utah, USA, including areas of known leakage from such reservoirs. For example, every day the Crystal Geyser near Green River, Utah, erupts CO₂-charged water through the pipe of an abandoned well into a fault zone above a natural CO₂ reservoir, emitting up to 350 kg CO₂ per minute during each eruption. Other nearby faults in this area leak large amounts of CO₂, as evidenced by CO₂-rich groundwater leaking from springs, geysers, and abandoned exploration wells, and by tufa and travertine deposits that fill the surface outcrops of these fault zones. We evaluated potential time-scales of such fault "self-sealing" using reactive transport simulation models of one particular fault zone, the Little Grand Wash fault zone, just a few km from Crystal Geyser. We also conducted laboratory CO₂-brine flow tests on example rock cores, and constructed reactive transport models of the flow tests. Comparison of the two sets of simulation models suggest very similar self-sealing mechanisms, with changes in pCO₂ followed by outgassing and carbonate precipitation. Finally, we are comparing results of this analysis to evaluate the potential for self-sealing behavior in other systems, including ongoing geologic storage test sites in southern Utah, northern New Mexico and western Texas, USA. These ongoing tests are part of the Southwest Regional Partnership on Carbon Sequestration testing program, funded by the U.S. Department of Energy and managed by DOE's National Energy Technology Laboratory.

The growth of pyramids and pits on hematite mediated by redox chemistry and bulk electron transport

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Under non-equilibrium conditions, electric potential gradients can be generated in hematite (α -Fe₂O₃), and the resulting electron transport, via a polaron hopping mechanism, couples the redox reactions that take place on different crystal surfaces. When hematite crystals are reductively dissolved at pH 2-3 in 1 mM Fe²⁺ in the presence of 10 mM oxalate, large (\approx 200 nm high) pyramids grow on the (001) surface while dissolution of the (012) and (113) surfaces results in the formation of etch pits. Electric potential measurements indicate that electrons are driven from the (001) surface to adjacent surfaces, facilitating the oxidation of Fe²⁺ to Fe³⁺ and its incorporation into the growing (001) surface.

Our efforts to understand the coupling between the surface chemistry, bulk electron transport and the evolving surface geometry has focused on the growth of pyramids on the (001) surface. Conceptual models based on slope dependent surface currents and orientation (relative to crystal symmetry axes) dependent surface energies and surface kinetics will be discussed, and progress towards simulation of the surface morphologies observed during the reaction of hematite, using kinetic Monte Carlo simulations and continuum surface growth models will be described.