Experimental and Theoretical Alteration of Basalt by Supercritical CO_2: Implications for CO_2 Sequestration

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Basaltic rocks have potential as repositories for sequestering carbon dioxide (CO_2) because of their capacity for trapping CO_2 in secondary carbonate minerals. In order to ascertain basalt utility in CO_2 sequestration, it is necessary to understand its reactivity with CO_2 under various pressure and temperature conditions. We carried out series of geochemical models and experiments, reacting tholeitic basalt with CO_2-charged fluids over a range of conditions from 50 - 150°C and from 100 - 300 bars. Experiments were carried out in both fixed-volume titanium reaction cells for batch processing and in a flexible gold-cell apparatus with a serial online fluid sampling capability to permit monitoring reaction progress. Basalt has a high reactivity to supercritical CO_2 and carbonic acid. Initial reactions caused a rapid drop in pH and an increase in dissolved cations including Ca, Mg, Fe, and Mn followed by a decrease in cation concentrations to steady-state values. CO2 decreased asymptotically to steady-state concentrations in experiments that were under-saturated with CO_2, a repeatable pattern following re-injection of CO_2 into the experiment, showing that basalt has a large capacity to react with CO_2. Batch experiments reacting olivine basalt with supercritical CO_2 at 100°C, 300 bars PCO_2 for two months resulted in almost complete alteration of the rock with 90% of the divalent cations (primarily Mg, Fe, and Ca) having been incorporated into carbonate minerals.

Reaction path simulations using the computer program CHILLER, which computes multi-component heterogeneous chemical equilibria, predicted calcite would initially precipitate, then later dissolve with the sequential addition of PCO_2 then be replaced by dolomite and siderite at equilibrium. SEM analyses of the solid reaction products found only Fe and Mg bearing carbonates consistent with the kinetic barriers to forming dolomite.

These studies indicate that basaltic rocks may quickly and effectively sequester CO_2. CO_2 sequestration can be accomplished either by mineral carbonation with mined materials or by in situ injection of CO_2 into a basalt formation. Mineral sequestration may be safer than traditional geologic carbon sequestration because it permanently changes the CO_2 into a solid, fixing it in a thermodynamically stable form that will not leak.