

Abstract

The potential dangers with increased concentration of CO₂ in the atmosphere, such as climate changes and sea level rise, have lead to an interest in CO₂ sequestration in geological formations. The thermodynamically most stable way to store carbon is as carbonate minerals. Carbonate mineral formation, however, requires divalent cations originating from a non-carbonate source. One such source is basaltic rocks which contain high concentrations of Ca²⁺, Mg²⁺ and Fe²⁺. The potential for forming carbonate minerals through the injection of CO₂ into basalt is under investigation in Iceland and several other places around the world. The aim of this thesis is to help optimize carbonate mineral precipitation in basalts during CO₂ injection through a series of related field and laboratory studies.

A detailed study of the chemical composition of the groundwater surrounding the Mt. Hekla volcano in south Iceland was performed to assess fluid evolution and toxic metal mobility during CO₂-rich fluid basalt interaction. These fluids provide a natural analogue for evaluating the consequences of CO₂ sequestration in basalt. The concentration of dissolved inorganic carbon in these groundwaters decreases from 3.88 to 0.746 mmol/kg with increasing basalt dissolution while the pH increases from 6.9 to 9.2. This observation provides direct evidence of the potential for basalt dissolution to sequester CO₂. The concentrations of toxic metals in these waters are low and reaction path modeling suggests that calcite and Fe(III) (oxy)hydroxides scavenge these metals as the fluid phase is neutralized by further basalt dissolution.

The rate limiting step for mineralization of CO₂ in basalt is thought to be the release of divalent cations to solution through basaltic glass dissolution. The dissolution rate of basaltic glass can be increased by adding ligands which complex aqueous Al³⁺. Aqueous SO₄²⁻ can complex Al³⁺ and the effect of SO₄²⁻ on the dissolution rate of basaltic glass was studied using mixed flow reactors at 3 < pH < 10 at 50 °C. Moreover, sulphur is often present in the flue gases of power plants and their disposal also poses an environmental challenge. If possible, co-injection of sulfur with CO₂ could provide a novel cost effective disposal method for industrial generated sulphur. Consistent with current models describing basaltic glass dissolution by aqueous solution composition, results show that SO₄²⁻ increases the dissolution rate of the glass in acidic conditions, while no effect was found in alkaline solutions. These results suggest both that 1) co-injection of sulfate may accelerate CO₂ mineralization in basalts, and 2) existing kinetic models provide an accurate description of basaltic glass dissolution.

To further assess the potential effect of SO_4^{2-} on the precipitation rate of carbonates, steady-state rates of calcite precipitation were measured in mixed flow reactors at 25 °C and pH ~9.1. The results show that 0.005 M Na_2SO_4 decreases the precipitation rate of calcite by ~40%. This result suggests that co-injected sulphate could slow calcite precipitation in the subsurface at pH conditions typical of calcite precipitation. Further experiments are planned to completely define these effects at conditions expected at subsurface CO_2 injection sites.