

Experimental Assessment of Basalt Dissolution Rates under Shallow Aqueous Carbon Dioxide Sequestration P_{CO_2} and Temperatures

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Sequestration by CO_2 mineralization in basalt formations is a greenhouse gas emission mitigation strategy. An advantage of this strategy is that CO_2 injected as a free phase or aqueous phase reacts with the basalt immobilizing CO_2 as carbonate minerals. Free phase injections are limited to pressures greater than the critical pressure of CO_2 whereas aqueous injections allow for shallower depths limited by CO_2 solubility under formation temperatures, pressures, and geochemical conditions. Pilot/field-scale demonstrations of injected supercritical CO_2 (Wallula Basalt Pilot Project, SE Washington, USA) or aqueous CO_2 (CARBFIX, Iceland) show that CO_2 rapidly reacts with basalts to form carbonate minerals over relatively short time frames.

We are planning to conduct a aqueous CO_2 injection into the basalts of the Eastern Snake River Plain (ESRP), Idaho USA. In preparation, we conducted twelve, 60-plus-day long batch laboratory water-rock- CO_2 experiments at temperatures of 21 and 40°C, P_{CO_2} of 15 and 45 bars (1.5 to 5.5 wt.% CO_2), and crushed basalt with effective grain diameters of 0.22 and 0.57 mm to assess the dissolution rates of ESRP basalt. Initial silica release rates were calculated from sampling-corrected experimental concentration vs. time profiles. Essentially no difference in rates were observed between experiments conducted under the same P-T conditions but with different grain sizes. This result is consistent with measured B.E.T. surface areas which differed from each other by less than 4%. Rates at 40°C and 45 bars were 1.8 ± 0.11 and 1.3 ± 0.06 times faster than for 21°C and 15 bars experiments, respectively. Using B.E.T. surface area normalized initial silica release rates, an empirical rate law of the form $r = A \cdot e^{-E_a/RT} \cdot (m_{\text{CO}_2})^n$ was derived where E_a is the activation energy (42.5 ± 5.0 kJ/mol) and n is the reaction order with respect to CO_2 molality. This rate law predicts that at constant m_{CO_2} the silica release rate is 2.9 times faster at 40 compared to 21°C and reflects the opposite effects of increasing temperature on basalt reaction rates (increasing) and CO_2 solubilities (decreasing). On going activities include assessment of release rates for other elements and considerations of the observed time dependency on rates.