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Feldspar Dissolution as a Source of Cations for Carbonate Growth in a Carbon Sequestration Context: EQ3/6 Modeling and Laboratory Experiments

Carbon sequestration by CO_2 injection into deep saline aquifers may result in long-term chemical alteration of host phases and eventual growth of carbonate minerals. EQ3/6 was used to model alteration scenarios with available kinetic data. Calculation predictions were tested in four experiments to determine if carbonate minerals would form under specific laboratory conditions as predicted by calculation. Each experiment used Dickson-type rocking gold bag autoclaves running for 3 to 5 months with 75-100 μm-sized An_{60} plagioclase grains at 150° C and pressures of 100 bars. Variables include initial CO_2 fugacity (from 0.15 to 70 bars), supporting electrolyte, alkalinity and pH. Calculations suggest that enough alkalinity could be produced by reaction with plagioclase to reach saturation with respect to carbonate minerals; higher fCO_2 conditions led to saturation with respect to dawsonite [$NaAlCO_3(OH)_2$] at pH 3.6, and lower fCO_2 conditions led to saturation with respect to calcite at pH 5.9 and at pH 6.9 (with and without 0.01 m starting alkalinity, respectively). The reacted plagioclase grains were analyzed by petrography, XRD, SEM/EDS, CO2 analysis, and AFM. Petrography and powder XRD did not reveal any carbonate precipitate. SEM and AFM images showed no evidence for secondary carbonate mineral precipitation, and analysis of the grains for CO_2 exsolution after acidification revealed no carbonate source. The plagioclase dissolution rates were highest initially then progressively slowed as the solutions became more saturated with respect to the feldspar. Because of the complex initial aqueous conditions, dissolution congruency could not be precisely established. This work highlights the need for longer-term field and laboratory experiments to test geochemical aspects of CO_2 sequestration, as well as for evolution of codes to better account for dissolution/growth inhibition, nucleation, and other processes.