

Deciphering coupled labradorite dissolution and secondary precipitation reaction kinetics using multiple isotope tracers

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Our current knowledge of geochemical kinetics is largely based on single mineral dissolution far from equilibrium. However, geologic systems that store CO₂, e.g., soils and aquifers, are multi-mineral systems at near-equilibrium. There is a lack of experimental data for such systems. Following our isotope doping studies of single minerals in the last ten years, we recently initiated multiple isotope tracer studies of the multi-mineral systems. Here we report the experiments of coupled labradorite dissolution, calcite precipitation, and clay precipitation in batch reactor systems. Plagioclase was chosen as the reactant because it is both a major component and among the most reactive minerals in basalts. ²⁹Si, ⁴³Ca, Li, and Ca¹³CO₃ seeds were introduced into the initial solutions. The isotope ratios in the experimental aqueous solutions were measured with MC-ICP-MS. Unidirectional rates of labradorite dissolution near equilibrium ($\Delta_r G = \sim -19.4$ kJ/mol to -4.2 kJ/mol) are about 10²~10⁴ times slower than the far-from-equilibrium rates reported in the literature. Calcite precipitation occurred very near equilibrium and the near-equilibrium precipitation rates were $\sim 10^4$ slower than the reported far-from-equilibrium rates in the literature. Imogolite or allophane precipitated near equilibrium. The experimental data show that the coupled reactions below arrested the system in a steady state that is close to labradorite equilibrium but not at equilibrium:

Labradorite + H₂O + HCO₃⁻ = Calcite + Imogolite + Aqueous ions

Overall, this study of multi-mineral reaction kinetics, together with our other studies also presented at this Goldschmidt on near-equilibrium modeling (Chen Zhu), multiple isotope tracer experiments on basalts (Janelle Cook et al.), and a comprehensive review of basalt-water-CO₂ experiments since 2000 (Peng Lu et al.), barite dissolution and precipitation and Ba and S isotope fractionation at solubility equilibrium (McKailey Sabaj et al.), aims to calibrate geochemical models before models are applied to the complex field systems.