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Evaluating the CO_2 Mineralization Potential of Ultramafic Rocks via Experimental Determinations of Near-Equilibrium Serpentinite Reaction Rates

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Peridotites and their hydrated counterparts (serpentinites) naturally react with CO_2 to form carbonate minerals; consequently, engineered versions of this reaction have been proposed as a climate change mitigation strategy. However, because most experimental studies are conducted at far-from-equilibrium conditions, the rates of serpentinite-fluid reactions at field-realistic, near-equilibrium conditions remain poorly constrained. While far-from-equilibrium experiments have provided useful indications of overall serpentinite reactivity, an improved understanding of near-equilibrium reaction rates will be vital to predicting the efficacy of carbon mineralization. To this end, we conducted a series of batch and flow-through experiments to measure the near-equilibrium reaction kinetics of a serpentinite sample from the Coquihalla serpentine belt in southwestern British Columbia, Canada. Batch experiments were conducted at 22°C and 1 bar, with fluid-to-rock ratios of 10 and 125, and initial pH conditions of ~3.5 and ~5.5. Flow-through experiments were conducted at 25°C, 55°C and 75°C and 1 bar, with initial pH conditions of ~3.5 and ~5.5. The flow-through system employed a novel design that enables recirculation of reacted fluid through the system, allowing us to target near-equilibrium conditions. The experiments starting at higher pH utilized a technique known as Si isotope doping, in which the experimental solution is spiked with $^{29}SiO_2$, allowing us to measure small concentration changes of Si isotopes to determine dissolution and precipitation rates simultaneously. Experiments show that dissolution and precipitation rates slow considerably as equilibrium is approached and thermodynamic drive decreases. We observe that the time to reach near-equilibrium conditions is significantly quicker for low fluid-to-rock ratio experiments and experiments starting at lower pH release more Mg. The rates determined in this study can be used to parameterize CO_2 mineralization models and improve predictions of CO_2 mineralization timescales in ultramafic rocks.