

Uptake of isotopic spike by Si-rich alteration layer during olivine dissolution

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Olivine dissolution was studied under conditions relevant for subsurface carbon dioxide storage (60 °C, 100 bar P_{CO₂}) for periods of 2 and 19 days in batch reactors in order to better understand the Si-rich surface layer that forms under these conditions. The initial aqueous reaction fluid was spiked with approximately 0.3mM ²⁹Si. Aqueous samples were withdrawn from the experiments at various times to track the reaction progress, and were analyzed for elemental concentration, alkalinity, and total dissolved carbon. The samples were analyzed for silicon isotopic concentrations using a multicollector ICP-MS. After conclusion of each experiment, the reacted olivine grains were analyzed with a high-resolution ion microprobe to measure the silicon isotope ratios in the solid as a function of depth. The ²⁹Si/³⁰Si ratio of the aqueous phase decreases with time as the spike is diluted by silicon from the dissolving olivine and after 19 days, approaches the natural ratio of 1.52. Correspondingly, the mineral surfaces reacted for 2 days show less ²⁹Si uptake than mineral surfaces reacted for 19 days. Interestingly, olivine reacted for only 2 days does have a Si-rich alteration layer on its surface; however, very little ²⁹Si is present (mineral ²⁹Si/³⁰Si=1.72, aqueous=3.56). This result indicates that the alteration layer forms very quickly and not from precipitation of Si from the bulk solution, but rather from leaching or a very localized dissolution/ precipitation process. Olivine grains reacted for 19 days have an alteration layer with a ²⁹Si/³⁰Si composition slightly greater than the aqueous fluid (mineral =1.88, aqueous = 1.54), suggesting that SiO₂ precipitation dominates at longer times. For the application of sub-surface carbon storage, the mechanisms that dominate over longer times are most relevant.