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Experimental Determination of Low-Temperature Ultramafic Silicate Alteration Rates Using Silicon Isotopes

LAUREN GASIOR, JUAN CARLOS DE OBESO, CHRISTOPHER R ANDERSON AND DIEGO P FERNANDEZ
University of Utah

The alteration of mantle peridotites into serpentinites at mid-ocean ridges and subduction zones, known as serpentinization, is an important global process that occurs in many different temperature and pressure regimes. However, past attempts at quantifying the rates of this reaction at geologically low temperatures (<150°C) have been unsuccessful due to slow kinetics, resulting in chemical changes below analytical uncertainty (Lamadrid, 2021). Constraining these rates can provide a better understanding of this process and inform the viability of using ultramafic mantle peridotites for mineral-based carbon sequestration and storage. Serpentinization is also the primary source of H_2 reservoirs on Earth, which are being explored as a carbon-free energy alternative, and thought to have supported early microbial life. Here, we present the results from low-temperature serpentinization experiments, utilizing a novel ^{29}Si doping technique (Gruber, 2013) to track mineral dissolution and precipitation over time using ICP-MS.

Using natural olivine minerals, experiments were run at temperatures from 20°C to 80°C with varying mineral grain sizes up to 106 μm to constrain the effects of temperature and surface area during surface alteration. The experimental solution was doped with ^{29}Si, which is rare in natural samples, to track mineral precipitation, and prepared close to serpentine saturation using $MgCl_2$. Starting materials were analyzed using an electron microprobe, and post-experimental grains were characterized using SEM and EDS. For grains <53 μm, experiments run at 20°C and 80°C show a rapid increase in ^{28}Si isotopes early in the experiments indicating rapid dissolution of ultrafine particles, followed by a drop in both silicon isotopes, indicating precipitation. With grains 53 μm-75 μm, silicon isotope behavior indicates a slower reaction. The activity diagram for these experiments indicates that the final solutions are saturated in serpentine and talc, and EDS analysis indicates the presence of both serpentine and brucite precipitates on the smaller grains. Calculated reaction rates closely match with extrapolated rates from previous literature. For grains <106 μm run at 80°C, EDS analysis indicates the presence of a brucite passivation layer, with no other precipitates detected, while solution chemistry indicates serpentine saturation.