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Dissolved SiO_2 Controls on H_2 Generation During Serpentinization

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The serpentinization of ultramafic rocks produces significant quantities of molecular hydrogen (H_2), a critical energy source for the subsurface biosphere and a potential carbon-free energy resource. H_2 generation during serpentinization of ultramafic rocks is highly dependent on fluid chemistry. Thermodynamic modeling predicts that dissolved silica, $SiO_2(aq)$, could exert a fundamental control on the formation of secondary mineralogy, Fe-bearing serpentine ± brucite ± magnetite, and the degree of oxidation of Fe^{2+} derived from the alteration of primary silicate minerals. In olivine-dominated lithologies, low $SiO_2(aq)$ activity promotes the formation of Fe^{2+}-brucite, which has the potential to suppress H_2 production. Conversely, higher $SiO_2(aq)$ activities, typical of orthopyroxene-bearing lithologies, are expected to suppress brucite formation while allowing $Fe^{2+,3+}$-serpentine and magnetite to form, potentially enhancing overall H_2 production.

We examined the control of $SiO_2(aq)$ activity on H_2 generation during the serpentinization of olivine by performing experiments at 200°C, 125 bar under flow-through conditions in passivated titanium reactors. The experiments employed high initial $pH_{25°C}$ (~10), $^{29}SiO_2(aq)$-bearing tracer solutions that vary in concentration, 10, 50, and 500 µmol/kg, to shift fluid chemistry increasingly further away from brucite-serpentine equilibria to prevent the formation of Fe^{2+}-bearing brucite and to promote the formation of $Fe^{2+,3+}$-bearing serpentine + magnetite. These experiments demonstrate that changes in $SiO_2(aq)$ activity during the serpentinization of olivine has a direct control on secondary mineral formation and H_2 generation rates. The lowest rates of H_2 generation occur under low $SiO_2(aq)$ activities, with solution chemistry representative of serpentine-brucite equilibrium, whereas the rate of H_2 generation increases directly with increasing $SiO_2(aq)$ activity further into the serpentine-only stability field.

Mineralogical analyses of the experimental products indicate that serpentine is the primary secondary mineral phase to form with magnetite present. Fluid chemistry and gas compositions partly constrain the reaction equilibria, and mineral alteration products are currently being analyzed by isotopic, magnetic, microscopy, and thermogravimetric methods to quantify the rate and degree of olivine alteration, the fate and oxidation state of Fe during secondary mineral formation, and coupled H_2 generation. Ultimately, these data will enable precise quantification of the kinetics of olivine alteration and secondary mineral precipitation coupled with H_2 generation rates.