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Examining the kinetics of Fe-brucite silicification and hydrogen generation during “re-serpentinization” reactions

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Serpentinization is a significant planetary process that has substantially impacted the biogeochemical evolution of both Earth and Mars. A hallmark of serpentinization is the production of molecular hydrogen (H_2), which serves as a basal energy source for life, supports chemolithoautotrophy in serpentinization-driven settings, and controls the geochemical evolution of planetary atmospheres. During serpentinization of olivine-rich rocks, partitioning of ferrous iron into brucite and serpentine limits the extent of hydrogen generation. However, later reaction of these minerals, especially highly reactive brucite, with SiO_2-rich fluids (i.e., silicification), can trigger “re-serpentinization” reactions and convert primary Fe(II)-bearing serpentine or brucite into secondary minerals such as magnetite and Fe(III)-bearing serpentine, both of which can yield significant H_2. These “re-serpentinization” reactions can help to explain the continued generation of H_2 in long-lived serpentinizing systems, such as Atlantis Massif and the Samail Ophiolite, where much of the primary olivine has already reacted to secondary minerals. Importantly, because “re-serpentinization” reactions do not require olivine, their apparent role in low-temperature H_2 production necessitates a reconsideration of our conceptual models of H_2 production in low temperature serpentinizing systems. However, the reactivity of ferroan brucite as a function of important variables, such as temperature and activity of aqueous SiO_2, remain poorly constrained. Here, we report the results of a series of flow-through experiments in which we reacted synthetic Fe-rich brucites (Mg#: 0.8, 0.5, and 0.2) with SiO_2-rich solutions under anoxic conditions at temperatures from ambient to 80°C. Throughout all experiments, ferroan brucite demonstrates exceptional reactivity, suggesting that “re-serpentinization” reactions can indeed produce significant H_2 at low temperatures. These results have far-reaching implications for the generation of H_2, the bioavailability of iron, as well as the fidelity of the geochemical signals that are recorded in serpentinized rocks. Our experimental results, therefore, demonstrate that it is essential to carefully consider the role of “re-serpentinization” reactions in regulating the production of H_2 in low temperature serpentinizing systems.