and Water-Saturated Supercritical CO₂ Conditions

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There is a growing demand for a solution to remove CO₂ from the atmosphere. Capturing carbon and storing it by injecting it into reactive rocks (e.g., basalt and peridotite) is gaining increased interest. In this process, the reaction of aqueous CO₂ with these crustal minerals results in the leaching of divalent cations and the subsequent precipitation of carbonate minerals. This approach offers a reduced likelihood of CO₂ leakage compared to alternative approaches that rely on the physical trapping of CO₂ in unreactive, high-porosity rock formations. While the crustal injection of CO₂-saturated water is routinely performed by the Carbfix program in Hellisheiði, Iceland and has successfully demonstrated quantitative carbonation, this process utilizes large quantities of water [1]. To address this concern, the injection of water-saturated supercritical CO₂ ("wet" scCO₂) directly into basaltic basement rocks may offer advantages such as lower water demands and the increased conversion of CO₂ into carbonate minerals; however, the reactivity of minerals upon exposure to scCO₂ is not well understood [2]. To assess the mineralization potential of utilizing wet scCO₂, we conducted a series of experiments at different temperatures comparing the rate of carbonation of basalt cores and ~100 micron-sized olivine grains in CO₂-saturated water and wet scCO₂. All experiments exhibited varying degrees of carbonation after three weeks of reaction at 90 bar CO₂, with carbonation occurring faster at higher temperatures in both the wet-scCO₂ and CO₂-saturated water experiments. Scanning electron microscopy (SEM) and energy-dispersive X-ray spectroscopy (EDS) confirmed the precipitation of magnesite on the sample surfaces. Microcomputed tomography was employed to examine the extent of carbonation within the interiors of the basalt cores, and thermogravimetric analysis was used to quantify the rates of olivine grain carbonation. The carbonation rates were observed to be slower than olivine dissolution rates, suggesting surface passivation is a potentially rate-limiting step in these processes. We will discuss how our data and observations may translate into sustainable solutions toward the mitigation of elevated atmospheric CO_2 .

[1] Gislason et al. (2010). International Journal of Greenhouse Gas Control, 4, 537–545

[2] Abdolhosseini Qomi et al. (2022). Nature Reviews Chemistry, 6, 598-613.