



## **Experimental carbonation of peridotite by percolation of CO<sub>2</sub>-rich fluids at 160°C and 120 bars**

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CO<sub>2</sub>sequestration by direct injection into geological formation is a highly regarded option for reducing greenhouse gas emissions. Carbonation of ultramafic rocks is the most efficient reaction to trap CO<sub>2</sub> into stable carbonate phases. Thus, widespread ultramafic bodies represent an immense storage capacity by long-term hydrothermal processes (slow-spreading ridges) and by industrial injection of CO<sub>2</sub>-enriched fluids. A critical issue for durability of both processes is the prediction of consequences of fluid circulation/injection on rock properties. Yet, fundamental parameters characterizing *in situ* reactions of CO<sub>2</sub>-enriched fluids in ultramafic rocks and their consequences on hydrodynamic properties are lacking.

We investigate these processes by scaled rock sample percolation experiments. We present results of experiments performed using sintered grounded dunite (96.5% olivine, 2% diopside, 1.5% spinel) flooded at constant specific discharge (0.6 ml/min) with CO<sub>2</sub>-enriched fluid up to PCO<sub>2</sub> = 95 bars, buffered with 0.6 M NaHCO<sub>3</sub> and under a confined pressure of 120 bars and a temperature of 160°C.

Permeability stabilizes after an increase period of 1h. The chemical composition of the outlet fluid is dominated by Si and is depleted in Mg relative to stoichiometric dissolution of olivine during the whole experiment. SEM and AEM/TEM are used to characterize the reactive interfaces and the neoformed materials. A porous and poorly crystallized material, enriched in Si and Fe and depleted in Mg, is observed around diopside and olivine grains near the sample inlet, i.e. where dissolution dominates. Near the sample centre, carbonate precipitation is observed. Siderite formation occurs

as aligned nanograins at the olivine dissolution front while Ca-magnesite precipitates locally as larger grains on olivine surface, preferentially in the vicinity of diopside. Thus, under those conditions, both olivine dissolution and carbonate precipitation occur at the sample scale. Mass balance calculations indicate that 60 mg of magnesite is formed (0.02 cm<sup>3</sup>) for 85 mg of altered rock (0.025 cm<sup>3</sup>). It is worth noting that the volumes of dissolved and crystallized minerals are similar. This can explain the constant permeability measured during the experiment, independently on the fluid-rock mass transfers. Olivine and carbonates are separated by a porous interface made of a poorly crystallized Si-rich gel whose composition varies between talc and serpentine stoichiometry. This porous reactive interface promotes fluid renewal at the olivine surface, and allows reaction to persist despite the carbonate formation. Long-lasting experiments are presently run to test the limit of this process.