

Carbonate mineralization in percolated olivine aggregates: Linking effects of crystallographic orientation and fluid flow

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ABSTRACT

In situ mineralization of CO₂ in ultramafic rock-hosted aquifers is one of the promising solutions for decreasing CO₂ concentrations in the atmosphere. Naturally altered ultramafic rocks suggest that carbonation processes are controlled by local heterogeneities in the structure of the rock and fluid transport at the water-rock interfaces. We studied the role of rock crystallographic anisotropy relative to the global fluid flow direction on the mineralization of CO₂ by means of electron microscope analyses from the macro- to the micrometer scale (EBSD-FIB). The sample used for the measurements was a hot pressed olivine core percolated by water enriched in CO₂ (pCO₂ = 10 MPa) at 180 °C. During the percolation experiment, olivine was dissolved and two types of carbonates, dolomite, and magnesite, were precipitated on olivine surfaces. The results showed that the dissolution of olivine is controlled by its crystallographic properties as shown by the development of etch-pits only on the (010)_{ol} planes and with elongated shapes parallel to the [010]_{ol} axes. In contrast, the precipitation of carbonates is governed by hydrodynamic properties. Carbonates are heterogeneously distributed in the percolated rock. They are mainly located along the moderate (for dolomite) and the minor (for magnesite) flow paths, both oriented parallel to the principal fluid flow direction, which allow carbonates to be supplied with divalent cations (e.g., Ca²⁺, Mg²⁺, and Fe²⁺). In these flow paths, carbonate growth is systematically oriented normal to the flow that facilitates the development of chemical gradients with cationic supersaturation conditions for carbonate precipitation near the walls.

In natural systems, the (010)_{ol} planes are parallel to the Moho and the (100)_{ol} planes are vertical; our study suggests that flow of CO₂-rich fluids will induce precipitation of carbonates localized along, and preferentially clogging, vertical flow paths while favoring olivine dissolution along horizontal fluid pathways. This dual control of structure and fluid flow on carbonation mechanisms could be an important parameter allowing sustainable CO₂ storage in peridotites, while limiting the risks of leakage toward the surface.

Keywords: EBSD-FIB, CO₂ mineralization, ultramafic rocks, etch-pits, oriented growth, crystallographic relationships.