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 carbonate 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 microprobe 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)₆ planes and with elongated shapes parallel to the [010]₆ 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 Fe³⁺). 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)₆ planes are parallel to the Moho and the (100)₆ 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 carbonate 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.

INTRODUCTION

In situ carbonation of ultramafic rocks is potentially an effective and safe means for mitigating atmospheric carbon dioxide (CO₂) concentrations (Bachu et al. 1994; Goff and Lackner 1998; Jun et al. 2013; Matter and Kelemen 2009; Oelkers et al. 2008; Seifritz 1990). Carbonation (or CO₂ mineralization) consists in transforming CO₂ into stable carbonates through a suite of chemical fluid-rock reactions; it is a process commonly observed in natural ophiolitic (for example, Kelemen and Matter 2008; Kelemen et al. 2011) and oceanic environments (for example, Alt and Teagle 1999; Bach et al. 2004; Kelley et al. 2005; Ludwig et al. 2006). These chemical reactions are coupled to the transport of reactants and products of reactions at the fluid-mineral interface: first, CO₂ reacts with water to produce H⁺ and bicarbonates/carbonates ions (HCO₃⁻/CO₃²⁻); the H⁺ ions then trigger the dissolution of silicate minerals and bicarbonates/ carbonates ions combine with the divalent cations provided by silicate dissolution and/or fluids precipitate carbonates (Kelemen et al. 2011). Ultramafic rocks are widely distributed at the Earth’s surface (Bodinier and Godard 2003; Deschamps et al. 2013); they also have the highest potential to mineralize CO₂ due to their high proportion of divalent cations (40–50 wt% MgO ± CaO ± FeO) and their high reactivity (Matter and Kelemen 2009; Oelkers et al. 2008; Xu et al. 2004). Thus they could represent one of the natural sinks for atmospheric CO₂ (Alt and Teagle 1999; Früh-Green et al. 2004). They are also a possible target for industrial CO₂ geological storage (Kelemen and Matter 2008; Matter and Kelemen 2009), provided the development of methods allowing to overcome the mechanisms limiting carbonation kinetics [e.g., formation of silica layer on dissolution olivine grains (Johnson et al. 2014; Sissmann et al. 2013)]; this requires a better understanding of the mechanisms driving carbonate reactions in ultramafic systems.

Laboratory experiments and natural observations of altered peridotites indicate that carbonate reactions are controlled by fluid flow and also by the structure of the reacting rocks (e.g., Andreani et al. 2009; Kelemen and Matter 2008). Kelemen and Matter (2008) reported a geometric distribution of carbonate veins in altered ophiolite outcrops (Samail ophiolite, Sultan-