

Mineralogical evolution of Fe–Si-rich layers at the olivine-water interface during carbonation reactions

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ABSTRACT

Recent studies investigating carbonation of iron-bearing silicates have shown that the rates of these reactions, although formally not depending on oxygen fugacity, are strongly different at different redox states of the system (Saldi et al. 2013; Sissmann et al. 2013). Here we provide a micro- and nanostructural characterization of the olivine/water interface during the carbonation of forsteritic olivine at 150 °C and $p_{\text{CO}_2} = 100$ bar. When the reaction starts under oxic conditions, the observed temporal sequence of interfacial layers consists of: a hematite/SiO_{2(am)} assemblage, Fe-rich phyllosilicates with mixed Fe valence and a non-passivating Fe-free amorphous SiO₂ layer, which allows the formation of ferroan magnesite. In contrast, starting at micro-oxic conditions, carbonation rates are much faster, with no real evidence of interfacial layers. Separate deposits of goethite/lepidocrocite in the early stages of the reaction and then formation of magnetite are observed at these conditions, while precipitation of siderite/magnesite proceeds unhindered. The evolution of the redox conditions during the reaction progress controls the sequence of the observed reaction products and the passivating properties of Fe–Si-rich interfacial layers. These findings have important implications for modeling the carbonation of ultramafic rocks under different oxygen fugacity conditions as well as for understanding the technological implications of adding accessory gases to CO₂ in carbon capture and storage mineralization processes involving ultrabasic rocks.

Keywords: Olivine carbonation, Fe–Si-rich interfacial layers, redox reactions, passivation, cronstedtite, dissolution/precipitation, Fe-oxides