

X-Ray microtomography analysis of the impact of pCO₂ on serpentinization reactions: A reactive percolation experimental approach

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Serpentinization is the main hydrothermal process driving the alteration of the mantle lithosphere by seawater at ridges. It consists in the alteration of olivine to serpentine and is associated to processes such as oxidation as well as carbonation when CO₂ is present. The sustainability and efficiency of the reaction requires penetration and renewal of fluids at the mineral-fluid interface. Yet the secondary low density minerals can fill the porous network, clogging flow paths efficiently. This study aims at better understanding the coupled hydrodynamic and chemical processes driving the earliest stages of alteration of the ultramafic basement, when seawater-derived hydrothermal fluids penetrate and interact with exposed mantle rocks at slow spreading ridges.

We investigate the structural changes of the rock in relation to dissolution-precipitation reactions triggered by the injection CO₂-rich seawater using an experimental approach.

The experiments simulate open conditions and were performed using the reactive percolation bench ICARE Lab 3 – Géosciences Montpellier. ICARE 3 allows to continuously measuring permeability changes during experiments and sampling the outlet fluids passing through the sample. We analysed the reacted samples before and after the experiments using a combination of geochemical (TGA-MS) and high resolution X-Ray microtomography (ESRF ID19 synchrotron beamline, Grenoble) approaches.

A series of experiments was carried out at 190°C and 25 MPa. CO₂ enriched natural seawater (XCO₂ 5.24 mmol/kg) was injected into Titanium capsules (2 mm diameter, 6 mm length) filled by pressed powdered San Carlos olivine (Fo90; grains 150-200 μm). The outlet section of the samples were analysed at 0.65 μm resolution using microtomography before and after the experiments. The reacted powdered sample was analysed by TGA-MS.

Comparison of microtomography images of reacted and unreacted samples shows evidences of olivine dissolution and secondary minerals precipitation during the 14 days long experiments. A new method based on image registration enables to identify the dissolution mainly localized at grain borders and the precipitation localizes in fractures and at grain borders. Dissolution appears to be dominant in the outlet section of the sample. The equilibrium of the reaction will be determined by the comparison of thermogravimetry (TGA-MS) analysis of the reacted sample after the experiment and thermodynamic modelling of the experiment in equilibrium.