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Micro-scale investigation of carbonation process in partially serpentinized peridotites

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The carbonation of ultramafic rocks is, theoretically, the most efficient reaction to trap CO₂ irreversibly in the form of solid carbonates, as predicted by equilibrium thermodynamic calculations. However, the success of industrial or natural carbonation in large ultramafic aquifers or oceanic ultramafic exposures does not only rely on the thermodynamic conditions of chemical reactions, but also on their feedback effects on the reactive surface area and on the local porosity and permeability. In addition, side processes like serpentinization, redox reactions, abiotic catalytic effects, and biological activity, can be expected in such complex natural system. Their occurrence and implications on carbon speciation and carbon transfers during hydrothermal alteration of oceanic peridotites have not been explored yet and requires detailed study of natural and/or experimental carbonation zones.

We have combined petrographic and electron microscopy with SIMS, Raman and FTIR microspectroscopy on partially serpentinized peridotites drilled during the IODP leg 304 (30°N, MAR) in order to characterize the mechanisms of peridotite carbonation at the fluid-mineral interface and identify the associated speciation of carbon (inorganic and organic carbon occurrences). We present first results on zones located close to talc-tremolite sheared veins in holes 1309B and D. Associations of carbonates, porous phyllosilicates and oxides are observed in close vicinity of relict olivines that underwent a previous stage of serpentinization. The olivine-carbonate interface is nanoporous which facilitates mass transfer between fluid and mineral. The phyllosilicate identified as saponite results from the metasomatic replacement, during the carbonation stage, of previously formed serpentine. These observations do not favour reaction-induced cracking but rather a transfer-controlled process in an open system. Among the submicrometric dark clusters widely-distributed in saponite and in serpentine, vibrational microspectroscopy reveals the presence of various types of organic compounds that tend to be located close to micrometric sulphides grains.

Those results underline the microscale variability of carbon speciation within hydrothermally altered peridotites. The association of reduced carbon phases with the carbonation texture suggests that CO₂ conversion may not be limited to solid carbonate formation in natural systems and that biological activity and/or abiotic CO₂ reduction, possibly catalyzed by Ni-rich sulphides, can occur contemporaneously. This complex association of reactions has to be unravelled further to determine the respective contribution of abiotic versus biological processes and integrate them in carbon transfers modelling through the oceanic lithosphere.