



Mineral replacements during carbonation of peridotite: implications for carbon dioxide sequestration in ultramafic rocks

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In contact with CO₂, ultramafic rocks are known to be reactive and eventually form ophicarbonates and listwaenites. Here we present observations from serpentinized peridotite clasts from the Solund Devonian Basin, SW Norway. These clasts show evidence for a stepwise reaction history starting with initial serpentinization and resulting in the formation of carbonates (mainly calcite and dolomite) and quartz. Thus, they represent a natural analogue for CO₂ sequestration in ultramafic rocks, which was proposed by the Inter Governmental Panel on Climate Change (IPCC 2005) as one possibility for long-term CO₂ storage. In several layers of the basin, the carbonatized ultramafic clasts are important constituents and account for up to 20 vol. % of the basin infill.

The investigated clasts show a concentric build-up with green to grey colored cores surrounded by mm to 10 cm thick zones of red to black shades. Textural evidence indicates the following alteration sequence: An early stage is represented by serpentinization of peridotite resulting in a typical mesh texture, with veins of serpentine and Ni-rich hematite surrounding compartments of relict olivine (Fo₉₀). Subsequently, relict olivine breaks down to form an alteration product which is significantly depleted in Mg relative to the precursor olivine. In the more advanced ophicarbonate stage, compartments are filled with calcite, quartz, and talc. In the most advanced stage, quartz, calcite, and hematite dominate and occur together with minor amounts of chromite, talc, and chlorite. The textural evolution is accompanied by a decrease in whole-rock MgO from 40 to 2 wt. % and a CaO increase from 1 to 35 wt. %. All clasts are characterized by high Cr and Ni (1000-4000 and 500-3000 ppm, respectively) revealing their ultramafic origin. Transmission electron microscopy (TEM) observations indicate that the alteration product after olivine is composed of an amorphous material, which is compositionally close to serpentine, together with poorly crystalline serpentine and extremely fine grained talc. Hydrothermal batch experiments (130-160 bar PCO₂; 200°C; 1-3 weeks reaction time) show that the alteration product after olivine is the favorable site of reaction presumably due to the large reactive surface area. In contrast, the olivine relicts have reacted to a significantly lesser extend, whereas the serpentine veins remain virtually unreacted. The dissolution of the compartment fillings is followed by nucleation and growth of calcite crystals also revealing that precipitation of calcite is strongly favored over magnesite as soon as the system contains Ca. The preferred precipitation of calcite is also supported by geochemical modeling (using Phreeqc), which shows that the Mg-bearing carbonates (dolomite, magnesite) only form if the fluid is sufficiently depleted in Ca. The compositional and textural differences between different samples as well as different run products from experiments indicate that the described clasts evolved from peridotite due to extreme mobilization of Mg, development of secondary porosity, and infill of carbonates. Mg removed from the clasts is partly consumed by replacement reactions in the vicinity of the clasts where Fe-minerals (almandine) are altered to Mg-minerals (talc). For basins containing abundant peridotite clasts, the outlined process will influence the CO₂ and MgO budget.

References:

IPCC Special report: Carbon Dioxide Capture and Storage, Summary for Policymakers, 2005.