



Multiscale solute transfer and porosity evolution during pervasive replacement of serpentine by carbonate and quartz - insights from the Oman ophiolite

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Pervasive carbonation of serpentinized peridotite to carbonate-quartz rock (listvenite) due to infiltration of CO₂-bearing fluid is a remarkable process because it can be geologically fast and it increases the rock's carbon content from initially zero to > 30 wt% CO₂. This pervasive conversion is related to an overall solid volume increase while at the same time requiring high time-integrated fluid rock ratios with permeability and diffusivity on all scales. Thus, porosity has to be created dynamically during reaction progress as otherwise fluid pathways become clogged by the reaction products carbonate and quartz, which is one of the major obstacles for artificial carbon storage by peridotite carbonation. Processes that can renew porosity and permeability during carbonation are fracturing and veining – in response to tectonic stress [1] or induced by reaction and crystal growth [2], or a combination of both –, reaction-enhanced ductile deformation [3,4], and spatial decoupling of dissolution and precipitation with solute transfer at different length scales.

Using SEM, EBSD, TEM and FIB nano-tomography, we investigated the microstructural record of local solute transfer and its role for porosity renewal in natural carbonate-bearing serpentinites, transitional serpentine-carbonate-quartz assemblages and listvenites from the Samail Ophiolite, Oman. The clearest indicators of pervasive replacement accommodated by local solute transfer are pseudomorphic replacement structures where carbonate and quartz occur in distinct microstructures corresponding to different inherited peridotite and serpentinite textures. A common pseudomorphic replacement structure in the Samail carbonated peridotite are quartz-fuchsite intergrowths replacing bastite (pseudomorphs after orthopyroxene) in listvenite with harzburgite protoliths. A local strong crystallographic alignment of quartz in each of the bastite/pyroxene pseudomorphs suggests that the anisotropic porosity structure of bastite serpentine favored oriented, epitaxial growth of quartz. Transitional serpentine-carbonate-quartz assemblages show that the first quartz and carbonate generations precipitate coeval but spatially separated, with distinct crystal habits. FIB nano-tomography, STEM analysis and high resolution SEM on ion-polished samples of a transitional serpentine-dolomite-quartz rock from the carbonation reaction front show nano-porous fluid channels in fibrous serpentine at high angle to a highly serrated carbonate-serpentine replacement contact. These nano-scale fluid channels

facilitated bidirectional mass exchange of Ca, C, Mg and Si bearing solutes between sites of preferential replacement, such as the cores of serpentine mesh textures, and larger-scale permeability networks along veins and fractures. These observations imply that massive and pervasive solute transfer through the reacting serpentine matrix is possible on a small scale, without clogging of porosity by immediate co-precipitation of quartz or Mg-silicates. Spatial decoupling of dissolution and precipitation was likely caused by the dynamically evolving composition of the reacting fluid and/or due to the influence of differential stress and volumetric strain – a mechanism that can compensate on a local scale for the volume expansion expected of isochemical carbonation reactions.

[1] Menzel et al., *Solid Earth*, 2022; <https://doi.org/10.5194/se-2021-152>

[2] Kelemen & Hirth, *EPSL*, 2012; <https://doi.org/10.1016/j.epsl.2012.06.018>

[3] Menzel et al., *Nature Communications*, 2022; <https://doi.org/10.1038/s41467-022-31049-1>

[4] Kelemen et al., *JGR*, 2022; <https://doi.org/10.1029/2021JB022352>

Funding: Junta de Andalucía (Postdoc_21_00791); DFG grants UR 64/20-1, UR 64/17-1; and EU Horizon 2020 Transnational Access EXCITE_C1_2022_34.