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Volumetric response of crushed dunite during carbonation reaction under controlled σ -P-T conditions

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Naturally, olivine reacts with CO₂-rich fluids, producing carbonates and silica. If in completion, this reaction will cause a large increase in the solid volume (~85%), which can generate a significant stress/force when it occurs in a confined space. This may be used to fracture the surrounding rocks in the context of the injection of industrially captured CO₂ into peridotites for permanent sequestration. Contrarily, this volume-increasing reaction may also clog transport paths and thus inhibit CO₂ access, leading to little or no volumetric increase at industrial time scales. Although observations from natural systems suggest that reaction-induced fracturing during peridotite carbonation can occur, the fracturing mechanism has not been experimentally reproduced under in-situ stress-temperature-chemical conditions. Here, we report 9 flow-through experiments performed on pre-compacted Åheim dunite (containing ~85% olivine) powders (grain size 36-50 μm) during carbonation reaction under controlled σ -P-T conditions. This was done using a purpose-built apparatus, consisting of a flow-through system accommodated with a uniaxial servo-controlled loading system. Before experiments, the dunite powders were compacted stepwise up to 250MPa to form a disc-shape sample with starting porosity of ~25%. The sample was covered by a thin Teflon sleeve plus Vaseline to reduce the friction against the vessel wall. The experiments were performed at a constant temperature of 150°C and constant (Terzaghi) effective stress of 1, 5, 15MPa, respectively. The sample was first exposed to deionized (DI) water at a pore fluid pressure of 10MPa, and then the DI water was replaced, maintaining constant pore pressure of 10MPa, by flow-through of a certain chemical fluid, such as CO₂ saturated brine (containing 1M NaCl plus 0.64M NaHCO₃, pH~3), CO₂ saturated water (pH~3), NaHCO₃ saturated solution (pH~9) and NaHSO₄ solution (pH~3). The permeability was measured for all experiments using the flow-through system by means of the steady-state method, and each experiment took 2-4 weeks. The experiments show that the samples exhibited 0-0.37% compaction strain when CO₂ saturated brine, CO₂ saturated water, and NaHCO₃ saturated solution flow through, independently of poroelastic effects, and the sample permeability drops in the order from 10⁻¹⁷ to 10⁻²⁰ m². By contrast, for the NaHSO₄ flow-through experiment where no carbonation reaction occurred, the sample permeability increased from 2*10⁻¹⁷ to 7*10⁻¹⁷ m², associated with 0.05% compaction. The sample mass after the NaHSO₄ flow-through experiment reduced ~5%, suggesting that magnesium and silica may be partly leached out from the sample. Microstructure observations

and XRD analysis on these samples demonstrate a drastic reduction in porosity of the reaction zone where CO₂ was integrated into the crystal structure of the product carbonates by means of carbonation reactions. The mechanism responsible for the observed behavior seems to be that the dissolution of olivine that occurred first at the grain contact surface leads to compaction, followed by precipitation of carbonates at porous that clogs the transport paths and thus reduces the permeability, though the detailed chemical analysis is still performing. As a result, our current findings suggest that the volume-increasing precipitation produced via the carbonation reaction under in-situ subsurface conditions will clog transport paths.