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Impact of CO_2 injection protocol on fluid-solid reactivity: high-pressure and temperature microfluidic experiments in limestone

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Geological sequestration of CO_2 has been proposed in the last decades as a technology to reduce greenhouse gas emissions to the atmosphere and mitigate the global climate change. However, some questions such as the impact of the protocol of CO_2 injection on the fluid-solid reactivity remain open. In our experiments, two different protocols of injection are compared at the same conditions (8.4 MPa and 45 C, and constant flow rate 0.06 ml/min): i) single phase injection, i.e. CO_2-saturated brine; and ii) simultaneous injection of CO_2-saturated brine and scCO_2. For that purpose, we combine a unique high-pressure/temperature microfluidics experimental system, which allows reproducing geological reservoir conditions in geo-material substrates (i.e. limestone, Cisco Formation, Texas, US) and high resolution optical profilometry. Single and multiphase flow through etched fracture networks were optically recorded with a microscope, while processes of dissolution-precipitation in the etched channels were quantified by comparison of the initial and final topology of the limestone micromodels. Changes in hydraulic conductivity were quantified from pressure difference along the micromodel. The simultaneous injection of CO_2-saturated brine and scCO_2, reduced the brine-limestone contact area and also created a highly heterogeneous velocity field (i.e. low velocities regions or stagnation zones, and high velocity regions or preferential paths), reducing rock dissolution and enhancing calcite precipitation. The results illustrate the contrasting effects of single and multiphase flow on chemical reactivity and suggest that multiphase flow by isolating parts of the flow system can enhance CO_2 mineralization.