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Laboratory-scale interaction between CO_2-rich brine and limestone and sandstone under supercritical CO_2 conditions

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A test site for a prospective CO_2 geological storage is situated in Hontomín (Burgos, northern Spain) with a reservoir rock that is composed of limestone (calcite) and sandstone (66 wt.% calcite, 28 wt.% quartz and 6 wt.% microcline). During and after CO_2 injection, the resulting CO_2-rich acid brine will likely promote the dissolution of carbonate minerals (calcite) and aluminosilicates (microcline). Since the reservoir Hontomín brine contains sulfate, gypsum (or anhydrite at depth) may precipitate. These coupled dissolution and precipitation reactions may induce changes in porosity and pore structure of the repository rocks. Percolations experiments with mechanically fractured cores (8.6 mm in diameter and 18 mm length) were performed under CO_2 supercritical conditions (Pfluid = 150 bar; $pCO_2 \approx 90$ bar and T = 60 ºC) in order to evaluate and quantify variations in fracture permeability, preferential path formation and fracture volume. The brine sulfate content and the flow rate were varied.

Regarding limestone, as the synthetic brines circulated through the fracture, the fracture permeability initially increased slowly, to thereafter increase rapidly. This change was due to a localized dissolution process (wormhole formation) along the core that occurred regardless gypsum precipitation. Nonetheless, the originated fracture volume in the sulfate-rich brine experiments was a factor of two smaller than that in sulfate-free brine experiments. Also, an increase in flow rate from 0.2 to 60 mL/h increased the volume of both dissolved calcite and precipitated gypsum. Regarding sandstone, permeability increased gradually with time. Nonetheless, this increase was not always continuous due to eventual fracture clogging. Formation of wormholes was observed.

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