

The role of mineral heterogeneity on the hydrogeochemical response of two fractured reservoir rocks in contact with dissolved CO₂

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In this study we compare the hydrogeochemical response of two fractured reservoir rocks (limestone composed of 100 wt.% calcite and sandstone composed of 66 wt.% calcite, 28 wt.% quartz and 6 wt.% microcline) in contact with CO₂-rich sulfate solutions. Flow-through percolation experiments were performed using artificially fractured limestone and sandstone cores and injecting a CO₂-rich sulfate solution under a constant volumetric flow rate (from 0.2 to 60 mL/h) at P = 150 bar and T = 60 °C. Measurements of the pressure difference between the inlet and the outlet of the samples and of the aqueous chemistry enabled the determination of fracture permeability changes and net reaction rates. Additionally, X-ray computed microtomography (XCMT) was used to characterize and localized changes in fracture volume induced by dissolution and precipitation reactions.

In all reacted cores an increase in fracture permeability and in fracture volume was always produced even when gypsum precipitation happened. The presence of inert silicate grains in sandstone samples favored the occurrence of largely distributed dissolution structures in contrast to localized dissolution in limestone samples. This phenomenon promoted greater dissolution and smaller precipitation in sandstone than in limestone experiments. As a result, in sandstone reservoirs, the larger increase in fracture volume as well as the more extended distribution of the created volume would favor the CO₂ storage capacity. The different distribution of created volume between limestone and sandstone experiments led to a different variation in fracture permeability. The progressive stepped permeability increase for sandstone would be preferred to the sharp permeability increase for limestone to minimize risks related to CO₂ injection, favor capillary trapping and reduce energetic storage costs.

2D reactive transport simulations that reproduce the variation in aqueous chemistry and the fracture geometry (dissolution pattern) were performed using CrunchFlow. The calcite reactive surface area had to be diminished with respect to the geometric surface area in order to account for the transport control of the calcite dissolution reaction at pH < 5. The fitted reactive surface area was higher under faster flow conditions, reflecting a decrease in transport control and a more distributed reaction in sandstone compared to limestone.