Geochemical Processes in the Interaction Between a Marly Shale and a CO_2-Rich Sulfate Solution Under Supercritical CO_2 Conditions

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Abstract

Geological CO2 sequestration at a pilot-plant scale is going to be performed at Hontomín (Spain). The Hontomín caprock is made up of marly shale, with an average composition (wt. %) of 71.2% calcite, 9.7% quartz, 7.1% illite, 6.5% albite, 2.8% clinochlore and trace amounts of gypsum, anhydrite and pyrite. The reservoir solution is rich in NaCl and sulfate (I = 0.6 M), being in equilibrium with respect to calcite and gypsum. The present study aims at evaluating the interaction between the shale and the CO2-rich solution of Hontomín under in situ conditions (pTotal = 150 bar, pCO2 = 61 bar and T = 60 °C). In particular, we focused on (i) the role of injected solution composition (S-free and S-rich solutions), (ii) the importance of mineral dissolution and precipitation processes and (iii) the effect of flow rate (0.2, 1 and 60 mL min-1) on the rock hydrodynamic properties. Under the experimental conditions, in both types of solution, dissolution of calcite, clinochlore and albite and precipitation of clays occurred, whereas precipitation of gypsum only took place in S-rich solution. In the S-free solution experiments, face and uniform dissolution was observed at low and high flow rate, respectively, but fracture permeability remained fairly constant independently of the flow rate. In the S-rich solution experiments, the fracture permeability decreased remarkably at the lower flow rates ($k_{final}/k_{initial} \leq 0.03$) due to precipitation of gypsum and clay minerals. However, at the highest flow rate, uniform dissolution occurred and the fracture permeability increased slightly ($k_{final}/k_{initial} = 4$) as dissolution of calcite predominated over gypsum precipitation.

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