Hydrodynamic and geochemical evolution of artificially fractured marl cores under supercritical CO_2 conditions

GABRIELA DÁVILA[123], LINDA LUQUOT[12], JOSEP M. SOLER[12] AND JORDI CAMA[12]

[1] Institute of Environmental Assessment and Water Research (IDAEA), CSIC, Jordi Girona 18-26, 08034 Barcelona, Catalonia, Spain.
[2] Associated Unit: Hydrogeology Group (UPC-CSIC).
[3] Department of Geotechnical Engineering and Geosciences, Universitat Politècnica de Catalunya (UPC), Jordi Girona 1-3, 08034 Barcelona, Catalonia, Spain.

The Hontomín (Spain) caprock is mainly composed of marl (70 wt.% of calcite). The aim of the present work is to study the alteration of this rock in contact with CO_2-rich solutions under supercritical CO_2 conditions (P_{Total} = 150 bar, pCO_2 = 61 bar and T = 60 °C) to assess its sealing capacity. Given the low permeability of the marl (k < 10^{-18} m^2), water-caprock interaction may take place along preferential pathways (e.g. fractures or microcracks).

Flow-through percolation experiments were performed using artificially fractured cores to elucidate (*i*) the role of the composition of the injected solutions (*S-free* and *S-rich* solutions) and (*ii*) the effect of the flow rate on fracture permeability. In addition, 2D reactive transport models have been used to interpret the results.

Results and discussion

Major dissolution of calcite (*S-free* and *S-rich* solutions) and precipitation of gypsum (*S-rich* solution), together with minor dissolution of the silicate minerals, contributed to the formation of an altered skeleton-like zone (mainly made up of unreacted clays) next to the fracture wall. Fracture permeability did not significantly change in the *S-free* experiments. Permeability decreased due to gypsum precipitation in the experiments performed under slow flow rates (0.2 and 1 mL h^{-1}) when using the *S-rich* solution.

A good match between the experimental and model variation in the outflow composition with time (except at high flow rate, 60 mL h^{-1}) needed a slight change in the initial D_{eff} values in the rock matrix (≈$1\text{-}3\times10^{-13}$ m^2 s^{-1} under slow flow rate) and in the calcite reactive surface area. Model results successfully reproduced major calcite dissolution and gypsum precipitation together with minor clinochlore dissolution. Minor precipitation of dolomite, kaolinite, boehmite and zeolites was also predicted by the model.