



# 1 Fluid-rock reactions along the CO<sub>2</sub> migration pathway in limestone reservoirs and fissured claystone caprocks

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The aim of the experimental work is to produce a data base for constraining the modelling of CO<sub>2</sub> injection and sequestration. A set of reactive percolation experiments with in-situ-like pressure and temperature conditions are presented. Experiments were designed to quantify reactions occurring (i) along the CO<sub>2</sub> path in the reservoir and (ii) in the claystone caprock under which residual CO<sub>2</sub> gas may accumulate.

(i) We report 4 percolation experiments ( $T=100^{\circ}\text{C}$ ,  $P=12\text{MPa}$ ) in limestone samples, all similar, to investigate mass transfers near the CO<sub>2</sub> injection zone where the aquifer fluid is saturated with CO<sub>2</sub> (CO<sub>2</sub> partial pressure  $P_{\text{CO}_2} = 10\text{MPa}$ ) and at increasing distances from the injection where the fluid is expected to contain progressively less CO<sub>2</sub> ( $P > P_{\text{CO}_2} > 0.7\text{MPa}$ ) and more divalent cations resulting from the rock dissolution along the fluid pathway. Results show that reactions produce high permeability channels close to the injection well, whereas precipitation inducing permeability decrease takes place far from the well. The permeability change due to dissolution can be expressed as a function of the porosity changes by the relation  $k \equiv \theta (\phi - \phi_c)^n$  where  $n$  is a function of the local Damköhler number,  $n = \alpha + \beta Da^{1.60}$ ,  $\phi_c$  is the percolation threshold porosity measured from the precipitation experiment and  $\alpha, \beta$  and  $\theta$  are fitted from the experimental results.

(ii) We realized cyclic percolation experiments of CO<sub>2</sub>-enriched fluid and CO<sub>2</sub> gas in a fractured sample of claystone. Results show that the flow of CO<sub>2</sub>-enriched fluid induces a large increase of porosity in the vicinity of the fracture due to dissolution of calcite and silica fractions, while permeability remains unchanged. Conversely, with cyclic flows of CO<sub>2</sub>-enriched fluid and CO<sub>2</sub> gas, the permeability increases after each episode of CO<sub>2</sub> gas flow. Strong concentration gradients triggered by the CO<sub>2</sub> gas lead to localized mass transfers that alter the cohesion of the clay matrix within a micron-meters thick layer. These transfer processes are controlled by the volume of rock affected by dissolution of both calcite and silica during the preceding episode of fluid flow. This progressive decrease of caprock sealing capacity is expected to persist.