



## Geochemical Model of CO<sub>2</sub> impact at the Well-Caprock-Reservoir interfaces

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The long-term safety of a CO<sub>2</sub> geological storage relies on the stability of the reservoir and on a proper assessment of CO<sub>2</sub> leakage and fluids displacement. Particular attention must be addressed to the injection wellbore as a human-made discontinuity of the geological medium, especially the caprock. Thus, the chemical and mechanical integrity of the Well-Reservoir-Caprock concrete sealing must be insured for the whole lifetime of a CCS project. These aspects are investigated following a 2-step approach within the Project INTERFACE (supported by French Research National Agency ANR through the Captage et stockage du CO<sub>2</sub> program). First, a geochemical model of the interface is developed that predicts the mechanisms of cement chemical alteration due to the interaction with the acid brine. Then, geomechanical simulations are run based on main results such as phase changes and concrete porosity evolution. Herein, the focus is on geochemical aspects solely.

The storage scenario assumes a CO<sub>2</sub> injection in carbonate formations caged by argillaceous rocks (case of the Dogger aquifer in the Paris Basin). The wellbore is cemented with an Oil Well Portland Cement (CSH, Portlandite, Katoite, Monosulfoaluminate, Calcite, Ferrihydrite) and chemically inert gravels (67% vol.), with a global porosity of 20%. Simulations have been run under the assumption of isothermal (75°C), isobaric (180 bar), and aqueous saturation conditions (SI = 1.0). Mass transfer is assumed to occur by molecular diffusion in the aqueous phase only.

Dissolution/precipitation reactions have been modelled both at equilibrium (cement minerals) and under kinetic constraints (reservoir and cap rock minerals). A sensitivity analysis has been performed with respect to the following parameters:

- Tortuosity (initially set to a single, material-specific value according to available data);
- Different porosity-diffusion coefficient retroaction laws;
- Kinetic parameters of cement minerals.

Numerical outputs indicate that the pH of cement pore-waters decreases in time as an effect of the propagation of the CO<sub>2</sub>-richer, more acid brines coming from the reservoir and the caprock. Each pH values correspond to the dissolution of different buffering minerals. In particular, Monosulfoaluminate, Portlandite, CSH and Katoite are dissolving in this order, allowing pH to decrease from ~11 down to ~5. Intermediate phases are precipitating and again dissolving such as Straetlingite, decalcified CSH, Zeolite CaP, Chabazite, Illite Mg, Hydrotalcite. The final (meta)stable assemblage comprises above all Calcite and Philippsite (Na, K). Final porosity is lower than the initial one, but highly dependent on the stage and location in the model.

Indeed, at the well-reservoir interface, the decalcification of Monosulfoaluminate into Katoite and Calcite is the only and temporarily cause for porosity increase. Portlandite Ca(OH)<sub>2</sub> then carbonates into Calcite CaCO<sub>3</sub> due to acid and carbonated brine intake, which has almost no influence on porosity. The final decalcification-carbonation of hydrated Silica Gel (CSH) decreases the porosity, as well as Katoite transformation into Zeolite. During these processes, some Calcium also migrates to the reservoir where it precipitates into Calcite. All these phenomena lead to a global reduction of the porosity at both sides of the interface, except at the acid plume propagation front.

However, at the well-caprock interface, the reactions are slower and most elements from dissolved cement materials migrate to the caprock side and precipitate, which increases the porosity at the well side and decreases it at the caprock side. This can significantly affect the behaviour of the storage sealing. We can finally deduce a time-space characterisation of the system with main mechanisms being simplified for a future export to a geomechanical study.