



CO₂-water-mineral reactions during CO₂ leakage into glauconitic sands: geochemical and isotopic monitoring of batch experiments

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The assessment of environmental impacts of carbon dioxide geological storage requires the investigation of the potential CO₂ leakages into fresh groundwater reserves. The Albian aquifer of the Paris Basin was chosen as a case of study because i) the Paris Basin contains deep saline Jurassic and Triassic aquifers identified as targets by the French national program of CO₂ geological storage and ii) the Albian aquifer is a deep freshwater resource of strategic national importance, above the Jurassic and Triassic formations.

An experimental and a geochemical modelling approach were carried out in order to better understand the rock-water-CO₂ interactions with two main objectives: to assess the evolution of the chemistry of the formation water and of the mineralogy of the solid phase during the interaction and to design a monitoring program for freshwater resources. The main focus is to select and develop suitable indirect indicators of the presence of CO₂ in the aquifer. We present here the experimental results, which combines both major and trace elements and isotopic tools, some of them new in the CCS field.

Batch reactors with a liquid/solid ratio of 10 made of appropriate materials (PTFE, stainless steel) were equipped with simultaneous controls on several parameters (pH measurement, gas phase composition, pressure, tightness...) after CO₂ injection (PCO₂= 2 bar; room temperature). Ten reactors were run simultaneously, over pre-determined durations of CO₂-water-rock interaction (1, 7, 15 and 30 days).

During the batch experiment, we observed major changes in several chemical parameters due to the CO₂ injection. A sharp drop in pH from 6.6 to 4.9 was noticeable, immediately after the injection, due to CO₂ dissolution in the water phase. Alkalinity varies from 1.3 mmol.L⁻¹ in the initial water to 2.0 mmol.L⁻¹ at the end of the 1-month experiment. Four types of ion behaviors are observed: (1) calcium, silicon and magnesium concentrations increase during the 1-month experiment; (2) dissolved iron strongly decreases immediately after CO₂ injection; (3) potassium, sodium and fluorine concentrations increase at the start of CO₂ injection and then stabilize to levels higher than the pre-injection concentrations, (4) chlorides and sulfates are stable. These variations indicate dissolution/precipitation and surface reactions involving mineral phases such as glauconite, siderite/iron hydroxide. The experimental results were interpreted and the geochemical mechanisms involved were included in geochemical modeling using PHREEQC, an essential step to quantify the overall effect of the combined individual reactions and processes. These mechanisms were corroborated with isotopic ratio variations. E.g. the variations of δ¹³C_{DIC} (from -15.7 ‰ to -21 ‰ vs. PDB) cannot be explained solely by the CO₂ dissolution, and indicate additional chemical processes. Likewise, shifts of δ¹¹B towards more negative values stress the implication of the glauconitic minerals, mainly B-bearing phase in the system.

These experimental results, and their numerical simulation, are promising for the development of our indirect geochemical and isotopic monitoring technique.