PhD Proposal : Study of Hydro-Chemical Degradation of Caprocks and Its Impact on the Long-Term Integrity of CO_2 Sequestration in Saline Aquifers

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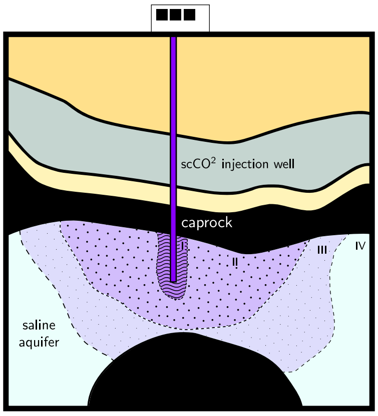
Context

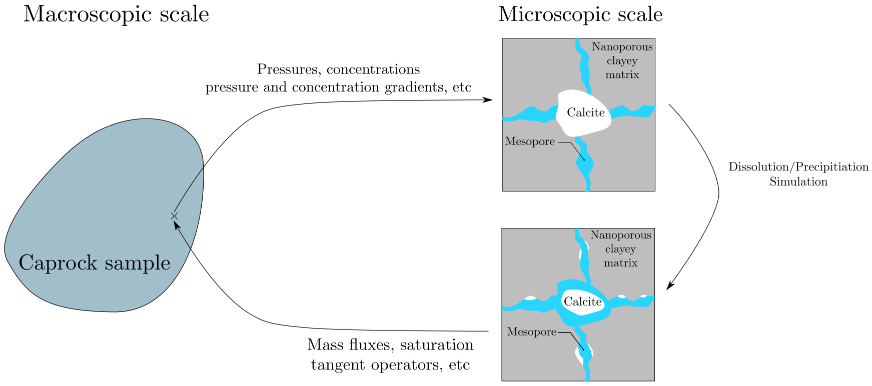
In connection with the IPCC AR6 Synthesis Report, Carbon Capture and Storage (CCS) in deep geological formations has emerged as a key option for reducing greenhouse gas emissions, and CCS facilities continue to expand across Europe. The goal of the ANR LOCCO project is to strengthen research on the long-term integrity of CCS by investigating the interaction between CO_2 injected into geological storage reservoirs and the caprock, which acts as an impermeable barrier preventing CO_2 upward migration and leakage. The long-term objective of this project is to provide tangible insights for improving the performance of current geological CO_2 storage technologies, with a focus on the role of multiphysical coupling processes occurring within the reservoir and at its interface with the caprock. Saline aquifers, which are widespread in France and more broadly in Europe, have the potential to store large volumes of CO_2 (according to the International Energy Agency, saline aquifers could store over 10,000 Gt of CO_2—enough to contain more than 100 years of global emissions). The main hydro-chemo-mechanical phenomena that could compromise the integrity of clay-rich caprocks will be studied in the laboratory (joint PhD) and modeled using new computational and modeling tools as part of this thesis. In geological storage, CO_2 is injected in liquid form (at low temperature and moderate to high pressure) and then transforms into a supercritical phase ($scCO_2$) once injected and heated to the geological formation temperature. However, $scCO_2$ tends to rise due to buoyancy (Archimedes' principle) and accumulate beneath the caprock. Four distinct zones are commonly identified in the reservoir: Zone I, fully saturated with $scCO_2$ near the injection well; Zone II, characterized by a two-phase mixture of $scCO_2$ and brine; Zone III, fully saturated with CO_2-acidified brine; and Zone IV, unaffected by CO_2 injection (see figure (a)). In each of these zones, caprock degradation is driven by different hydro-chemo-mechanical mechanisms, such as the dissolution of mineral compounds like calcite. These degradation processes affects the sealing performance (permeability, diffusivity) of the caprock and may lead to CO_2 leakage via diffusion or percolation (either diffuse or localized, such as fingering). This PhD project focuses particularly on the impact of dissolution on the long-term integrity of such sequestration systems.

Methods

This project aims to model the long-term integrity of CO_2 storage beneath a caprock whose microstructure may evolve due to the presence of calcite, which reacts when the interstitial fluid becomes acidified. To develop a model suitable for engineering practice, it will be necessary to account for the effective behavior of the porous medium. Rather than using an analytical constitutive model, we propose a double -scale approach. At the small scale (meso-pore level), a model of CO_2 advection-diffusion within the meso-pore network will be constructed, combined with a microstructure evolution model. A level-set or phase-field method will be employed to characterize the dissolution/precipitation of calcite. This



(a) Schematic view of different regions of the aquifer during $scCO_2$ injection



(b) Schematic representation of the two-scales modelling strategy considered in this proposal

work will require a comparative study of both approaches to select the most relevant for the application. Upscaling to the engineering scale will be achieved by integrating the results obtained at the small scale (representative elementary volume), assuming that the macroscopic effective behavior of the caprock is governed not by a set of analytical constitutive laws, but by finite element simulations conducted at the microscopic scale, following an FE^2 approach (see Figure (b)). This model will then capture the evolution of the caprock's hydraulic properties, in terms of resistance to CO_2 flow and diffusion induced by calcite dissolution. Once the model and computational tool are established, it will be possible to analyze the long-term integrity of storage in Zone III. The second part of the thesis will focus on modeling the two-phase flow of brine and supercritical CO_2 to characterize the risk of leakage (primarily via capillary fingering). To this end, the model developed in the first part will be extended to incorporate two-fluid flow (brine and $scCO_2$) using a Stokes-Cahn-Hilliard (phase-field flow) approach at the small scale. This model will inform engineering-scale simulations of the progression and stability of the CO_2 front, particularly in Zone II.

Objectives

This thesis presents three key objectives:

i) To develop a dissolution model coupled with two-phase flow at the small scale of a caprock.

ii) To use this micro-model to inform an engineering-scale model.

iii) To analyze the percolation front of CO_2 in a caprock undergoing dissolution, assess the contribution of dissolution to front instability, and identify local mechanisms that may trigger this instability.

About LOCCO Project

This thesis is part of the ANR (French National Research Agency) LOCCO project (hydro-chemo-mechanical LOCalization phenomena in CO_2 geological sequestration), which brings together three French partners (École Centrale de Nantes, École Nationale des Ponts et Chaussées, Université Grenoble Alpes) and international partners (University of Liège, University of Strathclyde, Queensland University). The proposed PhD project will therefore be conducted in close collaboration with these partners, involving the participation of the successful candidate in project-related events and providing an opportunity to develop his/her scientific network. At the local level (Grenoble), the project involves a team of five permanent members and two PhD students, ensuring a dynamic team environment and fully collaborative research work.

Applications and additional informations

Applications - A detailed CV, including transcripts from both Bachelor and Master degrees, along with a cover letter, should be sent to Quentin Rousseau (✉) and Pierre Bésuelle (✉). Letters of recommendation are welcome but not required.

Profils - Knowledge of continuum mechanics, fluid mechanics, the finite element method, and basic scientific computing skills (e.g., Python, numpy, matplotlib) are required to successfully carry out this project.

Location - The PhD will begin in autumn 2026. The successful candidate will join the 3SR Laboratory in Grenoble, specifically the Geomechanics team (website), which conducts cutting-edge research on the mechanics of geomaterials and associated multiphysical couplings.

Remuneration - The successful candidate will be remunerated according to current regulations.