Evolution of mechanical and hydraulic properties in a calcareous rock induced by CO_2 injection

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INTRODUCTION

Owing to the huge combustion of fossil fuels and land use for agriculture and livestock purposes, the concentration of atmospheric CO_2 has increased by about 45% since the industrial revolution (Rohmer et al., 2016). This significant increase has led to a considerable temperature rise (i.e., global warming) in near-earth-surface, entailing urgent global efforts to mitigate CO_2 emissions and moving toward clean, sustainable energy resources. Thus, the CO_2 Capture and Storage (CCS) in deep saline aquifers at depths larger than 850 m with temperatures in the range of 40-150 °C (i.e., low-temperature geothermal sedimentary systems) is considered as a part of the solution for concurrent CO_2 emissions mitigation and geothermal energy production (Newell and Ilgen, 2019).

Meanwhile, injection of the large volumes of CO_2 into saline aquifers is a highly complex process from the fluid-rock interaction perspective. Injected CO_2 dissolves in the host brine and results in the formation of carbonic acid, leading to the production of H^+ ions and pH reduction (Cama et al., 2019; Rohmer et al., 2016). The acidified brine could trigger geochemical reactions primarily concerning mineral dissolution and precipitation, which may induce alterations in the pore structure and hydraulic and mechanical properties of the host rocks (Clark and Varanrio, 2016). These alterations may, in turn, affect the long-term response of the geological medium to the CO_2 injection in terms of fluid injectivity and safety of a CCS project. Hence, the CCS operation success greatly depends on the comprehensive understanding and prediction of the coupled Chemo-Hydro-Mechanical (CHM) processes, necessitating the study of these processes at different scales from laboratory to field.

In this study, reactive transport simulations were conducted to evaluate the chemo-hydraulic response of the Pierre du Gard limestone (100 % calcite) to the injection of CO_2-rich water under supercritical CO_2 conditions (P = 100 bar and T = 60 °C). Simulation results have been then used to design the best experimental conditions to directly measure the chemically-induced alterations in the rock properties via percolation experiments.

METHODOLOGY

1D scoping simulations were executed to assess the chemical effects of a CO_2-rich water injected in the Pierre du Gard limestone core **(Fig.1)** using the CrunchFlow code (Steefel et al., 2015). Appropriate initial and boundary conditions (**Fig.2**) replicating the experimental procedure were considered. A cylindrical core with a diameter of 2.5 cm and a length of 5 cm was initially saturated with distilled water at 60 °C. CO_2-rich water was then injected into the core at the same temperature for about 360 h. The injection was carried out at constant flow rates spanning from 0.1 to 10 mL/min. The CO_2 concentration in the injecting solution was calculated using the PhreeqC code (Parkhurst and Appelo, 2013) in accordance with the CO_2 partial pressure (P_{CO_2} = 100 bar). Variations in the concentrations of chemical species, pH, and porosity along the core were calculated as a function of time in the simulations.

Figure 1. *A Pierre du grad core specimen.*

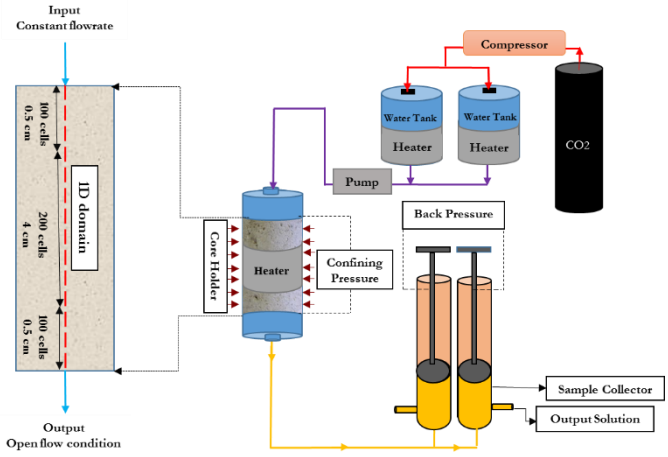

Figure 2. *Schematic of the experimental and numerical setups. The core sample is 5 cm long and 2.5 cm in diameter.*

SCOPING SIMULATION RESULTS

Simulation results revealed that, regarding the fast interaction between calcite as the building block of the Pierre du Gard specimens and CO_2-rich water, a continuous dissolution of calcite and porosity enhancement along the cores would occur. It is predicted that calcite dissolution and porosity increase will occur more dominantly in the inlet part of the cores (first 15 mm) since the pore-fluid evolves towards equilibrium as it passes through the cores. This effect is caused by Ca^{2+} and CO_3^{2-} ions and associated aqueous complexes produced in the inlet section, with corresponding higher pH values. Yet, it should be mentioned that equilibration of the injected solution becomes more pronounced with decreasing the flow rate, resulting in a lower porosity enhancement along the cores.

Albeit these calculations may give us a general overview of the chemically induced alterations in the properties of the Pierre du Gard cores, experimental results may be nonidentical to scoping simulations. Thus, we need to assess fluid-rock interactions utilizing percolation experiments planned to be done in the upcoming phase of this study.

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