

## Research domain

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Physics and chemistry of the Environment

## Title

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Capillary effects in heterogeneous porous reservoirs

## Context

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Deep geological repositories selected by several European countries to store their nuclear wastes are argillaceous formations known for their very low permeability that ensures the water containment. Their very low permeability is related to nanopores weakly connected to each other, which also promote the capillary state of trapped water [1]. Yet, capillary water super-solubilizes solid and gases [2], promotes cold boiling that changes the liquid-air distribution [3], and modify the stress field within the host matrix [4]. And these features are lacking in the standard conceptualization of the water-rock-gases interactions throughout the waste disposals [5]. This project is directed to fulfill this gap and establish which thermodynamic and poromechanical consequences can be expected throughout the formation.

The whole massif can be conceptualized as a dual porous medium with a strongly contrasted, nano-to-millimeter, pore size distribution (PSD). Observationally, the ventilation of the tunnels causes near-field desaturation of the formation with limited water loss, and triggers geochemical reactions mainly linked to the gases injected in the massif. The fractures of the EDZ play the role of preferential flow-paths giving assurance that only limited volumes of water and reactive materials are affected, and that the geochemical impact on porosity stays negligible. However, the geochemical dynamics induced by capillarity can challenge the common line of reasoning, especially when involving the metastable regime spontaneously settled in fractures behind nanoscopic menisci. Explosive bubble nucleation (or cold boiling, or cavitation) is triggered wherever the pore body exceeds the value fixed by the size of the liquid-air capillary bridges. Further percolation of these nucleated bubbles across the formation can be expected, therefore changing the flow paths. Meanwhile, the bubble nucleation is accompanied by relaxation of the thermochemical properties, generating brutal supersaturation that can initiate various geochemical (clogging) or poromechanical (crystallization pressure) processes.

Consequently, this study aims to shed light on the processes specific to double porosity systems when the sizes are very strongly contrasted (nm-mm). Experiments, at the scale of a decimetric reactive column, will evaluate the geochemical effects of the capillary state and its consequences on the porosity, the water-air distribution, and the dynamics of water-air-solid interactions. In other words, the main objective is to understand, on an experimental basis, how the two-phase distribution in a porous medium and the intensity of its capillary state controls the progress or passivation of solid-solution and gas-solution reactions, and the associated stress fields. The Ph. D. will involve experiments and reactive transport modeling.

The galleries of the nuclear waste storage area are a good example of an application, but other geological systems are possible, such as when a CO<sub>2</sub> bubble reaches the impermeable cover rock.

[1] Matray J.-M., Savoye S., Cabrera J. *Engin. Geology* 90, 1–16 (2007).

[2] Hulin C., Mercury L. *Geochim. Cosmochim. Acta* 252, 144-158 (2019a).

[3] Hulin C., Mercury L. *Geochim. Cosmochim. Acta* 265, 279-291 (2019b).

[4] Mercury L. et al. *ACS Earth Space Chem* 5(2), 170-185 (2021).

[5] Tremosa J. , D. Arcos, J.M. Matray, F. Bensenouci, E.C. Gaucher, et al. *Applied Geochem.* 2, 1417–1431 (2012).

## Objectives

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The project therefore aims at improving our mechanistic understanding of porosity-reactivity relationships in dual nanopores-bearing media under unsaturated conditions. Experiments, at the scale of a decimetric reactive column, will evaluate the geochemical effects of the capillary state and its consequences on the porosity, the water-air

distribution, and the dynamics of water-air-solid interactions. In other words, the main objective is to understand, on an experimental basis, how the two-phase distribution in a porous medium and the intensity of its capillary state controls the progress or passivation of solid-solution and gas-solution reactions, and the associated stress fields. The Ph. D. will involve experiments and reactive transport modeling.

## Means

The experiments will be performed at the core scale, bearing on an existing design developed to study carbonation reactions under unsaturated conditions with a limited water availability. Special columns will be composed and submitted to various water/gases fluxes. Phase transitions and cracking will be followed by X-ray tomography and non-intrusive sonometry. Chemical and isotopic measurements as well as Raman mapping will serve to localize the critical chemical interfaces.



Figure 1. Left: decimetric column and schematic diagram (left), and column before assembly with the CEMLab gas generator / mixer (Center, MIMAROC platform, BRGM); and right: porous medium with controlled distribution.

Reactive percolation modeling taking into account the capillary state will also be carried out to quantify the expected mass balances and the conceptual sketch of processes required to optimize them. The code used (PHREEQC 1 D column using the THERMO\_ZNS-produced capillary database) will extend the mechanistic conclusions within the framework of a representative elementary volume.

## Practical informations

Preferred starting date: 01/10/2016

Funding: Grant funded by French Ministry of Research and BRGM.

Salary: 1700 € net/month, with possible complementary teaching activities

Organization of employment: CNRS Orléans

## Contacts

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Interested applicants should submit their application in one pdf file that contains a cover letter, CV, and research statement.

Dead-line for application: 30 June 2022.

## Profile of the candidate

Masters with majors connected to some of the following: Water-rock interactions, Porous media, Physics of phases transitions, Thermodynamics, Micro-/nano-fluidics, Surfaces and interfaces.