

Experimental report

30/09/2025

Proposal: 1-07-14

Council: 4/2023

Title: Imaging water diffusion front in clay porous media

Research area: Engineering

This proposal is a new proposal

Main proposer: Eric FERRAGE

Experimental team: Lucas DESERT
Eric FERRAGE
Sebastien SAVOYE
Laurent MICHOT
Emmanuel TERTRE
Arnaud MAZURIER

Local contacts: Alessandro TENGATTINI

Samples: callovo-oxfordian clay
illite clay powder

Instrument	Requested days	Allocated days	From	To
NEXT	4	3	10/12/2023	12/12/2023

Abstract:

Predicting the long-term transport of water, ions and solutes in clay-rich media, such as clayey barriers for nuclear waste repositories, is a major environmental issue. In that context the prediction of time evolution of water diffusional front in fully or partially water-saturated clay porous media is still not fully understood. We wish to beneficiate from D2O/H2O image contrast on NeXT instrument to obtain such information. Using water through-diffusion set up and by recording, as a function of time, 3D image of H2O diffusion front on initially fully or partially (with different suction pressures) saturated samples with D2O, these experiments and the quantitative treatments of the images obtained should provide key insights for the correct description and prediction of the 3D diffusion front of water tracers in clay porous media.

CONTEXT

Predicting the long-term transport of water, ions and solutes in clay-rich media, such as clayey barriers for nuclear waste repositories, is a major environmental issue. In that context the prediction of time evolution of water diffusional front in fully or partially water-saturated clay porous media is still not fully understood.

For addressing this problem, water diffusion measurements are commonly obtained using through-diffusion techniques where the clay material (1cm diameter, 1.5 cm length) is compacted into a diffusion cell and a circulation is applied between two reservoirs (i.e., upstream and downstream reservoirs).

For water saturated conditions, experimental diffusion coefficients are consistent with those derived from numerical calculations. The situation differs in the case of partially saturated conditions, where a significant misfit (x10) between experimental and calculated diffusion coefficients is obtained, which raises several questions about the accessibility of water tracer in partially water-saturated conditions and on the possible presence of heterogeneities within clay samples leading to inhomogeneous diffusional front and non-accessible zones for the water tracers.

EXPERIMENTS AND DATA TREATMENT

In the framework of the Ph.D. of Lucas Desert, this proposal aimed at using NeXT for in-situ imaging in 3D the shape of the water diffusion front in fully and partially water-saturated conditions during the transient regime.

We considered 2 types of clay samples, i.e., a polymineralogic Callovo-Oxfordian sample and a pure illite reference, i.e., the most abundant clay mineral present in Callovo-Oxfordian material. Using aluminum through-diffusion setup, samples were compacted at a porosity of ~30% and equilibrated at 0 (i.e., fully saturated conditions) and 9MPa of suction pressures. Prior to neutron analysis, samples were equilibrated with D₂O in both upstream and downstream reservoirs. Then D₂O was changed to H₂O in the upstream reservoir at time=0, and the heterogeneous diffusional water front of H₂O obtained by image contrast was followed through time. Neutron tomography of the whole sample cell was recorded every 20min with a resolution of 28μm to image the diffusional fronts of H₂O.

As seen from Figure 1 for illite samples, the evolution of the H₂O front on selected slices from full neutron tomography is clearly visible. The difference in dynamics between fully and partially saturated conditions is also evidenced.

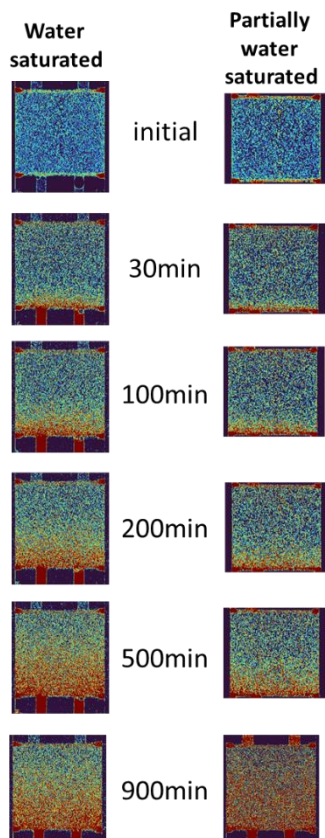


Figure 1: evolution with time of the propagation of H₂O in the diffusion cell (from the bottom to the top of the cell) in fully water saturated (left) or partially saturated (right) state for illite.

Using radial integration, 1D profiles of grey levels were derived to highlight H₂O front evolution within the samples as a function of time (Figure 2).

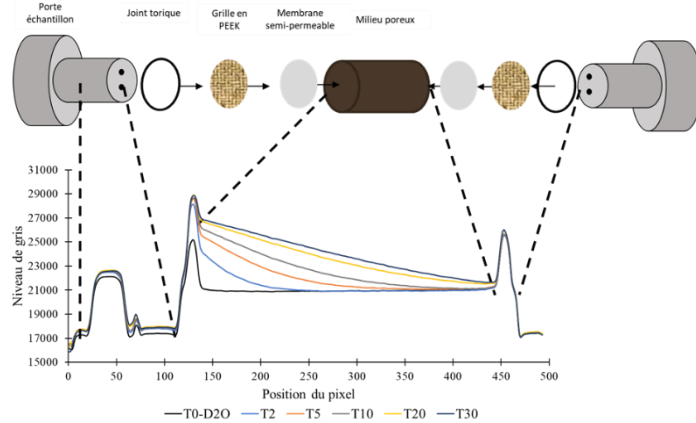


Figure 2: experimental evolution of 1D profiles of grey levels in water-saturated illite (bottom) and sketch of the cell elements (top).

Based on these profiles and by subtraction with the initial D₂O saturated profiles, the time-evolution of H₂O front was extracted and compared with 2D simulations (Figure 3).

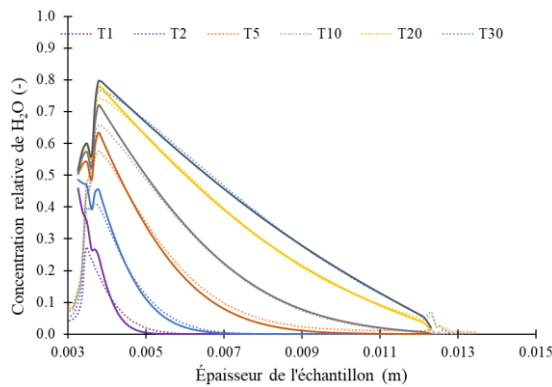


Figure 3: comparison between experimental evolution of 1D H₂O front and calculated profiles using CrunchClay software for water-saturated illite.

For these simulations performed with CrunchClay software, it was possible to evidence the need for a detailed description of all elements (joins, peek grid, injection points, etc.) to correctly reproduce the experimental data (Figure 4).

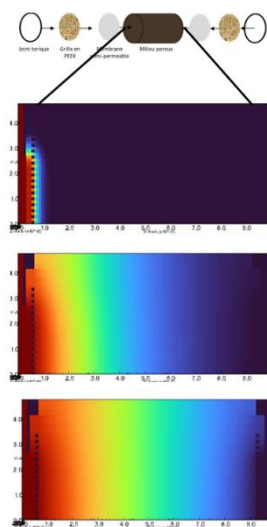


Figure 4: CrunchClay simulations and 2D evolution of the calculated H₂O front with exact sample geometry for water-saturated illite.

MAIN CONCLUSIONS

- The NEXT instrument worked perfectly and this instrument is perfectly adapted for investigating time-resolved water transfer in clay-rich rocks.
- For illite sample, the detail imaging of the transient regime allowed us to reveal the role played by the geometry of the sample and diffusion cell on the H₂O front evolution and thus on the diffusion coefficients obtained. Based on these more realistic simulations compared to what was done so far, a large part of the misfit between experiments and simulations is now understood.
- For Callovo-Oxfordian samples, the H₂O front evolution in water saturated conditions was also found consistent with simulation. Unfortunately, the partially-saturated sample showed strong inhomogeneity in water content (assigned to partial condensation during transport in winter) that made impossible further analysis.
- All these data represent a part of the Ph.D. of Lucas Desert and a manuscript is under preparation for publication.