

Visualising the coupled mechanics of CO₂ breakthrough

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1 Introduction

Successful geological CO₂ storage relies on the availability of an impermeable caprock formation that overlies the injection reservoir and prevents CO₂ migration to the surface through a combination of trapping mechanisms that are activated at different scales. Shales have been widely studied as potential caprock formations because of their favourable properties such as very low permeability and high capillarity. Shales are highly heterogeneous and anisotropic materials whose response is governed by strong Thermo-Hydro-Chemo-Mechanical (THMC) couplings that conventional constitutive models often fail to represent. In the context of CO₂ storage, characterisation of caprock integrity and sealing capacity is usually evaluated in terms capillary entry pressure [1, 2]. In addition to the hydromechanical response, CO₂ introduces a chemical component (mineral dissolution and/or precipitation) the impact of which on shales remains a challenging topic due to the extreme mass transfer limitations. In this study, the impact of CO₂ injection in a Swiss shale, the Opalinus Clay, is for the first time assessed with live x-ray tomography. This approach aims to identify and decouple the different THMC mechanisms that take place, through quantitative 3D image analysis.

2 Hydromechanical testing during live x-ray tomography

Opalinus Clay samples ($h = d = 5$ mm) are tested during in-situ x-ray tomography using an original high pressure and temperature cell [3]. The presented work is divided in two parts: (i) CO₂ injection at increasing pressure steps (u_{CO_2}) under isotropic confinement (p) and (ii) direct isotropic exposure to supercritical CO₂ (p_{CO_2}). In both series of tests, the tested Opalinus Clay sample has been progressively resaturated under free swelling conditions before being mounted in the x-ray compatible cell. The kinematics of the tested samples are acquired and quantitatively processed with 3D x-ray image analysis during different time intervals.

3 CO₂ breakthrough

CO₂ breakthrough under confined conditions (isotropic compression, $p = 10$ MPa) is assessed with live x-ray tomography ($7.8 \mu\text{m}/\text{px}$) and the volumetric response of the sample is evaluated with image analysis on the different scans before and after each loading phase. CO₂ is injected in the sample from the bottom ($h = 0$) under increasing constant pressure steps of 2 MPa ($u_{CO_2} = 2, 4, 6$ and MPa). Upon confinement application the sample contracts while after the first CO₂ pressure level (scan 02) no significant volumetric activity is measured. Further CO₂ pressure increase (scans 03 to 05) results in swelling response, which is attributed to CO₂ breakthrough, i.e., breakthrough pressure 2-4 MPa (consistent with [4]). The calculated volumetric maps (Figure 1) reveal an increasing localized swelling activity evolving from the bottom (injection) side of the sample upwards – it is particularly visible in scan 05.

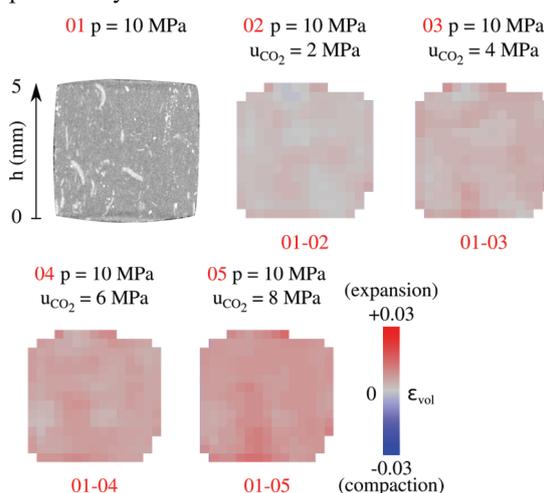


Fig. 1: Localised volumetric deformation at different levels of CO₂ injection pressure

The more localized swelling activity of the sample is further demonstrated in Figure 2, where the max. volumetric strain throughout the sample's height is plotted. When CO₂ breakthrough occurs ($u_{CO_2} = 4$ MPa) higher volumetric strain is measured close to the injection

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which further increases as injection increases. The varying pattern of max. vol. strain along the sample suggest the creation of additional breakthrough conduits. It is important to mention the pre-existence of micro-fissures in the sample initially under unconfined conditions. Even though these micro-cracks disappear after the application of confinement (at the given resolution) and are not any more visible even after breakthrough, the high swelling activity can be attributed to locations where high concentration of fissures is observed. These motivating results show that even at resolutions lower than the average pore size of the material, 3D image analysis can reveal important insight on the localised behaviour which in the context of CO₂ storage can be related to potential leakage paths.

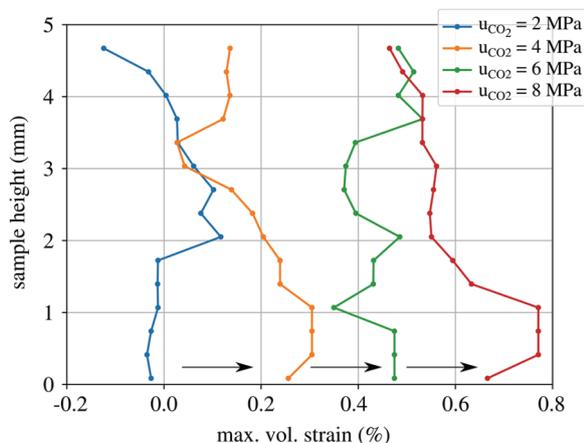


Fig. 2: Maximum volumetric strain along the height of the sample for different CO₂ injection pressures

4 Isotropic exposure to supercritical CO₂

Isotropic exposure of an Opalinus Clay sample to supercritical CO₂ (pCO₂ = 10 MPa pressure and 34°C) is evaluated with 3D x-ray imaging (5.7 μm/px) over a time period of 56 days. Direct CO₂ exposure results in initial swelling due to thermal loading, followed by slight compaction and local micro-fissuring due water evaporation in the anhydrous CO₂ (see Figure 3). This interaction can occur at the bottom of the caprock formation with the buoyant CO₂, leading to partial desaturation of the caprock which can threaten its mechanical integrity. Again a highly localised strain activity around the three pre-existing horizontal fissures is observed. Cracks are more prone than intact matrix to opening/closing upon THM loading which here is imposed both by thermal loading and CO₂ drying, they thus have a crucial role in the integrity of the entire storage system.

For the first time, local CO₂ concentration in the material has been measured based on the developed method explained in [5], where density variations attributed to constant mass volumetric deformation from Digital Volume Correlation can be directly compensated by applying an attenuation correction. Any remaining

grey value difference is attributed to mass exchanges, here CO₂ invasion. Sample swelling enables CO₂ invasion in the material, in particular through the fissured zones, while upon volume stabilisation CO₂ concentration continues to increase even in zones within the clay matrix. Remaining increased density regions around the cracks after depressurisation could correspond to chemical activity (mineral precipitation) from CO₂ exposure. While this cannot be confirmed from the post-exposure mineralogical analysis, these results reveal the potential of in-situ imaging of small size shale samples on the detection and understanding of the different coupled THMC phenomena.

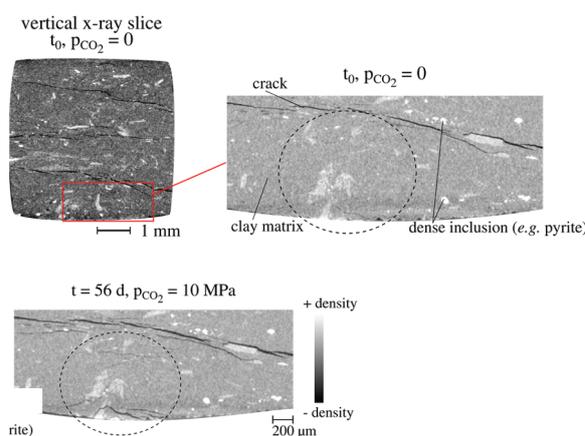


Fig. 3: Desiccation fissures due to contact with supercritical CO₂

References

1. Boulin, P. F., Bretonnier, P., Vassil, V., Samouillet, A., Fleury, M., & Lombard, J. M. (2013). Sealing efficiency of caprocks: Experimental investigation of entry pressure measurement methods. *Marine and Petroleum Geology*, 48, 20-30.
2. Espinoza, D. N., & Santamarina, J. C. (2017). CO₂ breakthrough – Caprock sealing efficiency and integrity for carbon geological storage. *International Journal of Greenhouse Gas Control*, 66, 218-229.
3. Stavropoulou, E., & Laloui, L. (2022). Evaluating the impact of effective stress on the CO₂ entry pressure of a caprock material: a multi-scale experimental approach. *under submission*
4. Minardi, A., Stavropoulou, E., Kim, T., Ferrari, A., & Laloui, L. (2021). Experimental assessment of the hydro-mechanical behaviour of a shale caprock during CO₂ injection. *International Journal of Greenhouse Gas Control*, 106, 103225.
5. Stavropoulou, E., Andò, E., Roubin, E., Lenoir, N., Tengattini, A., Briffaut, M., & Bésuelle, P. (2020). Dynamics of water absorption in callovo-oxfordian claystone revealed with multimodal x-ray and neutron tomography. *Frontiers in Earth Science*, 8, 6.