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INVESTIGATING UNDERGROUND CO₂ STORAGE IN POROUS MEDIA USING GEOLOGICAL LABS ON CHIP

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KEY WORDS

Microfluidics , high pressure , porous media , CO₂ geological storage , invasion processes

SHORT SUMMARY

CO₂ geological storage in deep saline aquifers represents a mediation solution for reducing the anthropogenic CO₂ emissions. Consequently, this kind of storage requires adequate scientific knowledge and tools at the pore scale to evaluate injection scenarios or to estimate reservoir capacity. In this context, porous media designed inside high pressure / high temperature microfluidic reactors (micromodel or geological labs on chip - GLoCs) turn out to be excellent tools to complement the classical core-scale experimental approaches to investigate the different mechanisms associated with CO₂ geological storage in deep saline aquifers).

This paper will first highlight the latest results obtained at ICMCB concerning the application of the GLoCs to study the invasion processes of CO₂ in water and brine saturated GLoCs. In particular, direct optical visualization and image treatments allow following the evolution of the CO₂/brine phase distribution within the pores, including displacement mechanisms and pore saturation levels. Finally, we will present some ongoing work aiming at integrating in situ spectroscopy techniques in HP microreactors to get information about the dissolution and mineralization trapping. Beyond CO₂ geological storage investigations, the GLoCs could also find wider applications in geological-related studies such as Enhanced Oil Recovery, shale gas recovery or geothermal energy.

EXTENDED ABSTRACT

Introduction

The sustainable management of anthropogenic CO₂ emissions is currently a key factor for stabilizing the global atmospheric content of this greenhouse gas. CO₂ management is a fragile equilibrium between the will to reduce the human impact over the environment and the political and economic context. In addition to enhance oil recovery through CO₂ injection, two strategies with different timescale are envisioned to address part of this issue: (i) the immediate use and transformation of small amount of CO₂ to beneficial products (usable products, energy, etc.) and (ii) the ultimate CO₂ Geological Storage (CGS) in which it is foreseen storing large quantities of CO₂ within deep underground formations. In the latter, deep saline aquifers are ideal candidate sites thanks to their large estimated storage capacities (over ten trillion tons of CO₂). Consequently, this kind of storage requires adequate scientific knowledge to evaluate injection scenarios, estimate reservoir capacity and asses leakage risks. Admittedly, there are four trapping mechanisms involved during the CO₂ storage within deep saline aquifers: (i) the stratigraphic trapping, (ii) the residual trapping, (iii) the solubility trapping and (iv) the mineral trapping. For each trapping mechanism, experimental data is required to be able to adjust

the numerous numerical models. Although conventional lab scale means (typically core-scale experiments) are adequate to demonstrate proofs of concept and to access some experimental data, there is a lack of in situ characterization techniques, thus limiting the monitoring and the understanding of the various involved processes. Therefore, new methods are needed to investigate deeply the fundamental mechanisms associated with these four trapping mechanisms. In this context, microfluidics approaches have brought several advantages over conventional experimental means, including fast screening of the parameters, fast heat and mass transfer, the ease of implementation of various characterization techniques (including optical), etc. [1] By adapting such approaches to porous media, several research teams have developed “micromodels” to investigate key parameters related to multiphase flows in porous media [2-5]. Recently, in the frame of a large scale French research project dedicated to the experimental and numerical investigation of CO₂ geological storage and gathering 8 laboratories (CGSμLab), our team has demonstrated the fabrication and use of the first high pressure / high temperature microreactors, able to withstand harsh conditions up to 20 MPa and 400°C [6], which were used in particular to investigate CO₂ / Water hydrodynamics at high pressure [7]. Herein, we focus on the results obtained at ICMCB by coupling these “new” microtools, which were adapted to studies dealing with fluidic within porous media, so called “Geological Labs on Chip – GLoCs” [8], with optical and spectroscopic characterization means to investigate key mechanisms related to CO₂ geological storage in deep saline aquifers.

Experimental approach

We have developed and used high pressure / high temperature microfluidic tools [6] for studying and characterizing typical issues related to CO₂ trapping mechanisms in deep saline aquifers – structural and residual trapping, solubilization and mineralization (Fig. 1). Our fabricated 2D porous networks are created in silicon/Pyrex microsystems to replicate the reservoir p,T conditions ($25 < T (^{\circ}\text{C}) < 50$, $50 < p (\text{MPa}) < 10$), geological and topological properties (porosity, brine composition). The simple microfabrication procedure, based on photolithography followed by wet etching of silicon provides flexibility in terms of designs (2D masks extruded in the third dimension at various depth). These microtools are able to mimic the actual storage conditions encountered in real storage reservoirs, while taking advantages of the fast heat and mass transfers provided by the microfluidic approaches.

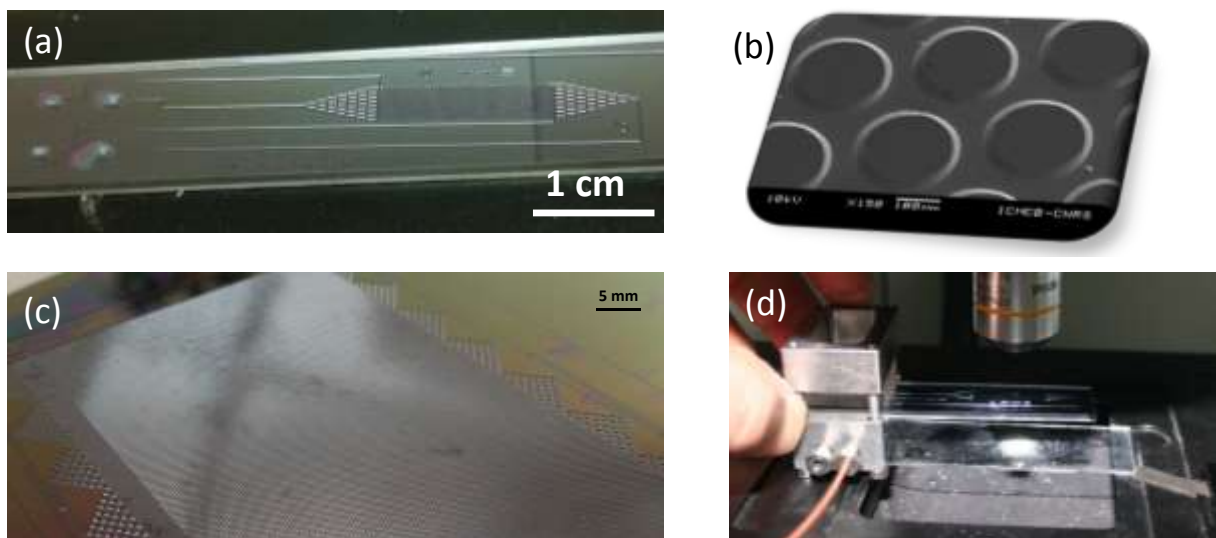


Figure 1: (a) Full view of a GLoC with a typical 2D porous pattern, (b) SEM images of the etched porous media, (c) larger porous medium on chip and (d) GLoC device placed under a confocal Raman spectrometer.

We have been using the GLoCs to investigate the different mechanisms associated with CO₂ geological storage in deep saline aquifers. Indeed, the ability to easily and quasi-instantaneously monitor fluid flow and pressure inside the HP micromodels, allows us to access data such as pressure drop that are linked to the permeability of the pore network. These two parameters have been studied experimentally with several type of GLoCs in order to determine the influence of each geometrical feature and p,T conditions. These experimental data are then compared to numerical modelling results.

Results

We have then focus on the use of GLoCs to investigate the capillary trapping mechanism at pore scale. Images of micromodels were analyzed using the ImageJ software to quantitatively evaluate the CO₂ saturation (SCO₂). We first validated the method by comparing the measured porosity from images with a full CO₂ saturated microsystem and the calculated porosity (by the measured circumference of the silicon plots and the design value). The comparison showed good agreement with errors < 2 %. The details of the method are reported in Buchgraber et al [9].

The direct optical visualization and image treatments allow us to follow the evolution of the CO₂/brine phase distribution of the injected CO₂ within the reservoir, including displacement mechanism (formation of pools, bridges or domes), fluid flow pathways, pore saturation levels, which are key parameters to be determined to estimate the true storage capacity of an aquifer as well as the stability of the stored volume. Additionally, the optical characterization techniques can be used to monitor the different behaviors of the CO₂/brine system under flow conditions as a function of time. When CO₂ enters the pores network saturated with water, we have identified three main mechanisms (Figure 2): (i) the invasion (the CO₂ invades the porous medium, breaking the capillary barrier in the pore and displacing water to occupy the pore space – Figure 2-b), (ii) the percolation, (when the CO₂ reaches the outlet of the micromodel, leaving behind some water due to a capillary forces) and (iii) the drying, (the dry injected CO₂ vaporizes the water remaining in the pore network) [10]. The setup is highly flexible to tune the p,T conditions, flow rates as well as chip geometry, depth and wettability. All these parameters have been investigated to study their influence on the CO₂/brine behaviors and the CO₂ saturations of the porous media (ex: Figure 2-c for the influence of the p, T conditions) and thus to evaluate their impact on the capillary and residual trapping mechanism.

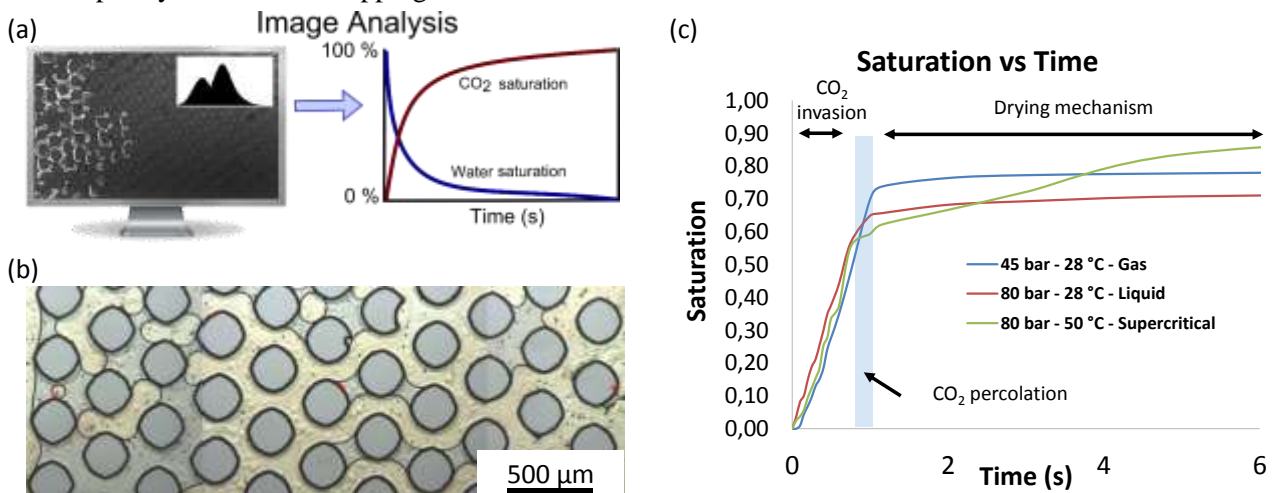


Figure 2: (a) General strategy based on direct image analysis to access CO₂ and water saturation inside a porous medium, (b) pores network during the invasion process, displaying both area saturated with water and CO₂, (c) evolution of the porous medium saturation in CO₂ as a function of pressure and temperature conditions (adapted from [10]).

Finally, it is possible to envision the coupling of GLoCs to spectroscopy and optical characterization to get information about dissolution and mineralization trapping [11]. The Raman spectroscopy directly applied on our chips gives us a fast and accurate way to identify and quantify dissolved carbon dioxide in water and salt water [12] or to study carbonate precipitation. In particular, by adding CaCl₂ to the water phase, we can observe the precipitation of calcite (characterized by In situ Raman spectroscopy) at the interface between water and CO₂, following the reaction: $\text{Ca}^{2+} + \text{CO}_3^{2-} \rightarrow \text{CaCO}_3$ (Figure 3).

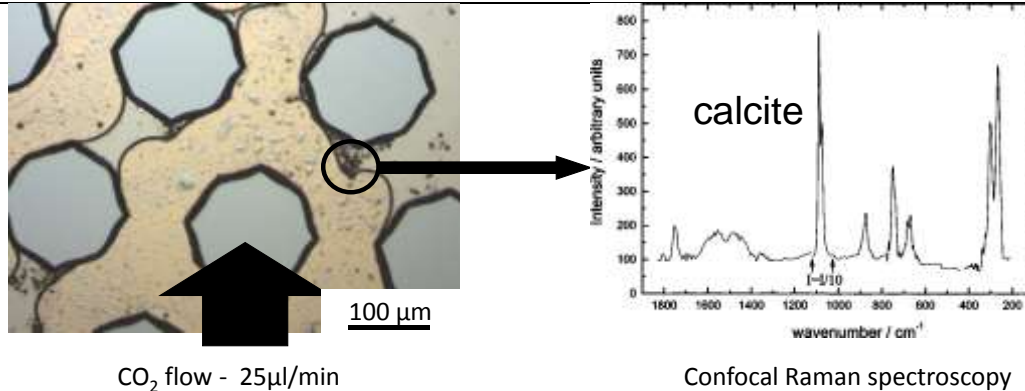


Figure 3: Calcite precipitation during the CO₂ invasion process of a CaCl₂ aqueous solution-saturated porous medium on chip (T = 35°C, p = 10 MPa, [CaCl₂] = 0,1 M, QCO₂ = 50 μL.min⁻¹) with additional *in situ* Raman characterization.

Conclusion

The recently developed Geological Labs on Chip (HP micromodels) have been confirmed as fast and accurate microtools to identify and quantify dissolved carbon dioxide in water and salted water, but also to study invasion mechanisms depending upon flow rates, porous medium characteristics and brine composition (in particular for studying carbonate precipitation and drying mechanisms). These approaches are proved viable in the laboratory, and can be used to simulate and analyze various CO₂ geological storage issues or other geological-related topics such as Enhanced Oil Recovery (EOR), shale gas recovery or geothermal energy. In the frame of the CGSμLab project, these tools were coupled to *in situ* X-rays to investigate crystallization processes [13], to optical fibers integrated in chip for CO₂ detection and to Brillouin spectroscopy. Additionally, they are used in a complementary way to conventional experiments using silica capillaries and macroscale high pressure cells. The obtained data are then used to feed the numerical models realized by our partners. Future works include the development of real 3D porous media on chip coupled to *in situ* X-ray tomography in order to get closer to real porous systems by coupling chemical reactivity to hydrodynamics and thermodynamic aspects.

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