

Perspective

Revealing subsurface dynamics: Imaging techniques for optimizing underground energy storage

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Abstract:

Subsurface processes play a crucial role in determining the efficiency and viability of key applications with significant technical and economic implications, including hydrocarbon production, CO₂/H₂ geo-storage, and environmental engineering. A comprehensive understanding of natural behavior including microstructures, morphologies, and various petrophysical properties at pore scale is vital for optimizing the utilization of underground energy storage formations. Despite ongoing efforts, the behavior of diverse natural phenomena in the subsurface remains inadequately understood. This work leverages imaging techniques in conjunction with flow displacement experiments in investigating various natural phenomena, such as CO₂/H₂ geo-sequestration and fracture propagation. Additionally, the significance of microfluidic experiments in studying the dynamics of multiphase flows are briefly underscored. As a conclusion, porous media characterisation at pore scale is valuable for the advance in the understanding of natural phenomenon in subsurface engineering and the subsurface sciences, and upscaling them across space and time.

1. Introduction

Utilizing a range of imaging modalities, as depicted in Fig. 1, allows for a thorough investigation into the microscopic behavior of underground energy within storage formations. This strategy improves our understanding of microstructures, morphologies, and diverse petrophysical properties across a range of scales.

Among the imaging technologies, micro-computed tomography (micro-CT) imaging offers a window into these microenvironments, enabling a detailed understanding of the physical and chemical processes that govern energy storage efficiency and safety (Blunt et al., 2013). Recently, micro-CT imaging, coupled with flow displacement experiments, are designed to visualize and quantify multiphase flow in porous rocks at pore scale. The schematic diagram depicting the typical protocol of flow experiment conducted in a flow cell with *in-situ* micro-CT imaging is illustrated in Fig. 2. Following tomographic

reconstruction, a sequence of 3-dimension (3D) images is generated. Subsequently, these images undergo processing and analysis to extract specific properties such as fluid saturation, relative permeability, capillary pressure, or temporal changes in topological characteristics. These findings serve as the foundation for theory development, model validation, and a variety of other applications (Berg et al., 2013; Berg et al., 2016; Gao et al., 2017; Zou et al., 2018a, 2018b).

The direct insight into microscopic details in particular access to the topology of fluids within the pores and the time evolution *in-situ* has led to unprecedented progress in the understanding of multiphase flow in porous media. This approach ranges from CO₂ geo-storage (Andrew et al., 2014; Herring et al., 2016), oil recovery (Wang et al., 2021), gas hydrate formation in sediments (Cai et al., 2020; Wang et al., 2020), deep coal seam behavior to geologic reactive transport (Iglauer et al., 2011; Al-Khulaifi et al., 2018; Zhang

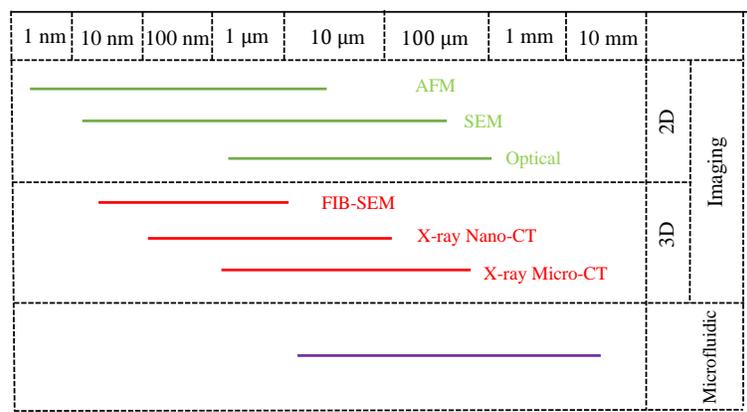


Fig. 1. Multiscale imaging and other characterization techniques (modification after (Ma et al., 2017; Arif et al., 2021)).

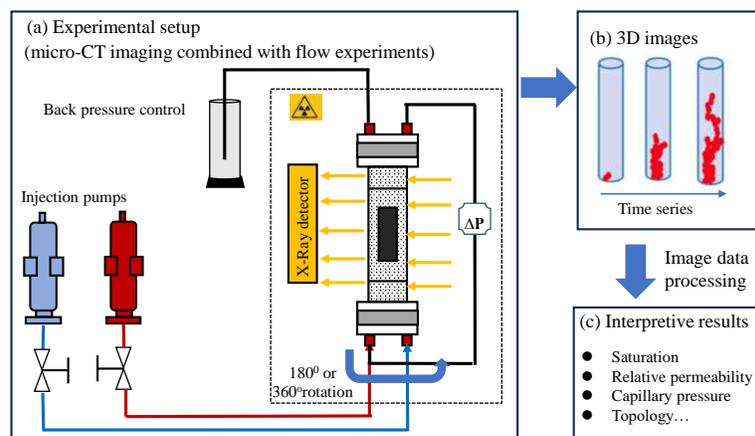


Fig. 2. (a) A conventional flow experiment employing micro-CT takes place within a flow cell, coupled with *in-situ* micro-CT imaging, (b) after the tomographic reconstruction, a series of 3D images is produced and (c) image data processing is then processed and analyzed to extract specific properties.

et al., 2019). The data acquired via micro-CT experiments can thus significantly improve fundamental understanding of such pore-scale phenomena and can also be used to develop and benchmark computer models.

This paper firstly provides the technical challenges associated with conducting fluid flow experiments using micro-computed tomography. A thorough insight into the key applications that have derived benefits from micro-CT imaging studies is then presented. Additionally, the microfluidic technique and its application in the context of multiphase flow are introduced.

2. Imaging techniques in underground energy storage

2.1 CO₂ geo-sequestration

The increasing urgency to combat climate change and transition to sustainable energy systems has brought underground gas storage to the forefront, especially for CO₂ geo-sequestration and hydrogen as a clean energy source. For CO₂ geo-sequestration technology, when CO₂ is injected into geological formations, it acidifies the resident brine, initiating

chemical reactions with minerals. These reactions can modify the hydraulic and mechanical properties of the rock, impacting reservoir and wellbore integrity, injectivity, long-term compaction, and caprock sealing capacity (Benson and Cole, 2008; Benson et al., 2013).

In CO₂ storage, pore-scale imaging has provided invaluable insights into the mechanisms that govern CO₂ behavior in subsurface environments. Key mechanisms include structural trapping (CO₂ is physically trapped beneath impermeable rock layers), residual trapping (CO₂ is trapped in pore spaces), solubility trapping (CO₂ dissolves in the formation water), and mineral trapping (CO₂ reacts with minerals to form stable carbonate compounds) (Wildenschild et al., 2011; Iglaier et al., 2015; Hu et al., 2017; Lebedev et al., 2017; Ahmed et al., 2019; Yu et al., 2019; Moghadasi et al., 2023).

2.2 Supercritical CO₂ injection into coal seam gas

In geological storage sites, injected CO₂ in deep reservoirs (>800 m) attains supercritical conditions (pressure >7.3 MPa and temperature >31.1 °C), resulting in a liquid-like density that enhances storage efficiency. However, the lower density

of supercritical CO₂ compared to resident brine creates buoyancy, necessitating low permeability caprocks for permanent underground containment. The processes for trapping buoyant CO₂ in the subsurface remain a widely discussed topic, with ongoing research exploring how CO₂ migrates through the micrometer-scaled pore space of the rock and interacts with both the rock and resident formation brine.

One of proposed storage sites for CO₂ storage is coal seams. CO₂ injection into deep un-mineable coal seams can enhance coalbed methane recovery, and this process has gained substantial interest in recent decades. In the work of Zhang et al. (2019), 3D micro-CT was adopted to investigate fluid-coal interactions at pore-scale at reservoir conditions via *in-situ* core flooding experiments. Supercritical CO₂ was injected into a heterogeneous water-bearing bituminous coal at reservoir conditions with 15 MPa confining pressure, 10 MPa pore pressure, and 323 K. Under these conditions, the coal's microstructure (mainly the calcite-fusinite mix phase) partially dissolved after CO₂ flooding. After the Supercritical CO₂ was injected into the water bearing coal, the Supercritical CO₂ mixed with the brine caused significant chemical dissolution and clearly wormholes were created. The dissolved volume significantly increased the porosity and pore connectivity in the coal sample. This process could serve as an environmentally friendly and relatively harm free acidizing method in enhanced coal bed methane, which can significantly increase permeability and thus methane production and CO₂ injectivity, at least when carbonates are present.

2.3 H₂ storage

Hydrogen is attracting global attention as a key future low carbon energy carrier. Large-scale hydrogen storage can help alleviate the main drawbacks of renewable energy generation, their intermittency, and their seasonal and geographical constraints (Ershadnia et al., 2023; Wang et al., 2023). The most common geological formations to store H₂ are mainly salt caverns and, aquifers, depleted oil and gas fields since they provide large scale volume for storing H₂ at higher pressure (thus high energy densities). Such geological hydrogen stores feature a porous and permeable reservoir formation, a caprock and a trap structure (Yekta et al., 2018; Heinemann et al., 2021; Muhammed et al., 2023). The injected hydrogen will displace the *in-situ* pore fluids, usually brine and/or residual hydrocarbons, and spread out underneath a low-permeable caprock capable of retaining the fluid (Zivar et al., 2021; Higgs et al., 2023).

Some recent studies on hydrogen have raised significant concerns regarding the integrity of storage sites, particularly concerning the risks associated with hydrogen embrittlement in surrounding rock formations and potential leakage pathways. For instance, researchers have utilized nuclear magnetic resonance imaging to analyze hydrogen saturation levels in reservoirs. This method helps in understanding how variations in pressure and temperature can impact the efficiency of hydrogen storage and retrieval (Al-Yaseri et al., 2022).

To reliably assess the behavior of hydrogen reliably in representative porous formations, it is crucial to observe in

situ the pore-space configurations of the fluids. Some studies have utilized pore-scale imaging techniques to understand how hydrogen migrates through and interacts with these geological formations. This approach has enabled the observation of trapped hydrogen inside rock otherwise saturated with an aqueous phase (Thaysen et al., 2023). These studies highlight the significant potential of the hydrogen trapping, which could hinder the efficiency of storage and retrieval.

Regarding hydrogen production, phenomenon in the pore space, known as Ostwald ripening, has the potential to lead to gas reorganization. The mechanism responsible for reconnecting trapped hydrogen gas is thoroughly investigated using high-resolution 3D micro-CT imaging techniques (Thaysen et al., 2023). In the work of Zhang et al. (2023), injection and withdrawal dynamics for hydrogen are analyzed in detail by micro-CT imaging techniques, comparing results for hydrogen with nitrogen experiments. Less trapping was observed with hydrogen because the initial saturation after drainage is lower due to channeling. Remarkably after imbibition, if the sample is imaged again after 12 hours, there is a significant rearrangement of the trapped hydrogen. Many smaller ganglia disappear while the larger ganglia swell, with no detectable change in overall gas volume. Compared with nitrogen, the fluid configuration is largely unchanged (see Fig. 3). This rearrangement, driven by concentration gradients of dissolved gas, is facilitated by Ostwald ripening, with estimated timescales consistent with experimental observations. Swelling of larger ganglia potentially enhances gas connectivity, resulting in reduced hysteresis and more efficient withdrawal.

By comparing pore-scale imaging findings for CO₂ and hydrogen storage, It is found that CO₂'s larger molecule size and higher reactivity with rock minerals necessitate a focus on chemical reactions and long-term mineral trapping, while hydrogen's smaller size and higher mobility require a more detailed examination of physical trapping mechanisms and the integrity of the storage formation. This comparative analysis highlights the need for tailored storage strategies for each gas, considering their unique properties and interactions at the pore level (Sordakis et al., 2018; Heinemann et al., 2021; Bai et al., 2024).

2.4 Microstructure and fracture propagation

The subsurface is increasingly being recognized as a critical component for mitigating the climate crisis, offering substantial storage capacity for seasonal energy and permanent waste disposal. However, subsurface characterization and prediction of its dynamic behavior is highly complex due to its multi-scale nature, intricate pore system, and complicated subsurface environment (Shi et al., 2017; Wang et al., 2017).

Rock microstructures are characterized and quantified from meter-scale to nano-scale even angstrom-scale, using multi-scale and multi-model approaches, including micro-CT imaging, focused ion beam scanning electron microscope, transmission electron microscopy tomography and atomic force microscopy and nano infrared spectrometers. Meanwhile, dynamic imaging of the rocks has provided the opportunity to characterize the thermo-hydro-mechanical-chemical coupling

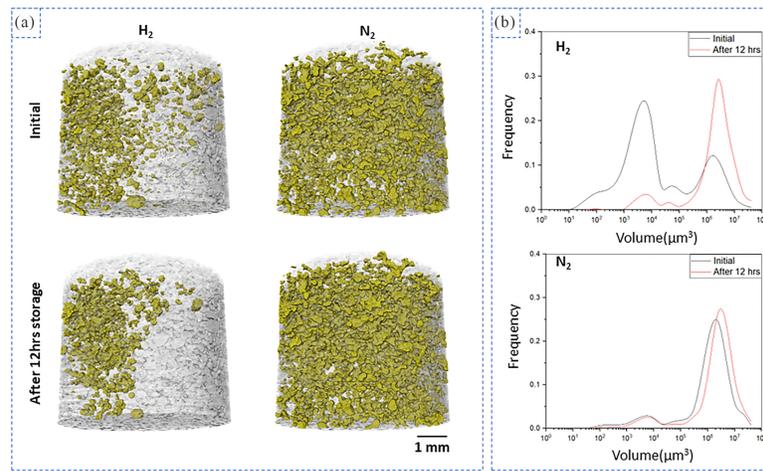


Fig. 3. (a) 3D images of the trapped gas ganglia before and after 12 hours for both the hydrogen and nitrogen experiment: yellow represents trapped gas ganglia. (b) The quantified ganglia size distributions are shown with equal bin sizes in logarithmic space: this is equivalent to a volume-weighted distribution. The area under the distributions remains the same, indicating no change in gas volume (Adapted from Zhang et al. (2023)).

mechanism of rocks in the complex subsurface conditions to investigate the storage efficiency and sealing ability under realistic reservoir conditions. These include high temperature (up to 1000 °C), high pressure (up to 65 MPa), fluids (diffusion and multi-phase flow) and complex chemistry environment (brine and injected fluids).

The advanced multi-scale and dynamic characterization approaches provide a critical foundation for unlocking the full potential of subsurface applications in energy decarbonization and waste disposal. By leveraging cutting-edge characterization techniques and a comprehensive understanding of the dynamic behaviors of complex subsurface environment, it can optimize energy storage efficiency and minimize environmental impacts while ensuring safe operations (Ma et al., 2016, 2019).

By means of time-lapse micro-CT (4D; 3D plus time) imaging, Ma et al. (2021) qualitatively and quantitatively investigates the 3D microstructure and pore system of the Bowland shale, and characterize the possible flow pathways and storage capacity of CH₄ and CO₂ in shales. Given the varied transport mechanisms of CH₄ and CO₂ in shale reservoirs, such as Darcy flow, surface diffusion, and molecular diffusion, dependent on pore size and network size, they further investigate shale samples at different scales. Based on the understanding, they proposed a flow pathways and storage mechanism shown as Fig. 4.

Initially, during CO₂ injection, high concentrations migrate from induced fractures to preexisting microcracks and macropores as free molecules. Subsequently, CO₂ infiltrates mesopores and micropores (<20 nm) via free molecules and surface diffusion, storing in the open pore space and adsorbing to the pore surface. As CO₂ concentration rises, CH₄ desorbs while CO₂ adsorbs, gradually penetrating smaller pores. Surface diffusion becomes predominant in finer pores. At equilibrium, CO₂ may be stored in shales via hydrodynamic trapping, adsorption to rock surfaces, solution trapping in

formation water, mineral trapping through precipitation, and residual or capillary trapping in immobile fractions. This study underscores the effectiveness of multi-scale 3D characterization in evaluating CO₂ storage feasibility in shale and enhancing gas recovery, suggesting its potential for further technical development to improve energy efficiency.

2.5 Microfluidic research on underground energy storage

Besides, experimental technology involving microfluidics on lab-on-a-chip systems offer an alternative means to visualize and investigate the influence of fluid properties and topological features of porous structures on fluid displacement. Microfluidics technology offer viable options for choosing materials, designing patterns, and treating surfaces. This shift allows studies to concentrate more on the fundamental characteristics of natural porous media. Consequently, micro/nanofluidic experiments conducted on lab-on-a-chip systems present a promising alternative for investigating the influence of fluid properties and topological features of porous structures on fluid displacement (Zhao et al., 2016). The insights gained from these experiments can be directly employed for validating and attempting to scale up mathematical or numerical models concerning observations of pore-scale fluid flow (Hu et al., 2018a, 2018b).

Microfluids offer enhanced detection resolutions of time (millisecond) and spatial scales (nano-level), allowing for intricate observations of pore-scale dynamics that enables the direct visualization of pore-scale phenomena in real time. Throughout the past few decades, microfluidic devices with porous media patterns representing porous rock have been used to study fluid flow for subsurface engineering. Images or 3D models of actual porous rock can be generated using micro-CT, focused ion beam scanning electron microscope or nuclear magnetic resonance methods (Alzahid et al., 2018). These images can then be transformed to a microfluidic

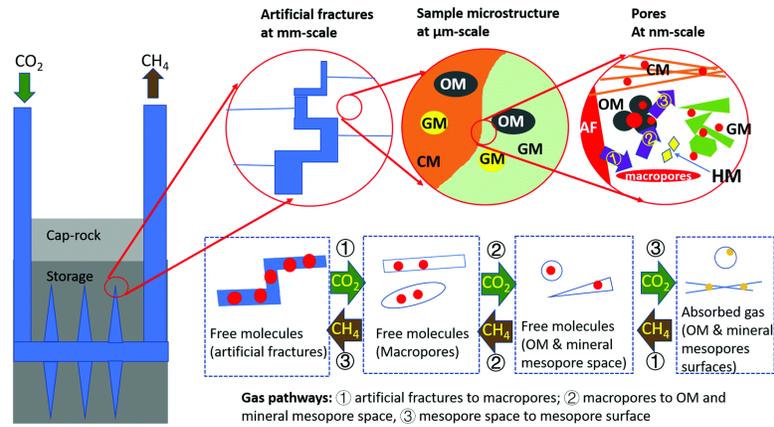


Fig. 4. Schematic diagram of the storage space and flow pathways for CO₂ and CH₄ (Adapted from Ma et al. (2021)).

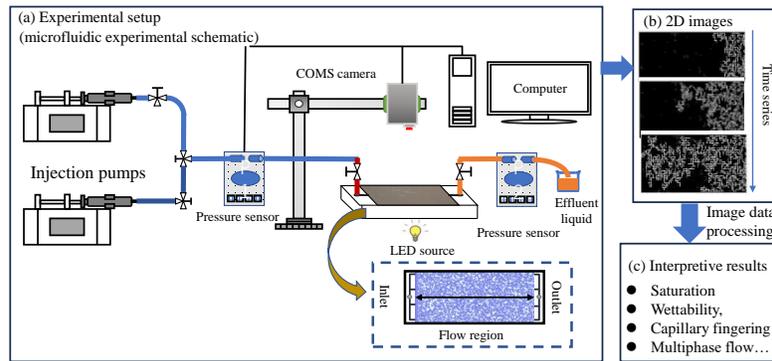


Fig. 5. (a) A typical microfluidic experiment utilizes a high-resolution camera and a light-emitting diode lighting positioned both above and below a flow cell, (b) following the acquisition of image dataset, a time series of 2D images is generated and (c) these images undergo subsequent processing and analysis to extract specific properties.

chip using standard photolithography or soft photolithography. These micromodels allow for the direct visualization of flow behavior through a two-dimensional porous medium that is geometrically representative of a real rock (Wen and R, 2015).

A basic microfluidic setup commonly consists of a pump, microfluidic device, pressure controllers and visualization system (Fig. 5). The fluid is injected into the chip pattern through the inlet tube where it displaces/interacts with resident fluid. By utilizing syringe pumps, high-pressure pumps, pressure transducers, flow sensors, pressure cells, heaters, etc., the setup can be modified for specific applications (Qiu et al., 2023). After acquiring the image dataset, a time series of 2D images is produced. These images are then subjected to further processing and analysis to extract specific properties, including fluid saturation and wettability characterization, with application in porous media and underground energy storage. Overall, the real-time visualization, quantification and/or control of flow and/or transport in the micromodel are considered a significant advantage of microfluidic studies and applications (Cottin et al., 2011; Lv et al., 2023). In particular, the visualization method is critical for a successful microfluidic experiment and can be used as both a qualitative and quantitative tool (Xu et al., 2014; Singh et al., 2017; Tanino et al., 2018).

However, microfluidic models generally do not allow for the connectivity of corners and crevices due to their 2D nature,

in addition to their lack of roughness, which can limit the type of flow processes that occur during immiscible displacement experiments.

3. Conclusions

Advancements in pore-scale imaging technology are expected to provide even more detailed insights into dynamic processes of underground energy storage which is an excellent tool as it can provide valuable 3D pore-scale information. Of particular importance are *in-situ* observations, which give direct evidence of the various phenomena occurring.

However, pore-scale imaging technology also pose many challenges, such as the need for high-resolution imaging without compromising sample size, the difficulty in interpreting the dynamic behavior of gases within these pores, and the complexity of correlating laboratory-scale observations to field-scale phenomena. A significant challenge lies in translating these pore-scale observations to predict large-scale field behavior accurately. Bridging this gap requires not only technological advancements but also the development of sophisticated modeling approaches that can integrate pore-scale data into field-scale simulations.

Overview, pore-scale imaging has emerged as a vital tool in advancing our understanding of underground gas storage for

both CO₂ and H₂. The insights gained from these imaging techniques are critical in optimizing storage strategies, ensuring safety, and enhancing the efficiency of these storage solutions. As moving towards a more sustainable and carbon-neutral future, the role of pore-scale imaging in underground gas storage will only grow in importance, guiding both technological and policy decisions in this crucial sector.

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Conflict of interest

The authors declare no competing interest.

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