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Perspective

A molecular perspective on the microscopic mechanisms of CO_2 injection and water films in fluid transport and enhanced oil recovery

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Abstract:
Molecular dynamics simulation has emerged as a powerful tool to shed light on the fundamental mechanisms that govern fluid behavior across multiple phases, such as gas, liquid and solid, at the molecular-scale. In shale reservoirs, understanding nanoscale phenomena such as microscopic friction, oil-gas interactions during CO_2 huff-n-puff processes, and the influence of co-solvents, is crucial for enhanced oil recovery and informing strategies for CO_2 geological sequestration in shale. This paper explores the unique role of molecular dynamics simulations in revealing the microscopic mechanisms of fluid transport and enhanced recovery during CO_2 injection, with particular attention to the effects of hydration film, CO_2 affected areas and co-solvents. The regulatory mechanism of hydration films on the friction behavior of montmorillonite provides new insights into interfacial mechanics, with implications for the mobility of confined fluids. In tight reservoir systems, the microscopic oil recovery mechanisms of CO_2 under varying sweep conditions water film thicknesses highlight the complexity of fluid displacement at the nanoscale. Furthermore, the co-injection of CO_2 with selected co-solvents is shown to enhance both oil mobilization and carbon storage efficiency within shale nanopores, offering a promising pathway to improve recovery outcomes under diverse reservoir conditions. By providing a molecular-level understanding of these critical processes, this work lays the groundwork for bridging atomistic insights with field-scale applications for unconventional resources development.

1. Introduction

With the global demand for shale oil and gas continuing to rise alongside escalating CO_2 emissions, Carbon Capture, Utilization, and Storage (CCUS) has gained traction as a promising pathway to balance energy production with climate mitigation goals (Guo et al., 2024). Shale formations are characterized by well-developed nanoporous structures with hybrid organic-inorganic compositions (Kuila et al., 2013; Pearce et al., 2022) where the behavior of confined fluids (water, oil/gas, CO_2, co-solvents) is governed by intricate interactions at the molecular scale. These systems pose unique challenges, as fluid-rock dynamics in nanoconfined environments – subject to variations in temperature, pressure, and mineral surface properties – show a dominant influence on macroscopic outcomes

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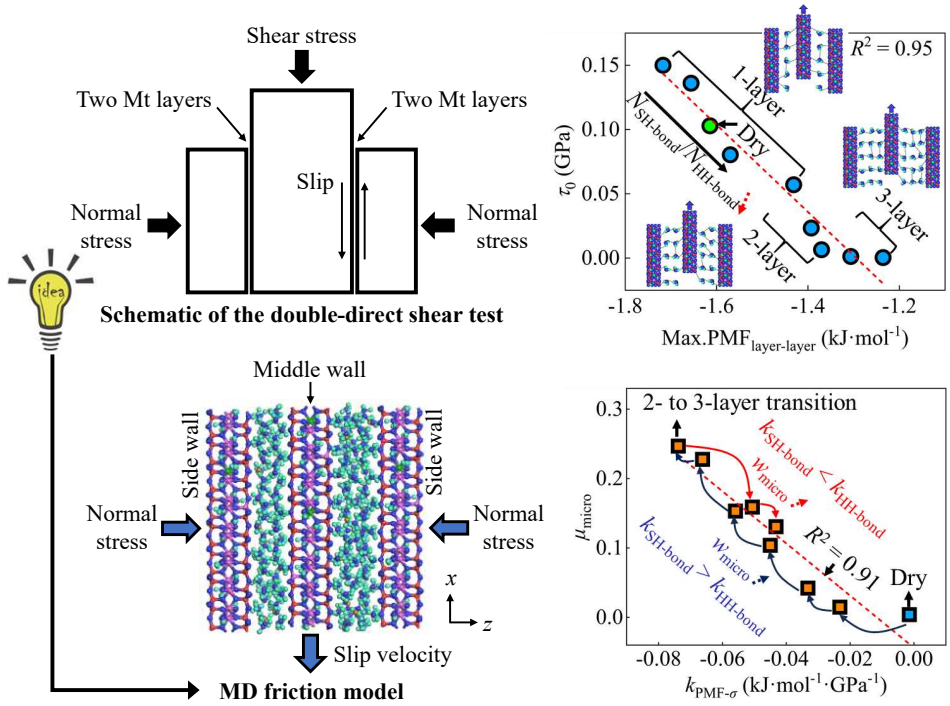

Fig. 1. The effects of hydration film on the microscopic frictional properties of montmorillonite (Du et al., 2024).

(Yang et al., 2020; Cai et al., 2024; Zhan et al., 2025; Zhou et al., 2025). Molecular Dynamics (MD) simulation has become an essential tool for exploring such nanoscale phenomena. By enabling atomistic modeling of fluid-solid interactions, MD fills a critical gap where experimental techniques are limited by resolution, accessibility, and replicability. It has been widely used to investigate fundamental mechanisms controlling the occurrence state and transport behavior during the Enhanced Oil Recovery (EOR) and CCUS in shale system (Zhan et al., 2024a; Zhan et al., 2024b).

In this work, a molecular-level investigation into three interrelated phenomena of critical relevance to unconventional reservoir development: (1) frictional behavior on mineral surfaces under hydration, (2) CO_2-induced oil recovery mechanisms in shale during huff-n-puff processes, and (3) the role of co-solvents in enhancing CO_2 performance for both shale oil recovery and carbon sequestration. By using MD simulations, this study aims to shed light on the fundamental mechanisms governing these processes, providing new insights to guide the design of more effective and efficient strategies for shale resource development and carbon sequestration.

2. Hydration film regulation of nanoscale friction in montmorillonite

Frictional behavior between layered clay minerals, such as montmorillonite, plays a crucial role in controlling fluid transport, permeability evolution, and fracture propagation in shale reservoirs. These factors are central to both shale oil recovery and the long-term integrity of carbon geological sequestration. The fricition behavior is typically described by Amonton's law, which assumes a linear relationship between normal load and frictional force in macroscopic scale (Mo et al., 2009), while this approximation may breaks down at the nanoscale, when the adhesive force dominate interfacial interactions. In subsurface environments, water molecules are often confined between clay layers and appear as nanometer-thick hydration films. These films adhere strongly to mineral surfaces, resist compression, and dynamically respond to shear. This hydration lubrication effect substantially reduces sliding friction and modulates particle-scale deformation and rearrangement (Ma et al., 2015). Such effects can significantly influence fracture closure, caprock sealing, and hydrocarbon mobility in tight formations.

Conventional experimental tools such as atomic force microscopy or surface force apparatus offer limited insight into hydration film structure and its evolution with water content. It's observed that the hydrated cations can enhance lubrication silica surfaces by the experiments, highlighting the complexity of hydration-mediated friction (Donose et al., 2005). Yet, how hydration film structure influences adhesive force and friction coefficients in realistic shale-like environments remains poorly understood, especially under variable water saturation typical of both EOR and CCUS.

Recent MD simulations have revealed that the frictional behavior of montmorillonite is co-regulated by the structural state of the hydration film and the strength of hydrogen bonding cross clay surfaces (Du et al., 2024). As shown as Fig. 1, the microscopic friction coefficient exhibits a two-stage behavior with respect to water content. A critical transition occurs when the hydration film evolves from a bilayer to a trilayer structure, beyond which increased water weakens adhesion and reduces friction. This transition not only alters interfacial shear resistance but also affects the ease of clay layer sliding, which affects the pore network connectivity and fluid accessibility in

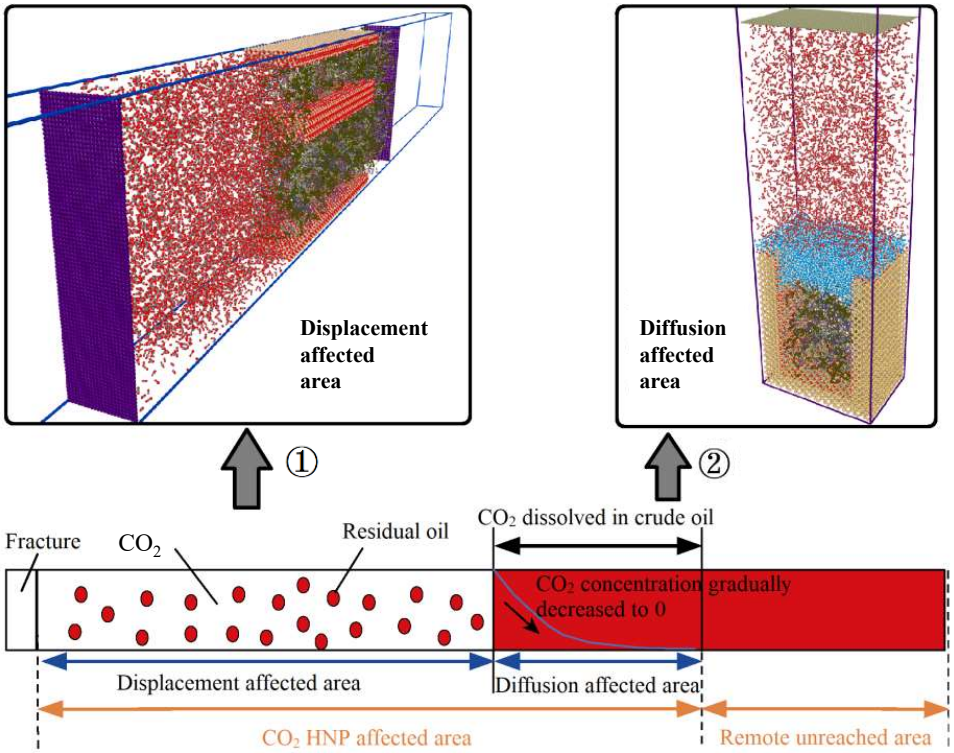

Fig. 2. Schematic of the displacement-affected area and the diffusion-affected area during the CO_2 huff-n-puff (Luo et al., 2023).

shale formations dominantly. Understanding this mechanism provides a molecular-level foundation for tuning water content in hydraulic fracturing fluids and CO_2-water mixtures to optimize subsurface flow behavior. It also informs that the design of injection strategies where controlling interfacial slip, matrix compaction, or fracture conductivity is essential.

3. Microscopic oil recovery mechanisms of CO_2 in different affected areas of tight reservoirs

Tight oil reservoirs are generally characterized by strong heterogeneity, ultra-low permeability and porosity, narrow pore throats and complex mineralogy. Despite the deployment of advanced techniques such as horizontal drilling and large-scale hydraulic fracturing, the primary recovery factor often remain below 10%, leaving vast quantities of crude oil trapped within the tight formations (Hoffman, 2018; Jia et al., 2019). Due to the inefficacy of conventional waterflooding in tight oil reservoirs, CO_2 injection has emerged as a leading EOR technique, offering both miscibility benefits and potential for in situ carbon storage. Understanding the microscopic mechanisms of CO_2-oil displacement in these environments is therefore critical to advancing EOR performance.

However, the complexity of nanoscale pore networks in tight reservoirs poses significant challenges for experimental characterization. The behavior of fluids within nanopores, especially across different CO_2-affected areas, remains poorly resolved (Feng et al., 2020). Previous studies have often relied on simplified systems involving single-component oil, which fail to capture the compositional complexity and phase behavior of actual crude oil. In reality, the interaction between CO_2 and multi-component oil systems can strongly influence phase transition, interfacial tension, and overall displacement efficiency (Moh et al., 2022; Zhang et al., 2024). Furthermore, the role of interfacial water films, which is common in water-wet shale systems, has been largely overlooked, despite their influence on both wettability and CO_2 diffusion pathways.

To address this gap, a conceptual framework can be introduced into the CO_2 huff-and-puff based on recent MD simulations (Luo et al., 2023), in which the CO_2-affected area can be divided into two distinct regions: the displacement-affected area and the diffusion-affected area, as illustrated in Fig. 2. The microscopic oil recovery mechanisms within distinct different CO_2-affected areas and under varying water film thicknesses can be explained in detail. While actual pore conditions are far more complex than those captured in the simulation, the phenomena observed based on MD could offer valuable explanations for field-scale behaviors.

4. Mechanism of cosolvent-enhanced CO_2 for improving shale nanopore crude oil recovery and carbon sequestration

Shale reservoirs are characterized by strong heterogeneity, extensive development of micro- and nanopores, complex oil occurrence states, and rapid decline in single-well which all pose significant challenges to efficient exploitation. Studies have confirmed that CO_2 can enhance shale oil recovery while simultaneously enabling geological carbon storage through various physicochemical mechanisms (Wang et al., 2023b). Supercritical CO_2, in particular, exhibits high injectivity in tight shale reservoirs and can rapidly diffuse through micro-

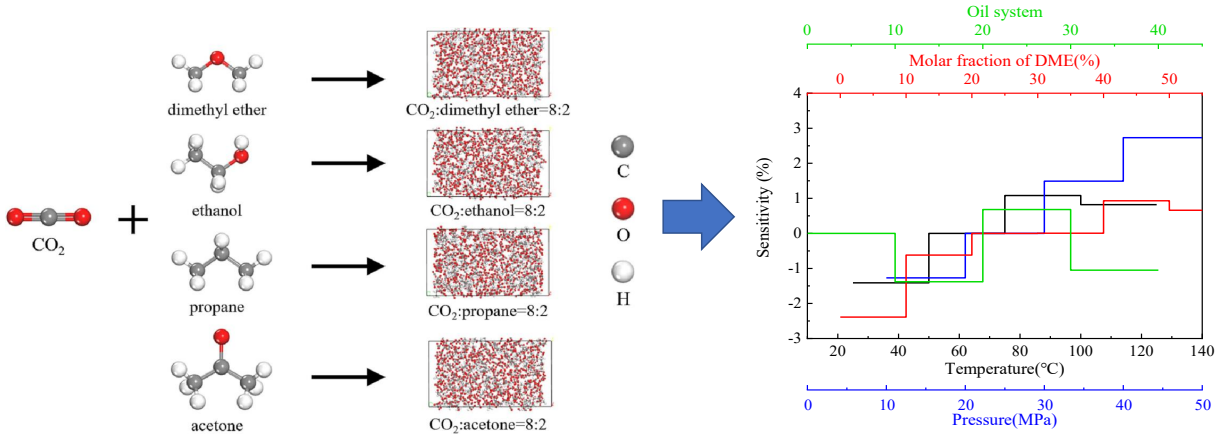

Fig. 3. Sensitivity of EOR effect to different influencing factors (Wang et al., 2023a).

and nanoporous media. However, in many shale reservoirs, the minimum miscible pressure of CO_2 exceeds the fracture pressure of the rock, and the presence of heavy hydrocarbons and colloids in the crude oil further complicates the displacement process (Zhao et al., 2021; Wang et al., 2024). These reservoir and fluid characteristics inhibit the solubility and miscibility of CO_2, making CO_2-EOR mechanisms highly complex and sensitive to operation conditions.

Recent advances suggest that introducing cosolvents, such as alcohols, ketones, esters, ethers, surfactants, and other nonpolar compounds can enhance the miscibility of CO_2 with crude oil and improve recovery efficiency (Gong et al., 2019). However, existing investigations on cosolvent-assisted CO_2-EOR remain fragmented and lack systematic evaluation. The relative importance of different influencing factors has not been fully ranked, nor has the governing rules that define each factor's impact on different CO_2-EOR mechanisms been clearly identified. Moreover, the specific influences of cosolvent-oil interactions across various shale oil components remain insufficiently understood. Although cosolvents can help mitigate gas channeling and improve sweep efficiency, their applicability and performance under diverse shale oil reservoir conditions still require further investigation.

To address this gap, the microscopic mechanisms for both EOR and enhance CO_2 sequestration when cosolvent mixtures with CO_2 injected could be explained using MD simulations (Wang et al., 2023a). As illustrated in Fig. 3. It is identified that the advantages of such mixtures across a range of reservoir conditions, including different temperatures, pressures, and oil compositions. These findings offer a molecular-level understanding of cosolvent-assisted CO_2-EOR and provide theoretical guidance for optimizing injection strategies in shale formations, contributing to both efficient resource development and long-term carbon storage.

5. Conclusions

This perspective emphasizes the critical role of microscale mechanisms, including frictional behavior at mineral interfaces, CO_2-induced oil recovery dynamics, and cosolvent-enhanced interactions, for enhancing shale oil production and advancing carbon geological sequestration. Molecular dynamics simulation serves as a powerful tool to probe solid-liquid interfacial phenomena at the micro- and nano-scales, offering unique insights into processes that are difficult to capture experimentally. By revealing how hydration films modulate nanoscale friction, how CO_2 interacts with multi-component oil across different pore environments, and how cosolvents improve miscibility and sweep efficiency, a theoretical foundation for optimizing enhanced oil recovery strategies and secure CO_2 storage could be revealed by molecular dynamics simulation. These microscale insights not only inform the design of reservoir-specific injection schemes but also offer the potential to be upscaled into predictive macroscopic models. Such integration of molecular understanding into field-scale applications paves the way for more efficient and sustainable development of unconventional energy resources.

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