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Perspective

Advances in multiscale atomistic modelling for enhanced oil and gas recovery and CO_2 sequestration

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Abstract:
A thorough understanding of gas-fluid interactions, competitive adsorption mechanisms, transport behaviour, and reactive transformations is fundamental for improving enhanced oil and gas production and ensuring permanent CO_2 storage. The combined application of classical and ab initio molecular dynamics, grand-canonical Monte Carlo, and emerging machine learning approaches enables a comprehensive elucidation these mechanisms across multiple spatial and temporal scales. This view integrates our recent multiscale modelling efforts with state-of-the-art computational techniques to chart an agenda for predictive subsurface workflows.

1. Introduction

Injecting CO_2 into reservoirs serves the dual purpose of boosting hydrocarbon recovery and sequestering carbon in deep formations (Fentaw et al., 2024). The success of these operations depends critically on microscale processes: CO_2 must displace oil and/or gas within water-containing nanopores, alter mineral wettability under reservior brine conditions, and undergo reactive interactions with the host rock while maintaining the integrity of the sealing formation (Gbadamosi et al., 2019). Experimental techniques alone cannot capture the rapid, confined nanoscale phenomena occurring under reservoir pressures and temperatures. Atomistic simulations bridge this gap by providing detailed insights into interfacial interactions, transport mechanisms, and reaction pathways in reservoirs.

2. Current state-of-the-art

2.1 Classical molecular dynamics coupled grand-canonical Monte Carlo

The sequential grand-canonical Monte Carlo and molecular dynamics (GCMC-MD) workflow has proven effective for evaluating both gas adsorption and transport behaviour in organic matter and silica nanopores. In this approach, GCMC simulations are first used to determine equilibrium gas loading at given pressure and temperature conditions. The resulting configurations are then passed to classical MD to investigate molecular diffusion, density distributions, and interfacial structural changes.

In our 2019 study (Shi et al., 2019), we examined mixed silica-organic matter slit pores at 313 K. CH_4 was initially adsorbed at 15 MPa using GCMC, followed by stepwise introduction of CO_2 and N_2 up to 100 MPa. The simulations showed that CO_2 preferentially displaces CH_4 from the central slit region, with subsequent removal from both matrix surfaces. MD trajectories confirmed that these newly vacated regions serve as diffusion pathways for the remaining CH_4, highlighting the importance of spatial desorption sequence in

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controlling transport.

Our 2020 work focused on dry and water-bearing kerogen II-D models (Gong et al., 2020). GCMC simulations showed that introducing 0.6-2.4 wt% water significantly reduced CH_4 uptake from 1.2 to 0.6 mmol/g due to water occupying adsorption sites. These results indicate that water molecules can form clusters within accessible pores and block key adsorption regions. MD simulations further demonstrated that CH_4 diffusivity decreases with increasing water content, while higher temperatures improve diffusion. Under reservoir-like conditions (358 K, 2.4 wt% water), the system exhibited favourable transport behaviour, suggesting an optimal trade-off between adsorption loss and enhanced mobility.

In our 2024 study (Shi et al., 2024), we extended this framework to new built kerogen models III-C, and III-B. GCMC simulations showed that type II-D exhibits the highest CO_2 selectivity over CH_4 across a range of pressures (0-20 MPa). Following MD simulations revealed that CH_4 diffusion varies by region: the central slit in II-D supports the fastest transport, while the more aromatic, structurally compact III-B and III-C kerogens impose stronger confinement, reducing diffusivity accordingly. These findings underscore how composition of shale organic matter and pore geometry jointly influence gas behaviour in organic rich shale systems.

2.2 Ab initio molecular dynamics

To capture the electronic-level mechanisms of CO_2 adsorption and water-mediated reactions in amorphous silica nanopores, we performed ab initio molecular dynamics (AIMD) using the generalized gradient approximation functional (Perdew et al., 1996) with dispersion correction (Grimme, 2006) and a double-ζ polarized Gaussian basis (VandeVondele and Hutter, 2007) as implemented in CP2K version 9.1 software package (Kühne et al., 2020). A 17 Å natural SiO_2 slit model contaning 15 wt% preadsorbed water was simulated in the NVT ensemble at 310 K upto 15 ps.

In our AIMD simulations of hydrated silica nanopores, confined water was found to perform a dual function: first, it undergoes rapid Grotthuss-type proton shuttling at under-coordinated silicon sites, generating transient $OH^-{-}H_3O^+$ complexes (O-O $\sim$ 2.4-2.5 Å) and freeing hydroxide ions that bind to the siloxane framework to create new surface silanol (Si-OH) groups (Shi et al., 2025); second, the same hydrogen-bond network that drives these proton transfers also acts as a reactive medium for CO_2 capture, facilitating the conversion of solvated CO_2 into carbonic acid (H_2CO_3) and its subsequent deprotonation to chemisorbed carbonate (${CO_3}^{2-}$) species coordinated to both silanol and siloxane oxygen atoms.

3. Challenges: From atomic to field scales

Despite advances in GCMC, MD, AIMD, and preliminary machine learning application (Zhang et al., 2024), significant bottlenecks remain in translating atomistic insights into robust field-scale CO_2 sequestration and enhanced shale oil and/or gas recovery strategies. Based on our integrated studies, three interlinked categories of challenges can be clearly identified:

3.1 Force field limitations

Most classical empirical force fields, such as COMPASS (Sun, 1998), ClayFF (Cygan et al., 2004), and TraPPE (Martin and Siepmann, 1998), lack the ability to capture reactive pathways involving CO_2 dissociation and mineral–fluid surface reactions. For example, in our AIMD work on hydrated silicate nanopores (Shi et al., 2025), we observed that CO_2 hydrolysis ($CO_2 + H_2O \rightarrow H_2CO_3 \rightarrow HCO_3^- + H^+$) phenomenon, but when simulated with COMPASS, this empirical force fields are not sufficient to observe microscopic bond breaking and building processes and describe the reactive interfaces. Fixed-charge force fields fail to account for local charge redistribution when water molecules adsorb onto kerogen or mineral surfaces. Water clusters can induce dipole moments on kerogen carbon sites, creating new hydrophilic sites that enhance CO_2 and CH_4 binding. Ignoring this effect can produce adsorption heat errors, which highlights the urgent need for polarizable or machine learning-based reactive potentials to describe fluid-solid interfacial charge transfer accurately.

3.2 Scale-integration bottlenecks

Processes like CO_2 mineralization and water-mediated carbonate formation were directly observed via AIMD within several tens of ps, but real reservoir stability must be projected over 100-1,000 years. There is still no mathematically rigorous framework to extrapolate short-time, atomistic reaction rates to predict long-term storage security under variable stress and brine composition. The quantum to continuum timescale bridging is still lacking. Meanwhile, microfractures and mesoporous networks (> 1 μm) dominate real flow behaviour during CO_2 injection. Current multiscale methods cannot yet couple nanoscopic chemical reactivity with mesoscopic percolation and fracture dynamics in a unified framework.

3.3 Missing multiphysics coupling

Current molecular simulations primarily address fluid-solid interactions under static or quasi-static conditions, often neglecting critical multiphysics feedbacks inherent in practical CO_2 injection and storage processes. Our MD and AIMD results demonstrate that CO_2 adsorption and water clustering can induce local swelling of organic matter or clay by 5-15% (Gong et al., 2020), thereby altering pore volume and connectivity in ways that significantly affect gas transport. However, standard MD approaches are unable to couple such swelling behaviour with stress redistribution or fracture propagation. Furthermore, during dynamic injection, local thermal fluctuations, chemical reactions (e.g., CO_2 hydration and carbonate precipitation), and evolving pore pressures interact nonlinearly. Yet, most GCMC and classical MD simulations assume constant temperature and rigid pore geometries, omitting such complex couplings. As a result, while local adsorption behaviour can be captured, these models fail to account for how chemical and mechanical responses jointly reshape pores scale flow paths and influence long-term storage integrity. Notably, during CO_2 injection and CH_4 displacement, the transient local adsorption states captured by MD do not always align with the instantaneous equilibrium profiles predicted by GCMC. This highlights the role of kinetic delays and local trapping in deviating from ideal equilibrium assumptions. Bridging atomistic insights with continuum scale models of geomechanics, heat transfer, and reactive transport remains a major challenge in accurately predicting reservoir behaviour.

4. Conclusion

Microscopic-scale simulations, including GCMC, MD, AIMD, and machine learning methods, have played an increasingly important role in studying CO_2 sequestration and enhanced oil and gas recovery. Through these methods, important insights have been gained into how pore size, water content, and mineral or organic surface structure influence gas adsorption, diffusion, and chemical reactions under reservoir conditions.

Our work demonstrates that GCMC and MD are effective in describing the competitive adsorption and transport behaviour of CH_4 and CO_2 in kerogen and shale systems, particularly when water is present. AIMD simulations further show that confined water not only affects gas distribution but also participates in surface reactions, such as silanol formation and carbonate precipitation, which are not captured by conventional force fields. These findings highlight the need to consider both physical and chemical effects in modelling subsurface processes.

To move forward, more accurate and transferable force fields, especially those incorporating reactivity or trained by machine learning, will be essential. At the same time, new modelling frameworks are needed to connect atomic-scale simulation results with larger-scale flow and geomechanical models. Combining simulation with experimental measurements under realistic reservoir conditions will also be important for validation. These developments will help ensure that microscopic modelling continues to support the practical design and evaluation of CO_2 enhanced oil and gas recovery and CO_2 storage technologies.

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Conflict of interest

The authors declare no competing interest.

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