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Perspective

Caprock wettability: A cross-process perspective from CO_2 sequestration to underground hydrogen storage

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Abstract:
Caprock wettability is a fundamental control on the sealing capacity and integrity of subsurface energy storage systems. In CO_2 geological sequestration, a strongly water-wet caprock ensures high capillary entry pressures, effectively preventing CO_2 migration. However, increased pressure and brine salinity can weaken hydrophilicity and compromise long-term sealing. In underground hydrogen storage, caprocks generally remain water-wet, yet the high diffusivity of H_2 reduce capillary sealing efficiency and may induce wettability alteration due to microbial or redox processes. Repeated injection-withdrawal cycles further cause transient wetting-drying and hysteresis, altering interfacial structures and capillary behavior. Understanding these dynamic wettability responses under varying physicochemical conditions is crucial for assessing storage security. Future studies should integrate in-situ characterization and molecular modeling to reveal reactive and reversible wettability mechanisms, providing a unified framework for CO_2 and H_2 storage systems.

1. Introduction

Caprock serves as the primary seal in subsurface gas storage systems, providing a barrier that prevents the plume upward migration and thereby ensuring the long-term integrity of the reservoir. The sealing efficiency of caprocks is influenced by their wettability, which controls capillary entry pressure and the stability of fluid interfaces across the storage–seal boundary (Fig. 1). In the context of geological CO_2 sequestration, extensive research has been devoted to understanding how pressure, temperature, mineral composition, and fluid chemistry affect caprock wettability and capillary sealing behavior. In contrast, the emerging field of underground hydrogen storage introduces additional physicochemical complexities due to the smaller molecular size and higher diffusivity of H_2 (Al-Yaseri and Jha et al., 2021). Moreover, dynamic variations in caprock wettability during repeated gas injection and withdrawal cycles remain poorly understood but are crucial for evaluating the long-term containment security and reversibility of these systems.

2. Wettability in CO_2 geological sequestration

In CO_2 sequestration, the caprock is ideally strongly water-wet, ensuring high capillary entry pressures that prevent CO_2 migration (Rücker et al., 2020). However, under elevated pressure, CO_2 can penetrate the pore network of the caprock, partially dissolve in the aqueous phase, and chemically interact

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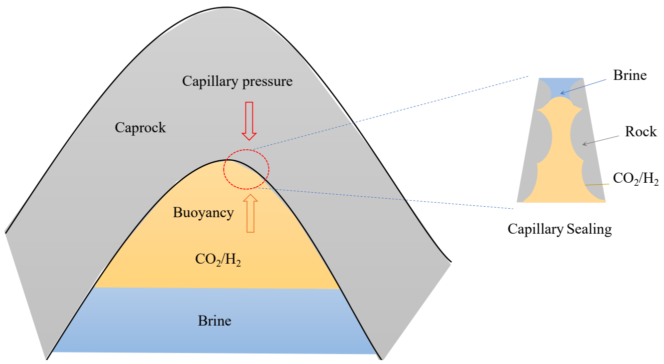

Fig. 1. Influence of caprock wettability on CO_2 sequestration and underground hydrogen storage.

with the pore surfaces of the rock, thereby inducing a gradual shift in rock wettability from strongly water-wet toward less water-wet or CO_2-wet (Alnili et al., 2018). Currently, a wide body of studies primarily investigates the influences of factors such as pressure conditions, mineral composition, fluid chemical properties, chemical reactions, and pore structure on rock wettability.

Studies on clay-rich caprocks (e.g., illite, smectite, and kaolinite) indicate that salinity and pressure can induce partial hydrophobicity due to CO_2 adsorption and desorption at the mineral surface. Such wettability alteration has been shown to reduce the capillary sealing efficiency of the caprock, as the transition toward CO_2-wet conditions lowers the capillary entry pressure and enhances CO_2 percolation through pore networks (Iglauer et al., 2015). Therefore, CO_2 sequestration security depends on the preservation of water-wet conditions under CO_2-brine-mineral equilibria, which may evolve over time due to physicochemical alterations.

3. Wettability in underground hydrogen storage

Geological formations suitable for underground hydrogen storage include depleted oil and gas reservoirs, salt caverns, and aquifers (Zhu et al., 2024). The capillary sealing capacity of the caprock is primarily governed by interfacial properties, including interfacial tension and wettability. In general, the interfacial tension of the hydrogen-brine-rock system increases with increasing temperature and brine concentration but decreases with increasing pressure. In contrast, the contact angle decreases with increasing temperature, and this trend becomes more pronounced at higher pressures (Zhu et al., 2023). Beyond these general trends, the mineralogical composition of caprocks strongly modulates wettability behavior in H_2 storage settings. Clay-rich caprocks (e.g., illite, kaolinite, smectite) tend to remain strongly water-wet due to abundant surface hydroxyl groups and high cation-exchange capacity. Their surface charge and hydration layers can stabilize thin water films around pore walls, suppressing hydrogen spreading and maintaining high capillary entry pressures. In contrast, carbonate minerals such as calcite may experience surface charge reversal or localized dissolution/precipitation under brine-gas interactions, which can alter surface roughness and modify contact angles. Quartz-dominated caprocks are generally hydrophilic, yet their wettability can be influenced by ionic strength-driven changes in the electrical double layer, potentially reducing water-wetness under high-salinity brines. Geochemical reactions induced by subsurface microorganisms can further reduce the contact angle (Esfandyari et al., 2025). For practical applications, as the storage depth increases, both the pressure and temperature within the underground hydrogen storage system rise accordingly. With this complex coupling effect, the capillary sealing capacity of the caprock is reported to be strongest at a depth of approximately 1,100 m, and weakest at around 3,700 m (Iglauer, 2022).

CO_2 and H_2 exhibit fundamentally different controls on caprock wettability due to their contrasting physicochemical behaviors. As discussed in Section 2, CO_2-brine-rock interactions under elevated pressures can induce wettability alteration. Such alterations are particularly significant in clay-rich caprocks, where CO_2 adsorption-desorption processes can reduce capillary sealing efficiency. In contrast, wettability in H_2 storage systems is governed mainly by physical rather than reactive mechanisms. H_2-brine interfacial tension increases with temperature and salinity but decreases with pressure, while contact angles generally decrease with temperature, reinforcing water-wetness. Due to its low solubility, H_2 causes minimal modification to mineral surfaces, and intrinsic mineral wettability dominates system behavior. Clay-rich caprocks remain strongly water-wet, whereas carbonates and quartz exhibit only limited changes related to surface charge or ionic strength. These distinctions show that CO_2 systems undergo chemically driven wettability alteration, whereas H_2 systems preserve more stable water-wet conditions.

4. Dynamic wettability during injection and withdrawal

During both CO_2 and H_2 injection, nonwetting phase invasion can displace brine and locally alter surface hydration structures. During the storage, the evaporation-induced salt precipitation can change pore wall microstructures, then affect wettability (Liu and Lei, 2023). Cyclic injection-withdrawal in H_2 storage leads to wetting-drying changes in charge or hydration state of mineral surface, resulting in in the variation of contact angles, which can complicate wettability heterogeneity and influence subsequent fluid flow and storage efficiency (Liu et al., 2025; Zhang et al., 2025).

Furthermore, the induced variations in wettability along the caprock-reservoir interface can significantly reduce the critical capillary entry pressure and facilitate the development of preferential gas pathways, thereby increasing the risk of gas leakage. Over time, repeated injection-withdrawal cycling may further compromise the caprock's effective sealing capacity and long-term integrity, particularly when wettability alteration leads to a decline in the capillary pressure threshold. Therefore, understanding the effect of dynamic wettability and its coupling with pore-scale flow, dissolution/precipitation phenomenon on caprock wettability alteration is vital for long-term gas sequestration/storage.

5. Outlook

Across CO_2 and H_2 storage systems, the common challenge lies in maintaining or restoring water-wet condition under fluctuating pressure, chemical, and saturation regimes. Future researches should focus on:

(1) In-situ characterization of dynamic wettability using advanced nuclear magnetic resonance, synchrotron, or atomic force microscopy methods under reservoir conditions.

(2) An exploration of the restoration effectiveness of caprock re-wettability, for instance, via the injection of surfactants or specific ion-composition brines capable of reacting with caprock minerals to generate water-wet mineral components.

(3) Molecular-level simulations to reveal hydrogen bonding, ion hydration, and gas adsorption mechanisms at mineral surfaces.

(4) Comparative frameworks linking CO_2 and H_2 wettability behaviors to develop unified models for gas-brine-rock interactions.

Overall, the long-term security of subsurface energy storage largely depends on the caprock's capacity to regain its hydrophilic state following repeated gas exposure.

6. Summary

Caprock wettability is a unifying yet evolving control factor on the integrity of CO_2 sequestration and underground H_2 storage. While injected CO_2 challenges the hydrophilic stability through adsorption and dehydration, H_2 introduces dynamic and cyclic effects that may weaken capillary sealing performance over time. Bridging insights from both systems will advance our understanding of wetting behavior in CO_2/H_2-brine-rock systems, which forms the foundation for safe and sustainable subsurface energy storage.

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Conflict of interest

The authors declare no competing interest.

References

Alnili, F., Al-Yaseri, A., Roshan, H., et al. Carbon dioxide/brine wettability of porous sandstone versus solid quartz: An experimental and theoretical investigation. Journal of Colloid and Interface Science, 2018, 524: 188-194.

Al-Yaseri, A., Jha, N. K. On hydrogen wettability of basaltic rock. Journal of Petroleum Science and Engineering, 2021, 200: 108387.

Esfandyari, H., Jozani, R. J., Hassanpouryouzband, A., et al. The microbial factor in subsurface hydrogen behavior: Implications for wettability and interfacial dynamics. Advances in Colloid and Interface Science, 2025, 346: 103647.

Iglauer, S. Optimum geological storage depths for structural H_2 geo-storage. Journal of Petroleum Science and Engineering, 2022, 212: 109498.

Iglauer, S., Pentland, C. H., Busch, A. CO_2 wettability of seal and reservoir rocks and the implications for carbon geo-sequestration. Water Resources Research, 2015, 51(1): 729-774.

Liu, L., Jiang, Y., Gao, X., et al. Exogenous and endogenous displacements in 3D porous media. Geophysical Research Letters, 2025, 52(17): e2025GL117416.

Liu, L., Lei, L. Contact angle on rough curved surfaces and its implications in porous media. Langmuir, 2023, 39(12): 4507-4517.

Rücker, M., Bartels, W., Garfi, G., et al. Relationship between wetting and capillary pressure in a crude oil/brine/rock system: From nano-scale to core-scale. Journal of Colloid and Interface Science, 2020, 562: 159-169.

Zhang, H., Mahmoud, M., Iglauer, S., et al. Field-scale investigation of CO_2 plume dynamics under spatial wettability variations: Implications for geological CO_2 storage. Advances in Geo-Energy Research, 2025, 15(3): 230-244.

Zhu, S., Shi, X., Yang, C., et al. Hydrogen loss of salt cavern hydrogen storage. Renewable Energy, 2023, 218: 119267.

Zhu, S., Shi, X., Yang, C., et al. Site selection evaluation for salt cavern hydrogen storage in China. Renewable Energy, 2024, 224: 120143.