Fingering Behavior and Residual Saturation Characteristics of Hydrogen-Brine Two-Phase Flow Under Different Pore-Throat Structures and Flow Velocity Conditions (Postprint)

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Date: 2025-07-22T00:00:00+00:00

Abstract

With the continuous growth in clean energy demand, underground hydrogen storage technology has gradually emerged as a research hotspot. Hydrogen exhibits stronger diffusion capability, lower density, and lower viscosity compared to air, and these physical properties cause its transport behavior in porous media to display a series of unique flow phenomena, among which the "fingering" phenomenon is a typical example. This phenomenon frequently leads to liquid entrapment within microscale pore structures, thereby significantly reducing ultimate hydrogen saturation and recovery efficiency. This study employs COMSOL to construct two-dimensional microfluidic porous media models with pore throat diameters of 0.05 mm and 0.08 mm, and varies the injection rate across a $\log_{10}$Ca range from -5.57 to -2.87. By extracting the morphological characteristics of the displacement front and the residual saturations after breakthrough and injection-production cycling, the differences in fingering patterns under viscous-force-dominated versus capillary-force-dominated conditions are systematically analyzed. The results demonstrate that under high Ca conditions, viscous forces dominate, forming multiple parallel viscous fingers; under intermediate Ca conditions, a transitional regime occurs where the number of fingering channels decreases, and hydrogen saturation in crossover regions is generally lower than that in capillary-dominated or viscous-dominated flow regimes; under low Ca conditions, capillary forces dominate, manifested as capillary fingering advancing along the minimum capillary threshold path, accompanied by Haines jumps. This research provides micro-mechanistic references for pore structure design in underground hydrogen storage media and optimization of injection-production parameters, contributing to enhanced storage capacity and recovery efficiency.

Full Text

Study on Capillary Fingering and Residual Saturation during Hydrogen Displacement of Brine under Different Pore-Throat Structures and Flow Rates

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Fingering behavior in porous media reduces the volume of hydrogen, leading to low hydrogen saturation in aquifers and limited storage capacity [7]. Conversely, during extraction, complete recovery of stored gas is challenging due to gas bubbles trapped in pores. Insufficient invasion and unrecoverable hydrogen significantly reduce storage capacity and recovery efficiency [8]. Therefore, studies on hydrogen fingering are crucial for ensuring the economic and operational feasibility of underground hydrogen storage. Fingering in porous systems is typically influenced by fluid properties (e.g., viscosity, injection velocity, fluid-solid interactions) and the microstructure of porous media. Understanding pore-scale fluid flow and transport properties is fundamental to reducing uncertainties related to dynamic behavior, volumetric capacity, and injection/withdrawal efficiency in reservoirs and groundwater systems.

This paper highlights the role of injection velocity and pore structure in hydrogen fingering patterns and the resulting saturation evolution. To understand the hydrogen-brine displacement process in porous media at the microscale, a numerical model was developed to simulate hydrogen injection into a two-dimensional saturated heterogeneous porous medium at different injection velocities. The influence of microstructure (pore-throat size) on fingering patterns is then examined.

The contribution of renewable energy, particularly wind and solar, to the global energy mix is expected to increase significantly in the future. The intermittency of these resources makes the development of large-scale energy storage facilities an essential component of future green energy systems [1]. Hydrogen (H_2) is considered an attractive energy carrier due to its high energy content per unit mass and clean combustion products. In 2021, global hydrogen demand reached 94 million tons, recovering to pre-pandemic levels (91 million tons in 2019), containing energy equivalent to 2.5% of global final energy consumption. Most growth came from traditional uses in refining and industry, while demand from new applications grew to about 40,000 tons, a 60% increase from 2020.

Due to hydrogen' s low density and being the smallest molecule, surface storage facilities cannot provide the capacity needed for large-scale energy storage [2]. Geological formations such as depleted oil and gas reservoirs, aquifers, and salt caverns have proven to provide safe storage options for gases like methane and carbon dioxide, and thus may offer potential solutions for hydrogen storage [3].

Among these, porous reservoir underground hydrogen storage has attracted considerable attention, with its feasibility largely depending on hydrogen flow and transport behavior. The injection and withdrawal cycles in reservoirs are controlled by complex pore-scale processes. Researchers have observed that when simulating hydrogen storage in sandstone reservoirs, hydrogen primarily occupies large pores, while brine occupies small pores, pore throats, and corners due to hydrogen’ s strong water-wetting properties. Pore-level observations provide strong preliminary indications that brine aquifers in sandstone reservoirs are favorable for hydrogen storage, as related experiments have demonstrated that hydrogen volume can remain unchanged for long periods unless relatively high rock-wetting gas is injected [4].

Underground porous media are complex multiphase systems, with macroscopic behavior influenced by physical phenomena occurring at the pore scale. When hydrogen is injected into water-saturated aquifers, the displacement process typically becomes unstable, leading to non-uniform hydrogen propagation in the porous structure. This finger-like instability, commonly termed fingering, arises from viscosity differences between hydrogen and water and the heterogeneous structure of porous media [5]. Hydrogen fingering leads to incomplete hydrogen invasion after injection [6].

2.1 Pore-Scale Multiphase Flow

Fundamental understanding of immiscible phase displacement at the pore scale dates back to the pioneering work of Lenormand et al. [9] in 1988. They found that under the influence of viscosity differences, two-phase displacement fronts often advance in a fingering pattern. Depending on the dominant forces, displacement processes can be classified into different flow regimes. As injection velocity increases, fluid systems evolve from capillary fingering-dominated to transitional regimes, ultimately entering a viscous fingering-dominated stage.

To reveal the evolution of displacement mechanisms, Lenormand et al. established a classical phase diagram model that delineates different displacement patterns through two dimensionless parameters—the capillary number (Ca) and viscosity ratio (M), as shown in [Figure 2: see original paper]. These parameters characterize the transitions among three typical behaviors: capillary fingering, viscous fingering, and stable displacement.

The capillary number (Ca) and viscosity ratio (M) are defined as follows:

[Figure 2: see original paper]

Table 1 Physical and mechanical parameters of relevant materials

Parameter	Value
$\gamma \cos \theta$	
Brine viscosity, μ_w	
Hydrogen viscosity, μ_h	
Interfacial tension, γ	2.00×10^3
Contact angle, θ	8.41×10^{-6}

Parameter	Value
kg/(m • s)	kg/(m • s)

where U is the injection velocity; μ_1 and μ_2 represent the viscosities of invading and invaded fluids, respectively; γ denotes interfacial tension; and θ is the contact angle.

2.2 Two-Dimensional Model of Porous Media

Two sets of micro-models were developed to simulate hydrogen-brine displacement patterns in porous media. A two-dimensional rectangular domain of 10 mm × 10 mm was modeled ([Figure 3: see original paper]). Impermeable circular particles were distributed in an equilateral triangular pattern in the micromodel, representing heterogeneous solids in sandstone aquifers. Pore-throat size served as the controlling parameter of the microstructure, as shown in [Figure 3: see original paper].

Figures 3(a) and 3(b) represent porous media with the same porosity of 0.262 [10]. Figure 3(b) was obtained by scaling up Figure 3(a) by a factor of 1.6 to maintain the same porosity while increasing pore-throat size. Expanding the diameter of a portion of randomly distributed particles by 10% and reducing another portion by 10% slightly perturbs the medium’ s uniformity. Particle surfaces are wetting walls with a certain contact angle. Brine in the model was displaced by hydrogen injected through the left inlet. Six fluid injection velocities were used in the study, ranging from 1×10^{-4} m/s to 5×10^{-2} m/s. During injection, the model was initially fully saturated with brine. Hydrogen was then injected at a constant flow rate from the left side of the model, allowing brine to exit from the right side. Conversely, during withdrawal, brine was injected from the right side of the model, releasing hydrogen from the left.

3 Numerical Results

Hydrogen-brine two-phase displacement with different injection velocities was simulated in the designed micro-models. The distribution of hydrogen in the micro-model at breakthrough was recorded. Breakthrough time denotes the point when the invading fluid reaches the micro-model outlet. Parameters used in the numerical simulations are listed in .

3.1 Hydrogen Displacing Brine

Numerical simulations of hydrogen injection were first performed on models with pore-throat sizes of 0.05 mm and 0.08 mm. Since the capillary number Ca is related to injection velocity, its variation significantly influences hydrogen-brine displacement behavior. [Figure 4: see original paper] shows typical displacement patterns formed at breakthrough under different injection velocities (ranging from 1×10^{-4} to 5×10^{-2} m/s, corresponding to $\log_{10}$Ca values from -5.57 to -2.87). Based on morphological characteristics of the displacement front, three typical flow patterns can be identified: viscous fingering, transitional regime fingering, and capillary fingering, marked by green, blue, and yellow boxes in [Figure 4: see original paper] respectively (solid boxes for 0.08 mm throat micro-model, dashed boxes for 0.05 mm throat micro-model).

When the injection velocity is 0.01 m/s ($\log_{10}$Ca = -3.57), multiple slender primary fingers can be observed advancing almost parallel from inlet to outlet, a characteristic typical of viscous fingering, as shown in the green box in [Figure 4: see original paper]. As Ca decreases, the number of primary fingers significantly reduces, showing a trend of shifting from the original injection direction to the longitudinal direction, while hydrogen saturation also drops notably (blue box region in [Figure 4: see original paper]). When Ca further decreases, hydrogen preferentially advances along paths with larger local pore-throat radii and minimal capillary resistance, forming a single dominant channel (yellow box in [Figure 4: see original paper]) or exhibiting deflection of some fingering paths (yellow box in [Figure 4: see original paper]). The displacement front shows obvious reverse invasion phenomena, marking the capillary force-dominated displacement stage, i.e., capillary fingering. At this stage, partial regression occurs in finger structures, indicating that low-velocity injection enhances the influence of capillary forces on displacement patterns.

During Haines jumps, bridging between primary fingers and connecting branches may break ([Figure 5: see original paper]), facilitating the formation of isolated bubbles. Repeated Haines jumps significantly increase hydrogen bubble size and trigger hydrogen bubble generation.

[Figure 5: see original paper] Haines jumps causing bubble formation, $\log_{10}$Ca = -5.57

3.2 Saturation at Breakthrough

Breakthrough hydrogen saturation depends on injection velocity and porous medium microstructure. [Figure 6: see original paper] shows the relationship between Ca and saturation.

It can be observed that in the capillary fingering stage (yellow box in the figure), hydrogen more easily forms a horizontal dominant channel that breaks through directly in the 0.05 mm throat micro-model, while in the 0.08 mm throat micro-model, hydrogen more readily undergoes upward or downward longitudinal migration, with the front more prone to bending and stronger directional expansion. This is because for the 0.05 mm throat, capillary pressure is relatively large and capillary force dominance is strong, making the interface more likely to advance along the path of minimal capillary pressure, producing a narrow and stable dominant channel. For the 0.08 mm throat, capillary pressure decreases and capillary force dominance diminishes, making the interface more unstable during displacement, with disturbances more likely to develop, resulting in interface warping or bifurcation, causing more longitudinal migration and front bending, and even “Haines jump” phenomena [11].

When $\log_{10}$Ca = -2.87, typical viscous fingering is observed, with three primary fingers advancing parallel along the injection direction. The propagation of multiple fingers thus creates high hydrogen saturation (i.e., 0.306). When $\log_{10}$Ca = -5.57 or $\log_{10}$Ca = -4.87, the displacement pattern is in the capillary fingering regime, with non-wetting phase invasion controlled by capillary forces. Hydrogen advances in a primary finger, and under capillary pressure, hydrogen more readily undergoes upward or downward longitudinal migration, with the front more prone to bending and stronger directional expansion, leading to high saturation in capillary fingering (i.e., 0.344), as shown in [Figure 4: see original paper].

It should be noted that under 0.05 mm throat conditions, although the number of branches at the displacement front is smaller and the overall propagation path is more concentrated, the interface is irregular and fluctuates significantly, with obvious Haines jump phenomena observed during propagation. This indicates that such fingering behavior remains dominated by capillary forces [12], revealing a nonlinear coupling relationship between microstructure and displacement mechanisms. Displacement patterns in the transitional regime exhibit behavior transitioning from viscous to capillary fingering, with relatively low saturation in transitional fingering. Due to the interaction between viscous and capillary forces, the number of primary fingers is limited and most branches propagate forward. Therefore, hydrogen saturation in transitional fingering is generally lower than in capillary- or viscous-dominated flow regimes.

3.3 Residual Saturation of Hydrogen

Continuous cyclic injection and withdrawal of hydrogen is important in underground hydrogen storage. Reservoir storage capacity and hydrogen recovery efficiency at the microscale are primarily influenced by initial hydrogen saturation after drainage and residual hydrogen saturation after imbibition [13]. Higher hydrogen saturation after invasion means greater storage capacity, while higher residual hydrogen saturation after withdrawal indicates more hydrogen trapped in the porous medium, thereby reducing overall hydrogen recovery efficiency.

[Figure 7: see original paper] shows the effect of Ca on displacement patterns during invasion-withdrawal cycles. This section simulates hydrogen injection-withdrawal cycles in a micro-model with porosity of 0.262 and throat size of 0.05 mm (see [Figure 7: see original paper]). To investigate the effect of Ca on hydrogen saturation during injection-withdrawal processes, three different injection velocities were set, corresponding to $\log_{10}$Ca values of -3.87, -3.57, and -2.87.

Each simulation underwent one cycle, with fluid distributions during corresponding invasion and withdrawal stages shown in [Figure 7: see original paper]. Measured initial and residual hydrogen saturations for each cycle are quantified in the bar chart in [Figure 8: see original paper].

At higher velocities, the high mobility of the hydrogen front promotes pore occupation, leading to increased hydrogen storage capacity. During withdrawal, hydrogen is extracted along viscous fingers, leaving very limited hydrogen in the micro-model. This suppression of bubble formation results in low residual saturation and contributes to high recovery efficiency, defined as hydrogen saturation before and after drainage.

Conclusions

This study, based on the COMSOL multiphysics simulation platform, constructed two-dimensional regular circular particle porous medium models with throat diameters of 0.05 mm and 0.08 mm, systematically investigating fingering behavior and saturation evolution during hydrogen displacement of brine under $\log_{10}\mathrm{Ca}$ conditions ranging from -5.57 to -2.87, and conducted injection-withdrawal cycle simulations. The main conclusions are as follows:

(1) At high Ca ($\log_{10}\mathrm{Ca} \geq -3.57$), viscous forces dominate, forming multiple parallel viscous fingers; at intermediate Ca ($-5.57 < \log_{10}\mathrm{Ca} < -3.57$), the transitional regime shows reduced fingering channels with front bifurcation and bending, where hydrogen saturation in transitional fingering is generally lower than in capillary- or viscous-dominated flow regimes; at low Ca ($\log_{10}\mathrm{Ca} \leq -5.57$), capillary forces dominate, exhibiting capillary fingering that advances along the path of minimal capillary resistance, accompanied by Haines jumps.

(2) Pore-throat scale exhibits coupling effects. In the capillary fingering stage, the 0.05 mm throat, due to higher capillary resistance thresholds, forms a concentrated, narrow, and stable dominant channel; while the 0.08 mm throat, with lower thresholds, shows greater interface instability with more bending and bifurcation, and more frequent local Haines jumps.

(3) Injection-withdrawal cycle results show that both initial and residual saturation decrease significantly with increasing Ca; when $\log_{10}\mathrm{Ca}$ increases from -3.87 to -2.87, residual saturation decreases from approximately 0.22 to 0.15, indicating that high-velocity injection helps reduce hydrogen trapping and improve recovery efficiency.

This study reveals the nonlinear coupling laws of pore-throat geometry and injection rate on hydrogen-brine two-phase flow fingering behavior and saturation evolution under dual capillary-viscous driving forces, providing micromechanistic insights for pore design and injection-withdrawal parameter optimization in underground hydrogen storage media, which can help improve reservoir hydrogen storage capacity and recovery efficiency.

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