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Original article

The role of Capillary Hysteresis in Enhancing CO₂ Trapping Efficiency and Storage Stability

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ABSTRACT

Background: The intensifying impact of climate change demands innovative approaches to reduce atmospheric CO₂ levels. Carbon Capture and Storage (CCS) offers a viable solution by sequestering CO₂ in geological reservoirs. However, understanding the role of capillary hysteresis in CO₂ trapping is critical for optimizing CCS performance.

Aim: This study aims to investigate the influence of capillary hysteresis on CO₂ trapping efficiency in saline aquifers using detailed simulation models and varying hysteresis values.

Materials and methods: Advanced CMG simulation software was utilized to model CO₂ injection and migration in saline aquifers spanning depths of 1200–1300 meters. The model, initially saturated with brine, applied water-alternating-gas (WAG) injection at hysteresis values of 0.2, 0.3, 0.4, and 0.5 to evaluate their effect on CO₂ trapping efficiency.

Results: The simulations demonstrated a direct positive correlation between hysteresis values and CO₂ trapping efficiency. At a hysteresis value of 0.5, nearly 100% CO₂ trapping was achieved. This increased efficiency was attributed to stronger capillary forces immobilizing CO₂ more effectively and reducing mobility towards caprock, thereby minimizing leakage risks.

Conclusion: The study highlights the key role of capillary hysteresis in enhancing CO₂ sequestration. Higher hysteresis values improve long-term storage stability, emphasizing the need for optimized WAG injection strategies in CCS applications.

Keywords: CO₂ storage; capillary hysteresis; saline aquifers; WAG injection; climate change mitigation.

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Оригинальное исследование

Роль капиллярного гистерезиса в повышении эффективности улавливания и стабильности хранения CO₂

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АННОТАЦИЯ

Обоснование. Усиление воздействия изменения климата требует инновационных подходов к снижению уровня CO₂ в атмосфере. Улавливание и хранение углерода обеспечивает действенное решение путём секвестрации CO₂ в геологических коллекторах. Понимание роли капиллярного гистерезиса в улавливании CO₂ имеет решающее значение для оптимизации эффективности улавливания и хранения углерода.

Цель. Цель данного исследования является изучение влияния капиллярного гистерезиса на эффективность улавливания CO₂ в солёных водоносных горизонтах с помощью детальных имитационных моделей и переменных значений гистерезиса.

Материалы и методы. Для моделирования закачки и миграции CO₂ в солёные водоносные горизонты глубиной 1200–1300 м было использовано современное программное обеспечение CMG. В модели, первоначально насыщенной рассолом, применялась поочередная закачка воды и газа при значениях гистерезиса 0,2, 0,3, 0,4 и 0,5 для оценки влияния этих значений на эффективность улавливания CO₂.

Результаты. Моделирование показало прямую положительную корреляцию между величиной гистерезиса и эффективностью улавливания CO₂. При значении гистерезиса 0,5 было достигнуто почти стопроцентное улавливание CO₂. Такое повышение эффективности объясняется тем, что более сильные капиллярные силы эффективнее иммобилизуют CO₂ и снижают его подвижность в сторону покрова продуктивного пласта, тем самым минимизируя риски утечки.

Заключение. Исследование подчеркивает ключевую роль капиллярного гистерезиса в повышении эффективности секвестрации CO₂. Более высокие значения гистерезиса улучшают долгосрочную стабильность хранилища, подчеркивая необходимость оптимизации стратегий поочередной закачки воды и газа в системах улавливания и хранения углерода.

Ключевые слова: хранение CO₂, капиллярный гистерезис, солёные водоносные горизонты, поочередная закачка воды и газа, смягчение последствий изменения климата.

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Түпнұсқа зерттеу

Капиллярлық гистерезистің CO₂ тұту тиімділігі мен сақтау тұрақтылығын арттырудағы рөлі

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АННОТАЦИЯ

Негіздеу. Климаттың өзгеруінің әсерін күшейту атмосферадағы CO₂ деңгейін төмендетудің инновациялық тәсілдерін қажет етеді. Көміртекті ұстау және сақтау геологиялық коллекторларда CO₂ секвестрлеу арқылы тиімді шешімді қамтамасыз етеді. Капиллярлық гистерезистің CO₂ тұтудағы рөлін түсіну көміртекті ұстау және сақтау тиімділігін оңтайландыру үшін өте маңызды.

Мақсаты. Бұл зерттеудің мақсаты – капиллярлық гистерезистің толық имитациялық модельдер мен гистерезис мәндерінің айнымалылары арқылы тұзды сулы горизонттардағы CO₂ тұту тиімділігіне әсерін зерттеу.

Материалдар мен әдістер. Тереңдігі 1200-1300 м тұзды сулы горизонттарға CO₂ айдау және кезуін модельдеу үшін заманауи CMG бағдарламалық жасақтамасы қолданылды. Бастапқыда тұзды ерітіндімен қаныққан модельде бұл мәндердің CO₂ тұту тиімділігіне әсерін бағалау үшін гистерезис 0,2, 0,3, 0,4 және 0,5 мәндерінде кезектесіп су мен газ айдау қолданылды.

Нәтижелері. Модельдеу гистерезис мөлшері мен CO₂ тұту тиімділігі арасындағы тікелей оң корреляцияны көрсетті. Гистерезис мәні 0,5 болған кезде CO₂-ны жүз пайызға жуық тұтуға қол жеткізілді. Бұл тиімділіктің артуы күшті капиллярлық күштер CO₂-ны тиімді иммобилизациялайтындығымен және оның өнімді қабаттың қақпағына қарай қозғалғыштығын төмендететіндігімен түсіндіріледі, осылайша ағып кету қаупін азайтады.

Қорытынды. Зерттеу капиллярлық гистерезистің CO₂ секвестрінің тиімділігін арттырудағы негізгі рөлін көрсетеді. Гистерезистің жоғары мәндері көміртекті тұту және сақтау жүйелерінде су мен газды кезек-кезек айдау стратегияларын оңтайландыру қажеттілігін көрсете отырып, ұзақ мерзімді сақтау тұрақтылығын жақсартады.

Негізгі сөздер: CO₂ сақтау, капиллярлық гистерезис, тұзды сулы горизонттар, кезектесіп су мен газ айдау, климаттың өзгеруін азайту.

Дәйексөз келтіру үшін:

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Introduction

Industrialization, urban growth, and migration to cities significantly drive up carbon dioxide (CO₂) emissions [1]. CO₂ absorbs heat from the sun and traps it in the atmosphere, leading to ozone layer depletion and alterations in atmospheric circulation patterns [2]. CO₂ geological storage has emerged as an effective approach to reducing carbon footprints and addressing environmental concerns, providing a solution for managing future emissions as part of a comprehensive strategy to combat climate change [3]. Carbon emissions are captured from power plants and permanently stored underground in saline aquifers or abandoned hydrocarbon reservoirs, known for securely storing gases [4]. Four main mechanisms – structural trapping, capillary trapping, solubility trapping, and mineral trapping–hydrodynamically or geochemically immobilize CO₂.

Structural and stratigraphic trapping, prominent in the initial stages of a CO₂ storage project, relies on an overlying caprock to prevent capillary leakage of CO₂ [5]. Capillary trapping occurs when CO₂ becomes immobile, forming isolated ganglia within pore spaces, enclosed by brine in storage aquifer formations [6]. Solubility trapping is considered a secure storage method, where CO₂ bubbles dissolve in the aqueous phase, creating carbonic acid [7]. This acid interacts with metal ions (Ca²⁺, Fe²⁺, Mg²⁺) within the geological structure through geochemical reactions, producing durable solid carbonate minerals known as mineral trapping [8]. However, structural and stratigraphic trapping, which relies on the presence of an overlying caprock, may encounter geological complexities and may not be feasible in all geological formations. Similarly, dissolution trapping requires time for significant storage, and mineral trapping, involving the formation of solid carbonate minerals, is a slow process, further delaying effective CO₂ storage. In contrast, capillary trapping, a rapid process, occurs early in storage, offering an immediate solution and serving as a key element for successful CO₂ storage. This mechanism involves water entering pore spaces, displacing CO₂ and leaving isolated pockets or droplets behind. Residual trapping is crucial for securely storing CO₂ underground over time, significantly enhancing storage efficiency and encapsulation within geological formations.

Previous studies have employed two methods to examine residual trapping behavior. The first method involves utilizing different ratios of vertical to horizontal permeability [9], as well as varying injection rates, temperatures, and pressures for a specific set of relative permeability curves [10]. The second method isolates the impact of changes in relative permeability curves by measuring the variations in trapped gas saturations. This is done by altering endpoint values such as residual gas saturation, critical gas saturation, irreducible water saturation, and wetting conditions, while maintaining other factors constant [11]. This research employs

the second method by using different hysteresis values in relative permeability curves to investigate CO₂ capillary trapping. The commercial CMG simulator is utilized to monitor the distribution of CO₂ after injection into an aquifer, followed by alternating water injections. The influence of four different hysteresis values (0.2, 0.3, 0.4, and 0.5), which reflect differences between drainage and imbibition relative permeability curves, is systematically studied to assess the CO₂ plume shape and trapping efficiency underground.

Model Characteristics

A The aquifer model comprised 2000 blocks: 100 in the i-direction, 1 in the j-direction, and 20 in the k-direction, each block measuring 10 meters in length and width and 5 meters in thickness. Stratified between 1200 and 1300 meters depth, the model had an initial pressure of 1800 psi at 1200 meters (Fig. 1) and maintained a constant temperature of 55°C, characteristic of deep saline aquifers. The aquifer was initially saturated with 6% salinity brine, with water compressibility at 3.102×10^{-6} psi⁻¹ and rock compressibility at 3.793×10^{-6} psi⁻¹.

A uniform porosity of 0.13 was applied across all layers to accurately simulate fluid flow, while permeability was set at 60 millidarcies in all directions to model CO₂ plume movement. To represent an infinite reservoir, boundary cell pore volumes were exponentially increased using a volume modifier of 1000, allowing unrestricted fluid flow. Using the CMG-GEM simulator, CO₂ was injected at a rate of 10,000 m³/day to a depth of 1285–1300 meters for one year, followed by a year of water injection at 50 m³/day to 1220–1235 meters after a one-year pause. A 10-year observation period tracked CO₂ migration, focusing on structural and capillary trapping. The simulation revealed that CO₂ displaces water initially but is later trapped as water re-injection lowers CO₂ permeability, achieving residual water saturation. The effectiveness of structural and residual trapping depends on CO₂'s mobility through the rock relative to water, controlled by relative permeability curves hysteresis, which is critical for predicting and optimizing CO₂ storage strategies.

Wetting relative permeability (krw) and non-wetting relative permeability (krg) were derived from the experimental study conducted by Edlmann et al. [12]. They injected water into strongly water-wet sandstone cores until reaching steady-state flow, marking the primary imbibition phase, followed by CO₂ injection representing the primary drainage phase. This alternating injection process was repeated for five cycles, each revealing a progressive hysteresis effect on the relative permeability curves. They employed a critical CO₂ saturation (S_{gc}) and irreducible water saturation (S_{wr}) of 0.05 and 0.2, respectively, to determine drainage relative permeability. Initially, S_{gr} was fixed at 0.2 for the first imbibition relative permeability curve and then shifted by 0.1 for the next five cycles of imbibition for the sandstone cores. The relationship between water saturation (Sw) and relative permeability

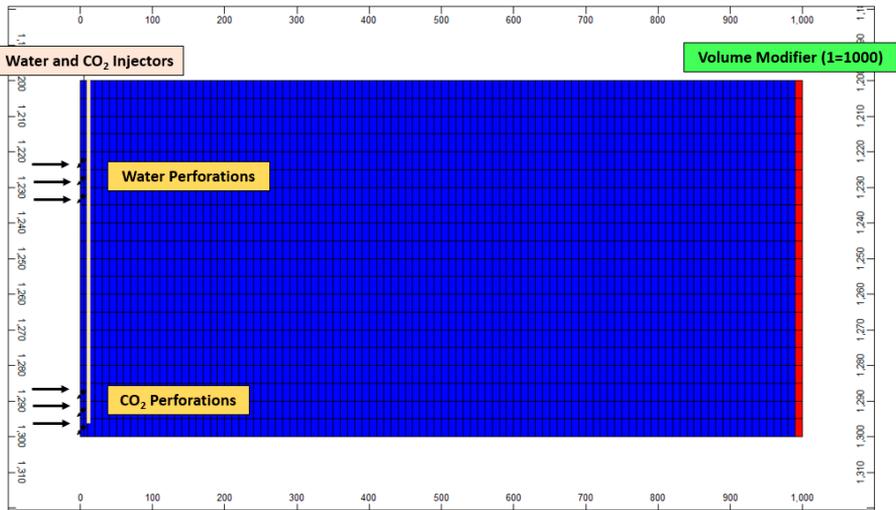


Figure 1. 2D aquifer model with perforations at a depth of 1220–1235 meters for water injection and 1285–1300 meters for CO₂ injection. An infinite boundary was also established by applying a volume modifier of 1000 to the right boundary

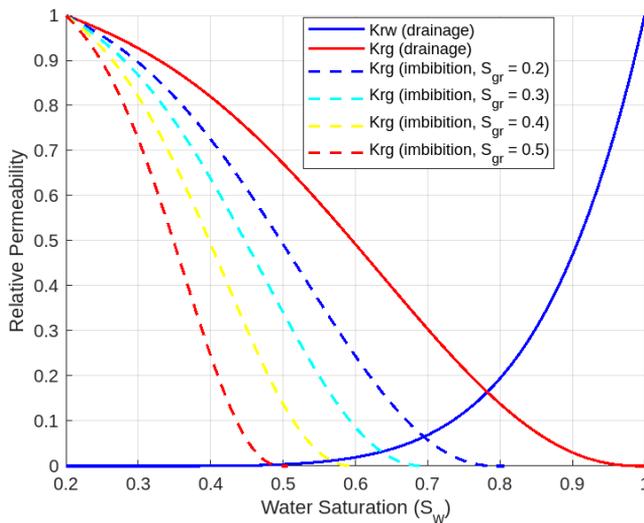


Figure 2. The utilized relative permeability curves for both water and CO₂ in highly water-wet sandstone cores through the primary drainage and imbibition phases, as inferred from the experimental study by Edlmann et al. [12] and mathematically represented using the Brooks-Corey-Moalem model [13]

for both the wetting (water) and non-wetting (CO₂) phases in water-wet sandstone is visually summarized in Fig. 2. This figure includes the observed hysteresis effect, evident throughout successive experimental cycles, which is the shift from drainage to imbibition relative permeability. The relative permeability curves are used in this study to examine the role of hysteresis in the efficacy of CO₂ capillary trapping within geological formations.

Results and Discussions

The study meticulously examines the role of hysteresis in the efficacy of CO₂ trapping within geological formations, an integral component

of carbon capture and storage (CCS) initiatives. Using CMG simulation software, a comparative analysis is conducted, contrasting four scenarios with hysteresis values of 0.2, 0.3, 0.4, and 0.5, which mimic drainage and imbibition processes through WAG injection.

Saturation Profiles

Fig. 3 illustrates saturation profiles for the first drainage process in a strongly water-wet aquifer, initially saturated with water and subjected to CO₂ injection. The hysteresis value in this process is assumed to be 0.2, as established in the lab study by Edlmann et al. [1]. Injected CO₂ from the bottom

left corner displaced the water and moved upward due to buoyancy, eventually reaching beneath the caprock, which acts as a no-flow boundary. This upward movement is clearly shown in the total gas saturation profile (Fig. 3a), where the highest gas saturation values are near the bottom left corner. The no-flow boundary at the caprock forces the CO₂ to spread horizontally, resulting in a broad distribution of gas saturation. As the system is strongly water-wet, the displaced water tends to return to pore spaces invaded by CO₂. The returning water moves back from lower layers with lower gas saturation, effectively snapping off and trapping CO₂ in isolated phases. The trapped gas saturation profile (Fig. 3b) shows a high concentration of trapped gas near the injection point and lower layers where the returning water has immobilized the CO₂ in the pore spaces due to capillary forces. The results of water injection following the first drainage process are illustrated in Fig. 4.

This injection, simulating the imbibition phase with a hysteresis of 0.3, pushes CO₂ into areas with high saturation, leading to more gas being trapped within the aquifer. The injected water displaces the CO₂, causing it to become trapped in isolated pockets. This process is driven by capillary forces, which are stronger during the imbibition phase due to hysteresis. During alternating drainage and imbibition cycles, the relative permeability curves shift, reflecting changes in the wetting and non-wetting phase saturations. This shift, known as hysteresis, results in a different saturation path during imbibition compared to drainage. Specifically, the non-wetting phase (CO₂) becomes trapped in the pore spaces during imbibition as the wetting phase (water) re-enters the pores and isolates the CO₂. The presence of this hysteresis-induced trapping reduces the mobility of the CO₂, preventing further migration due to buoyancy.

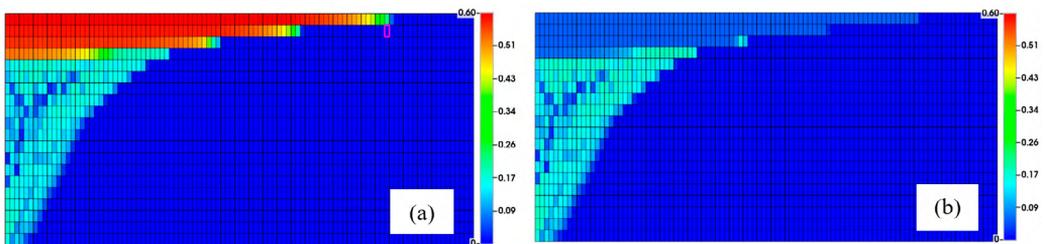


Figure 3. (a) Total gas saturation profile showing CO₂ injected from the bottom left corner spreading upward and horizontally beneath the caprock. (b) Trapped gas saturation profile showing CO₂ immobilized by returning water in lower layers

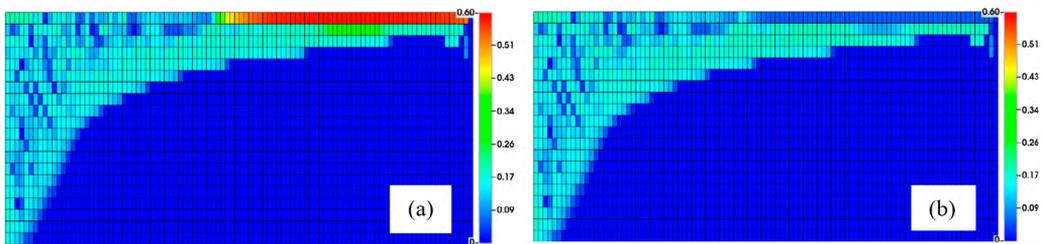


Figure 4. (a) Total gas saturation profile after water injection, showing reduced gas saturation at the top; (b) Trapped gas saturation profile, illustrating the hysteresis effect enhancing CO₂ immobilization during the imbibition phase with a hysteresis of 0.3

Fig. 5 presents the results of a subsequent drainage process where CO₂ is injected again, visualized in the saturation profiles. This time, the hysteresis value was increased to 0.4, up from the previous 0.3, reflecting an additional 0.1 increment. The total gas saturation (Fig. 5a) and trapped gas saturation (Fig. 5b) indicate significant changes compared to the previous drainage cycle. In Fig. 5a, the total gas saturation profile reveals that less CO₂ has moved to the top layers, with only a small section in the top layer exhibiting a light orange color, indicating a gas saturation of around 45%, whereas it was 60% and more extensive in the previous cycle. This reduction in gas saturation at the top layer suggests that CO₂ mobility has decreased

due to the increased trapping from the prior cycles. Fig. 5b highlights the trapped gas saturation, showing a significant increase in the amount of CO₂ immobilized by capillary forces. This enhanced trapping results from the hysteresis effect observed during the alternating drainage and imbibition cycles. As the relative permeability curves shift, the wetting phase (water) re-enters the pores, further isolating and trapping the CO₂. The increased trapping efficiency ensures more CO₂ remains securely immobilized within the aquifer, reducing the risk of CO₂ migration and enhancing long-term storage stability.

In the final simulation, water was injected again to simulate the imbibition process,

with the hysteresis in relative permeability set to 0.5. The resulting saturation profiles are shown in Fig. 6, with Fig. 6a representing the total gas saturation profile and Fig. 6b depicting the hysteresis-trapped gas profile. The profiles indicate that all the injected CO₂ has been effectively trapped, leaving no free gas in the system. This complete trapping is due to the increased hysteresis effect, which enhances the capillary forces during the imbibition phase, ensuring that the returning water isolates and immobilizes the CO₂ more effectively. The increased hysteresis value contributes to a stronger trapping mechanism, resulting in the complete immobilization of the CO₂ within the pore spaces. In conclusion, water-alternating-gas

injection progressively traps more gas, eventually leading to the absence of mobile gas.

Capillary Trapped Gas Efficiency

Fig. 7 presents the capillary trapped gas percentage as a function of dimensionless time (t_D) for varying hysteresis values, with the main plot on the left and an enlarged view on the right to accentuate the initial trapping phase. The data elucidates a positive correlation between hysteresis and CO₂ trapping efficiency, indicating that an increase in hysteresis enhances capillary forces, thereby augmenting CO₂ entrapment within the reservoir’s pore network and subsequently reducing post-injection mobility.

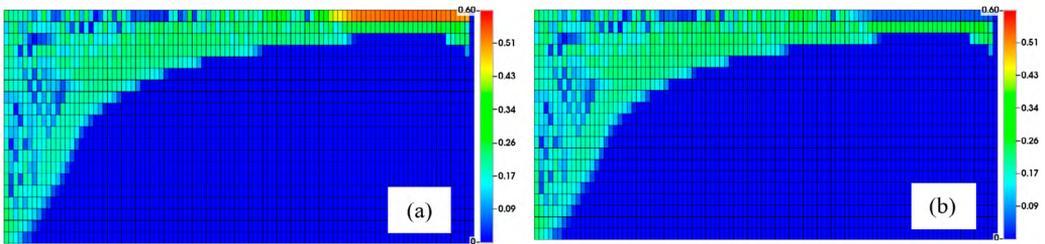


Figure 5. (a) Total gas saturation profile from the drainage process showing less CO₂ movement to the top layers, with a smaller section exhibiting a gas saturation of around 45%; (b) Trapped gas saturation profile indicating increased CO₂ immobilization due to the hysteresis effect during the drainage process

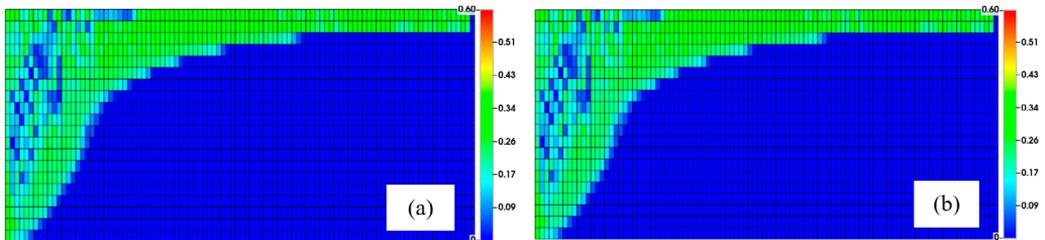


Figure 6. (a) Total gas saturation profile showing complete trapping of injected CO₂ with no free gas remaining; (b) Hysteresis trapped gas profile illustrating the enhanced capillary trapping due to a hysteresis value of 0.5 during the imbibition process

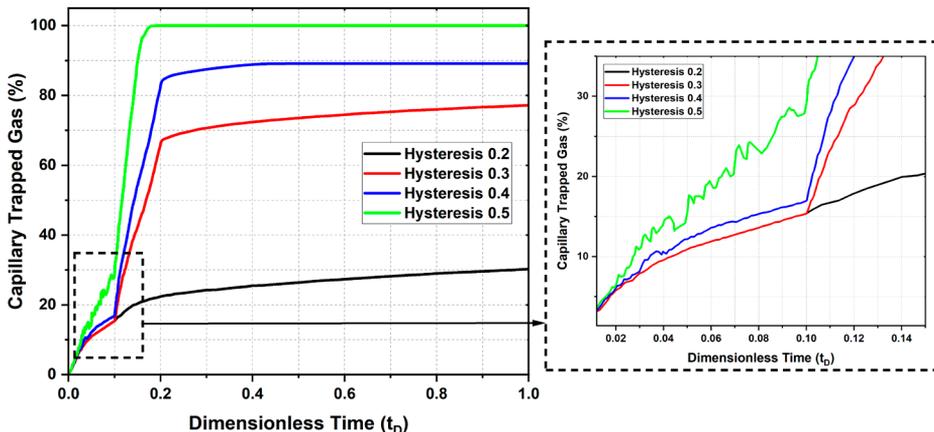


Figure 7. Capillary trapped gas percentage as a function of dimensionless time for different hysteresis values

In the primary plot, the capillary trapped gas percentage is depicted for hysteresis values of 0.2, 0.3, 0.4, and 0.5. A hysteresis value of 0.2 reflects the drainage process, wherein CO₂ displaces water from the pore spaces. The trapping efficiency for this value initiates at a low level and increases gradually, achieving a maximum trapped gas percentage of approximately 30% by the end of the simulation period. This gradual increase suggests that lower hysteresis results in less efficient trapping over time. In contrast, the trapping efficiency for a hysteresis value of 0.3, simulating imbibition through the post-injection of water, escalates more rapidly with a plateau around 75%. This indicates improved trapping efficacy through the snap-off of CO₂ by water in pore spaces.

For a hysteresis value of 0.4, the trapped gas percentage increases swiftly, reaching roughly 80%, demonstrating that a higher hysteresis value significantly enhances capillary forces, resulting in more efficient gas trapping. The highest hysteresis value tested, 0.5, exhibits the steepest rise in trapping efficiency, nearly achieving 100%. The rapid increase and elevated plateau imply that the highest hysteresis value results in the most efficient trapping. The zoomed-in plot on the right highlights the initial phase of the trapping process. All curves commence at zero, reflecting the absence of initial trapped gas. The green line (hysteresis 0.5) shows the most rapid increase in trapped gas percentage, followed sequentially by the blue (hysteresis 0.4), red (hysteresis 0.3), and black (hysteresis 0.2) lines. The fluctuations observed in the zoomed-in plot for higher hysteresis values (0.4 and 0.5) can be attributed to the dynamic interplay between capillary and viscous forces during the trapping process. Higher hysteresis engenders stronger capillary forces that effectively trap CO₂ in the pore spaces. However, as CO₂ injection proceeds, the viscous forces associated with the injection can momentarily reconnect trapped CO₂ clusters, causing them to form a stream and be released from the pore spaces, resulting in the observed fluctuations. These fluctuations are absent for lower hysteresis values (0.2 and 0.3) due to weaker capillary forces,

leading to more stable and gradual trapping without significant interplay between capillary and viscous forces.

The plots unequivocally demonstrate that increased hysteresis through post-water injection enhances the capillary trapping efficiency of gas within the aquifer. Elevated hysteresis values amplify capillary forces, which trap more gas more rapidly. As hysteresis intensifies, the ability of the wetting phase (water) to isolate and trap the non-wetting phase (CO₂) improves, owing to the increased capillary forces associated with higher hysteresis values, thereby preventing the mobilization of CO₂ and resulting in higher trapped gas percentages. These findings hold significant implications for CO₂ sequestration projects, where maximizing the trapped gas is pivotal for ensuring long-term storage stability. Implementing processes that augment hysteresis, such as WAG injection, can enhance the efficiency and security of CO₂ storage.

Conclusions

This study highlights the crucial role of hysteresis in enhancing CO₂ capillary trapping within a water-wet aquifer. The analysis reveals that lower hysteresis values (e.g., 0.2) lead to gradual and less efficient CO₂ trapping during the drainage process, achieving a maximum trapped gas percentage of around 30%. In contrast, higher hysteresis values, such as 0.3 and beyond, considerably improve trapping efficiency, with imbibition processes reaching up to 75% and subsequent drainage and imbibition cycles approaching nearly 100% trapped gas. The intensified hysteresis enhances capillary forces, ensuring CO₂ remains immobilized within the pore spaces, thereby reducing its mobility and preventing further migration. These findings are essential for CO₂ sequestration projects, suggesting that techniques like WAG injection can substantially improve storage security and long-term stability by amplifying hysteresis effects. Overall, strategic management of hysteresis through appropriate injection methods can maximize CO₂ trapping efficiency, contributing to effective and reliable CO₂ storage solutions.

ADDITIONAL INFORMATION

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